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Radiation treatment of gaseous and liquid effluents for contaminant removal

Proceedings of a technical meeting held in Sofia, Bulgaria, 7–10 September 2004

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FOREWORD

The Technical Meeting on Radiation Processing of Gaseous and Liquid Effluents conducted in Sofia, Bulgaria, 7–10 September 2004, discussed and evaluated issues related to the status and future trends in radiation application for environmental protection. Five experts from Bulgaria, India, the Republic of Korea, Poland, and the United States of America were invited to provide their experiences in this field. Twenty cost-free participants and observers — from Bulgaria, India, Lithuania, Poland and Ukraine — joined the meeting, and 15 papers in total were presented.

Research and development in radiation processing of gaseous and liquid effluents is undertaken in three fields: electron beam flue gas treatment (SO_X and NO_X removal), wastewater purification and sewage sludge sterilization. Wastewater or sludge treatment and flue gas purification all differ from technological points of view, but they are common services and applications of environmental radiation technology applications, based mostly on electron accelerators.

The technical meeting discussed new development in the field of radiation applications in environmental service, especially the status and prospects of radiation processing of gaseous and liquid effluents. Progress in the field of electron accelerators and gamma sources is crucial for routine application of the technology. Cost reduction and improvement of technical reliability are substantial especially for high power of accelerators and high activity of the sources needed for environmental applications. Environmental applications were carefully reviewed in accordance with the existing regulations and state of the art knowledge. The comparison with conventional commercial technologies was addressed as well. In flue gas treatment, applicability of the technology using different fossil fuels (coal, lignite, oil, etc.) was reviewed. The elaborated materials cover the technical and economical evaluation of the technologies. The possible applications of radiation technology for environmental preservation were presented during the meeting.

The electron beam (EB) technology for flue gas treatment was developed in Japan in the early 1980s. Later on, this process was investigated in pilot scale in the USA, Germany, Japan, China and Poland. Commercial EB flue gas treatment installations are operating in coal-fired plants in China and Poland. The plant in Poland treats approximately 270 000 Nm^3/h of flue gases. High efficiency of SOx and NOx removal (up to 90% for SOx and up to 70% for NOx) is achieved and by-product is a high quality fertilizer. The advantage of this technology over conventional ones has been clearly demonstrated from both the technical and the economic points of view. Further, its implementation depends on technical development of supply and operation of reliable, high power accelerators with minimal maintenance.

In accordance with a three party contract between the IAEA, Japanese Atomic Energy Research Institute (JAERI) and the Bulgarian Government a pilot plant for EB flue gas treatment was constructed in Maritza, Bulgaria to treat high humidity, high SOx and NOx gases from combustion of low-grade lignite. Flue gas of 10 000 cubic meters per hour was irradiated with three high energy accelerators, each 35 kW and 800 keV. The plant has been operating since January 2004. The efficiency of pollutant removal ranges of 90–99 % for SOx and 85–90% for NOx.

Materials presented in the report will serve as basis for the preparation of guidelines and feasibility studies including cost analyses for full-scale process implementation. Public awareness and technology acceptance are other factors to be considered in furthering the dissemination of the technology.

The IAEA wishes to thank all the meeting participants for their valuable contributions. The IAEA officer responsible for this publication was A.G. Chmielewski of the Division of Physical and Chemical Sciences.

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CONTENTS

SUMMARY

1. BACKGROUND

The problems of environmental damage and degradation of natural resources are receiving increasing attention throughout the world in recent years. The increasing population, increasing urbanization and enhanced industrial activities of mankind are all leading to degradation of the environment. For example, fossil fuels, such as coal, natural gas and petroleum, which are the main primary sources of heat and electrical energy production, are responsible for emitting a large number of pollutants into the atmosphere with off-gases from industries, power stations, residential heating systems and vehicles. All these fuels contain major constituents (carbon, hydrogen, oxygen) as well as other components, such as metals, sulphur and nitrogen compounds. During the combustion process different process residues and pollutants such as fly ash, sulphur oxides (SO_2 and SO_3), nitrogen oxides ($NO_x = NO_2 + NO$) and volatile organic compounds are emitted. Fly ash contains diverse trace elements (heavy metals). Air pollution caused by particulate matter and other pollutants not only directly impacts the environment, but also contaminates water and soil and leads to their degradation. Wet and dry deposition of inorganic pollutants leads to acidification of the environment. These phenomena affect human health, increase corrosion and destroy plants and forests.

Widespread forest damage has been reported worldwide. Many cultivated plants are not resistant to pollutants either, especially in the early period of vegetation. SO_2 and NO_x are oxidized in presence of water vapour forming sulphuric and nitric acids. Fog and droplets result in so called “acid rain” i.e. acid precipitation. In recent years investigation has shown that emission of volatile organic compounds (VOC) to the atmosphere can cause stratospheric ozone layer depletion, ground level photochemical ozone formation, and toxic or carcinogenic human health effects. VOCs contribute to the global greenhouse effect.

Ironically, coal, the dirtiest fuels among natural hydrocarbons, is expected to remain the principal fossil fuel for the next two centuries (Table I). Nonetheless, increasing use of fossil fuel will be needed to meet the increasing demands of the developed and developing countries. Thus, there exists an urgent need develop technologies that reduce or minimize the pollution associated with this increasing coal use [1].

TABLE I. PREDICTION OF NET INCREASE IN FUEL CONSUMPSION

Fuel	Power in GW		
	Installed in 1995	Predicted for 2020	Net increase
Coal	870	1836	966
Gas	435	1296	861
Oil	435	648	213
Hydro	667	1026	359
Nuclear	348	378	30
Other	145	216	71
Total	2900	5400	2500

The dramatic increase in global population (Table II) combined with industrialization, urbanization, agricultural intensification and water-intensive lifestyles is resulting in a global water supply crisis. While water is a renewable resource, it is only a finite resource. Only 3% of the world’s water is fresh, of which one-third is inaccessible. Presently, about 20 per cent of the population lacks access to safe drinking water. Yet, on one hand water is being used with abandon, and on the other, available supplies are increasingly becoming contaminated due to discharge of industrial and human waste into fresh water bodies.

Global freshwater consumption rose six-fold between 1900 and 1995 — at more than twice the rate of population growth. Moreover, water resources in exposed and underground collectors are being polluted with industrial and human waste. Up to a few decades ago most of the wastes discharged to water bodies comprised animal and human excreta and other organic components from industries. In areas with low population density and without sewage systems such problems were alleviated to a great extent by the natural self-purification capacity of the receiving water body. However, increasing urbanization in the last two centuries has been accompanied by subsequent expansion of sewerage collectors without any (or adequate) treatment. Liquid waste loads have become so large that the self-purification capacity of receiving streams of large human settlements can no longer prevent adverse effects on water quality. These wastes now constitute significant sources of water pollution. The industrial effluents carry chemical contaminations like heavy metals, organic pollutants, petrochemicals, pesticides and dyes, while discharge of sewage and sludge give rise to microbiological contamination of the water bodies. Some pollutants are synthesized in situ, as for example chloro-organic compounds originating from chlorine application for water/wastewater disinfections. The discharge of such materials into water bodies is potentially responsible for risk of infection, health effects caused by contaminated drinking water and offensive ardors.

TABLE II. GROWTH OF WORLD POPULATION BY CONTINENT AND REGION

Population (in millions)	Year		
	1980	1990	2020
North America	251.9	275.9	326.4
Latin America	262.7	448.1	716.3
Western Europe	433.5	454.1	489.2
Central and Eastern Europe	95.3	100.2	111.0
CIS	265.5	288.6	343.9
Middle East and North Africa	200.3	271.0	543.3
Sub-Saharan Africa	370.0	502.6	1195.3
Pacific	1559.2	1806.9	2428.4
(includes CPA)	(1084.7)	(1248.4)	(1652.5)
South Asia	909.5	1146.0	1937.9
Total	4347.9	5293.4	8091.7

About one-third of the world's population already lives in countries with moderate to high water supply stress — where water consumption is more than 10 per cent of the available renewable freshwater supply (Fig. 1). If the present consumption pattern continues, two third of the human population will live in water-stressed conditions by the year 2025. The declining state of the world's freshwater resources, in terms of quantity and quality, may prove to be the dominant issue on the environmental and developmental agenda of this century. The available world supply of freshwater cannot be increased; more and more people therefore will depend on this fixed supply in future. Water security, like food security, will become a major national and regional priority in many areas of the world in the decades to come. Here too, there thus exists a need to develop improved technologies that can control the pollution of this precious resource.

It is becoming increasingly clear that mankind’s environmental problems are no longer just local or regional, but have become continental in scope. Economically and technically feasible technologies for control of pollution from gaseous and liquid effluents streams are being sought by technologists working in a variety of areas. Radiation technologists are no different. Over the last few decades, radiation technology based techniques have been developed, demonstrated and deployed to alleviate some of environmental problems associated with gaseous and liquid effluent waste. It has been demonstrated that radiation technology offers an advanced solution to selective environmental problems.

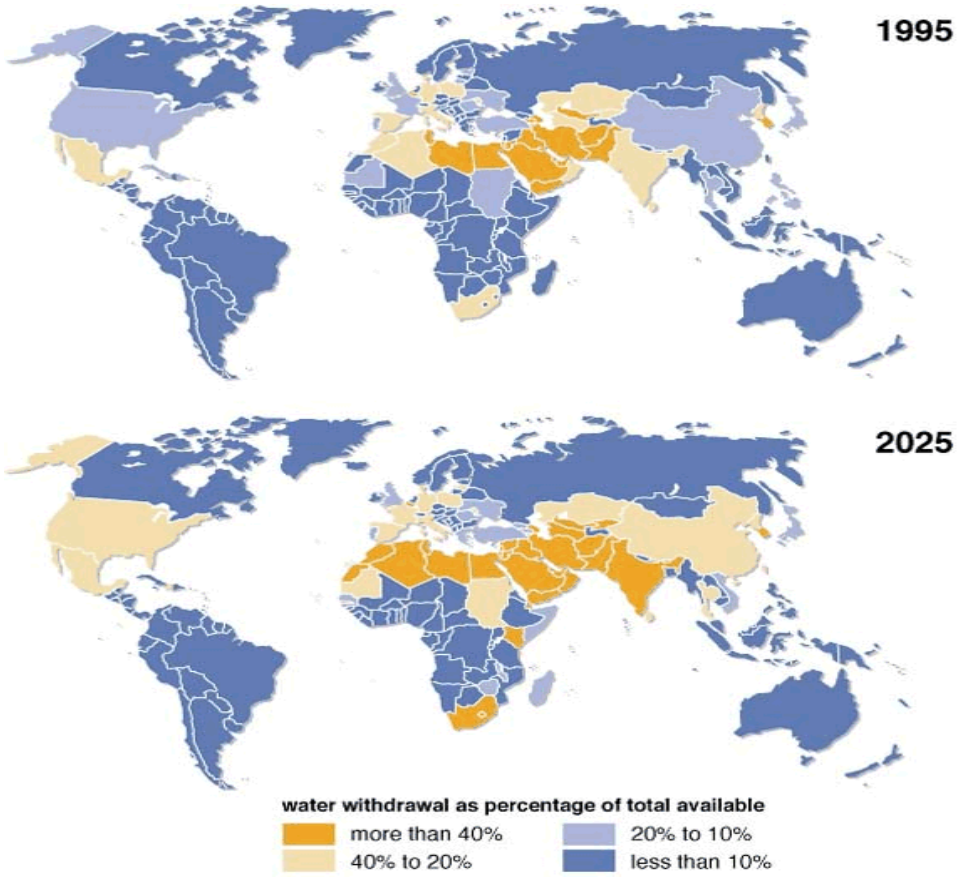

FIG. 1. Global water stress; Global environment outlook (GEO) 2000 by UNEP.

High-energy radiation was discovered more than one hundred years ago. Since then, properties of radiation to modify physico-chemical properties of materials have found many applications. Radiation technologies applying gamma sources and electron accelerators for material processing are well-established processes [2]. There are over 160 gamma industrial irradiators and 1300 electron industrial accelerators in operation worldwide that are being widely used for sterilization, food irradiation and polymer processing. Indeed, radiation processing today is a well established multi-billion dollar industry worldwide that is providing unique high value products for mankind in an environmentally friendly manner.

In recent years, large metropolitan cities in the world are facing the challenge of increasing environmental pollution resulting from ever increasing population and industrial activities. As a result, issues regarding environmental pollution, be it air, liquid or solid, are becoming a matter of concern. The realization that such pollutants pose a serious threat to human health has necessitated the need for development of cost effective and environmentally friendly technologies to overcome the problem. Radiation technology, with the unique ability to produce highly reactive species in an efficient manner, has the capability to alleviate some of these problems and thus play an important role in offering sustainable solutions.

Over the last few decades, extensive work has been carried out for utilizing radiation technology for environmental remediation. This includes application of radiation technology for simultaneous removal of sulphur dioxide and oxides of nitrogen from flue gases, purification of drinking water and wastewater purification and hygienization of sewage sludge for use in agriculture.

2. STATUS AND TRENDS IN RESEARCH AND DEVELOPMENT

The electron beam technology for flue gas treatment was developed in Japan in the early 1980s [3]. Later on, this process was investigated in pilot scale in the USA, Germany, Japan and Poland. The process was demonstrated in a pilot plant in Kaweczyn, Poland (20,000 Nm^3/h) using two accelerators (50 kW, 700 keV each) [4]. Recently, another pilot plant (flow 10,000 Nm^3/h; 3x30 kW, 800 keV) has been constructed in Bulgaria to treat high humidity, high SO_X gases from combustion of low-grade lignite [5]. The plant has achieved very good process parameters and efficiency of pollutant removal, which ranges from 90 to 97% for SO_X and 85–90% for NO_X. The fertilizer by-product obtained is of good quality with moisture content less than 1%. The Nitrogen content is approximately 21% or higher (recommended number for commercial fertilizer 21%). 96–97% of the product is ammonium sulphate and 2% nitrate.

Electron beam flue gas treatment plants are also operating in the coal-fired plants in China and Poland. Since the power of accelerators installed at the Polish plant is higher than 1 MW, it is the largest irradiation facility ever built. The plant treats approx. 270,000 Nm^3/h of flue gases. High efficiency of SO_X and NO_X removal was achieved (up to 95% for SO_X and up to 70% for NO_X) and by-product is a high quality fertilizer. The total investment cost was about US$ 18-20 million. The investment cost of the Chinese project was about US$ 11 million, since it treats SO_X mostly (accelerators of low power capacity). The other possible application of the technology is VOC and PAH treatment, e.g. in flue gas purification units of municipal waste incinerator plants.

Sewage sludge is the waste left over after municipal wastewater treatment plants have done their work. It is a rich source of many micronutrients and a source of carbon that make it a valuable fertilizer. However, it is often contaminated by pathogenic microorganisms that limit its use as a fertilizer for agricultural applications. A plant for liquid sludge sterilization, using gamma radiation from a cobalt-60 gamma source, has been in operation in India since 1992 [6]. The plant has been designed to treat 110 m^3 sludge per day fed from a conventional treatment plant with which it has been integrated. The operational experience of the plant has shown that the process is simple, effective, and easy to integrate with existing sewage treatment plants, and the radiation sterilized sludge can be beneficially utilized as a fertilizer in the agricultural field. Similarly, an electron beam accelerator can also be used for treatment of dewatered sludge.

A pilot plant for treating textile dyeing wastewater, equipped with an electron accelerator has been constructed in Republic of Korea, and an industrial project aiming at the treatment of 10 000 m^3/day is in progress [7, 8]. Based on the data obtained in laboratory experiments, the suitable doses are determined to be approximately 1–2 kGy for the flow rate of 10 000 m^3/day. Therefore, an accelerator with a power of 400 kW is applied for cost effectiveness and compactness of the plant. The cost for the high power accelerator is around US$ 2.0–2.5 million and building, piping, other equipment and construction works could be estimated at US$ 1.0–1.5 million. Considering the additional costs for tax, insurance and documentation of about US$ 0.5 million, the overall costs for plant construction are approximately US$ 4.0–4.5 million.

In all these applications, the advantages of radiation technology over conventional technologies have been clearly demonstrated from both, the technical and the economic points of view. It is therefore appropriate that the experience gained at these facilities be shared among the Member States, and the future strategies are defined so that the processes can be deployed on a larger scale.

The IAEA has organized meetings and coordinated research projects (CRPs) in which trends and new developments concerning applications of environmental radiation technologies were discussed.

Recent development in this field was elaborated by representatives of industry, universities and research institutes during the International Meeting on Radiation Processing (IMRP), held in Chicago, U.S.A, in September 2003 [1]. Some aspects of the process were reported during the Symposium on

Radiation Technologies in Emerging Industrial Applications organized by the IAEA in Beijing, China, in 2000 [1], which followed the Symposium on Radiation Technology for Conservation of the Environment, held in Zakopane, Poland, in 1997 [9].

The main research and implementation developments concentrated on three fields: electron beam flue gas treatment (SO_X and NO_X removal), wastewater purification and sludge sterilization. Since separation and enrichment technologies play a very important role in the products' recovery and pollution control, the possibility of radiation synthesis of stimuli-responsive membranes, hydrogels and adsorbents is being investigated as well [2]. Finally, besides already applied technologies for flue gas and wastewater treatment, further research is going on in the field of organic contaminant treatment in both, gaseous and liquid phases.

Reporting on the development of radiation sources to be applied in environmental radiation technologies mostly concerns high power electron beam machines. The technology has to be competitive with conventional ones, both from economic and technical points of view. Therefore, any developments concerning cost reduction and technological improvements regarding technical components, especially reliability of accelerators plays a very important role for further process implementation.

3. PURPOSE OF THE MEETING AND DISCUSSION TOPICS

The Technical Meeting on Radiation Processing of Gaseous and Liquid Effluents conducted in Sofia, Bulgaria, 7–10 September 2004, brought together outstanding experts in this field to discuss and evaluate issues related to the status and future trends concerning radiation application in environmental protection.

The major objectives of this meeting were the following:

- To share experience on design and operation of the facilities
- To evaluate and enumerate the advantages of radiation technology over conventional technology
- To assess the economic benefits and the limitations of the process
- To identify the technical needs in future deployment of the technology
- To identify the future potential areas wherein similar technology can be deployed
- To discuss the preparation of guidelines and feasibility studies including cost analyses for full-scale process implementation.

4. CONCLUSIONS

4.1. General conclusions

The programme of electron beam treatment of gaseous and liquid effluents, especially flue gases, wastewater purification and sewage sludge sterilization has been well established in the European countries as well as in China and Japan. Many of the fast developing countries in the East and West Asia, Africa and Europe regions are interested in the technology conducting feasibility studies and looking for assistance in training and capacity building [10–15].

The future of the radiation applications for environmental purposes depends on technical developments in electron beam technology, especially in designing and manufacturing of reliable compact accelerators with high power efficiency and minimal maintenance. This will reduce the operation cost and make the radiation technology very competitive for environmental applications [2].

4.2. Gaseous effluents

The successful operation of the electron beam treatment plants for flue gases in China and Poland for last few years has demonstrated the advantages of using this technology for removing NO_X and SO_2 from flue gases under varying conditions. The by-product in the form of ammonium sulphate and ammonium nitrate has been found to be of high purity and directly marketable.

The operational experience of the Maritza East-2 pilot scale plant recently established in Bulgaria has demonstrated that the electron beam treatment process is especially useful for treating emissions from lignite that has very high sulphur content (5–6%). An electron beam processing, removal efficiency of > 85% has been achieved for SO_2 and NO_X removal from the flue gas at a low dose of 2–4 kGy. The greatest strength of the technology under these conditions is that ammonium sulphate can be obtained at a relatively low cost.

Although the efficacy of the electron beam treatment has been well established for medium (2–4%) and high (5–6%) sulphur containing coals, the process needs to be studied further for plants using very low (<0.5%) sulphur containing coals.

Experiments conducted in recent years in Japan, Poland and Ukraine have demonstrated that besides SO_2 and NO_X, other potentially harmful chemicals, especially volatile organic compounds (VOCs) produced during burning process, can also be significantly reduced by electron beam irradiation.

During the discussions at the meeting, it also emerged that the presence of mercury in the flue gas also poses potential threat to human health, and studies should be conducted to investigate the effect of electron beam irradiation on mercury removal from the flue gases.

4.3. Liquid effluents

The main focus of radiation treatment of industrial wastewater is to convert non-biodegradable pollutants into biodegradable substances. In the use of high power electron accelerators substantial wastewater flow-rate duty is usually required to make radiation treatment cost effective. Radiation processing for industrial wastewater treatment is under investigation in many countries and is close to implementation. Preliminary results obtained in Republic of Korea in a textile dyeing wastewater have demonstrated the advantages of radiation technology over conventional techniques.

Radiation disinfection of effluents from a municipal wastewater plant for re-use has been successfully demonstrated by a number of researchers. Research activities have shown that inactivation of fecal coli-forms in secondary effluents from municipal wastewater plants can be obtained with doses of less than 1 kGy. While conventional disinfectants are adversely affected by the water chemistry matrix, radiation processing for bacteria inactivation is generally unaffected by the matrix. Therefore radiation processing has a clear advantage over the existing methods for municipal wastewater disinfection. Application of such low doses also leads to the degradation of synthetic and natural endocrine disrupters usually present in trace amounts in municipal wastewaters. However, at present there is no full scale radiation treatment plant in operation.

4.4. Sludge sterilization

The successful operation of a gamma irradiation facility in India for sterilization of sewage sludge for the last 10 years has demonstrated that this technology is simple, effective and can be easily integrated with use of conventional technology for sewage treatment. The end product, being free from pathogenic bacteria, can be easily inoculated with benign bacteria like Rhizobium and can be

converted into a biofertilizer. The recent large-scale field trials in agricultural farms in Egypt, India and Khazakstan, have shown that enriched sterilized sludge offers benefits such as higher crop output, lower water requirement and better soil characteristics. The radiation sterilized sludge has been well accepted by farmers. A number of Member States, especially in East and West Asia, and African countries, have shown keen interest in utilizing this technology.

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PRESENTATIONS

APPLICATION OF IONIZING RADIATION IN ENVIRONMENTAL PROTECTION

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Abstract

The radiation technology may contribute to the environmental protection to a great extent. The electron beam industrial installations for flue gases containing SOx and NOx treatment have been already built in China and Poland. The same technology for high sulfur and high humidity off-gases (low quality lignite) has been successfully tested in industrial pilot plant in Bulgaria. The pilot plant test performed in Japan have illustrated that by application of electron beam for municipal waste incinerator off-gases treatment concentration of dioxins can be reduced by 80 %, other Persistent Organic Pollutants can be depredated as well. The positive results of electron beam wastewater treatment are the basis of the full scale industrial plant being built in the Republic of Korea. The pilot gamma plant for sludge irradiation producing high grade organic fertilizer is in operation in India. All this achievements are reported in the paper.

1. INTRODUCTION

The municipal and industrial activities of man lead to environment degradation. The pollutants are emitted to the atmosphere with off-gases from industry, power stations, residential heating systems and vehicles. Fossil fuels, which include coal, natural gas, petroleum, shale oil and bitumen, are the main source of heat and electrical energy. Ironically, coals, which are the dirtiest fuels among hydrocarbons, will be the main fossil fuel for the next two centuries [1].

All these fuels contain major constituents (carbon, hydrogen, oxygen) as well as other materials, such as metal, sulphur and nitrogen compounds. During the combustion process different pollutants as fly ash, sulphur oxides (SO_2 and SO_3), nitrogen oxides (NO_x = NO_2 + NO) and volatile organic compounds are emitted. Fly ash contains different trace elements (heavy metals). Gross emission of pollutants is tremendous worldwide. These pollutants are present in the atmosphere in such conditions that they can affect man and his environment.

Air pollution, caused by a particulate matter and other pollutants, not only affects directly on environment, but also contaminate water and soil and leads to their degradation. Wet and dry deposition of inorganic pollutants leads to acidification of environment. These phenomena affect human health, increase corrosion, and destroy plants and forests. Widespread forest damages have been reported in Europe and North America. Many cultivated plants are not resistant to these pollutants either, especially in the early period of vegetation.

Mechanisms of pollutants transformation in atmosphere are described by environmental chemistry. Photochemistry plays an important role in these transformations. SO_2 and NO_x are oxidized, sulphuric and nitric acids, which are formed in presence of water vapour, fog and droplets.

Another problem caused by human activities is emission of volatile organic compounds to the atmosphere. These emissions cause stratospheric ozone layer depletion, ground level photochemical ozone formation, and toxic or carcinogenic human health effects, contribute to the global greenhouse effect, accumulate and persist in environment [2].

Waters in the open and underground reservoirs are being polluted, cultivated soil and forests degraded. Most of the plants, especially coniferous trees, are not resistant to sulphur oxides discharged from municipal and industrial facilities. Water pollution used to be primarily a local problem, with identifiable sources of pollution by liquid waste. Up to a few decades ago most of the wastes discharged to waters came from animal and human excreta and other organic components from industry.

In areas with low population density and without sewerage systems such problems are alleviated to a great extent by the natural self-purification capacity of the receiving water. However, with the

increasing urbanization of the last two centuries and a subsequent expansion of sewerage systems without any or adequate treatment, liquid waste loads have become so large that the self-purification capacity of receiving water downstream of large human settlements can no longer prevent adverse effects on water resources.

The other problem concerns industrial effluents, which carry out chemical contaminations, heavy metals, organic pollutants, most often petrochemicals, pesticides, dyes etc. Some pollutants are synthesized in situ, as for example chloroorganic compounds originating from chlorine application for water/wastewater disinfections. The results of discharges of such materials include dying living water reservoirs inhabitants, risk of infection, health effects caused by contaminated drinking water and offensive smells. Over the years, the pollution load of most receiving waters has further increased. In addition to impacts from point sources, pollution from non-point (diffuse) sources, for example leaching and runoff from agricultural areas and long-range transported air pollutants, have become increasingly important [3].

Consequently, the associated problems are no longer just local or regional, but have become continental in scope. The situation regarding environment contamination is becoming critical. Therefore the economically and technically feasible technologies for pollution control, gaseous and liquid effluents streams are searched for. The radiation offers advanced solutions to the selected problems as well [4, 5].

2. RADIATION PROCESSING OF GASEOUS SYSTEMS

2.1. Background

Wet flue gas desulphurisation (FGD) and selective catalytic reduction (SCR) can be applied for flue gas treatment and SO_2 and NOx emission control. Volatile organic compounds (VOC) are usually adsorbed on carbon, but this process is rarely used for lean hydrocarbon concentrations up to now. All these technologies are complex chemical processes and wastes, like wastewater, gypsum and used catalyst, are generated [6].

Electron beam technology is among the most promising advanced technologies of new generation. This is a dry-scrubbing process of simultaneous SO_2 and NOx removal, where no waste except the by-product is generated. Researches show that irradiation of flue gases with an electron beam can bring about chemical changes that make removal of sulphur and nitrogen oxides easier. The main components of flue gases are N_2, O_2, H_2O, and CO_2, with much lower concentration of SOx and NOx. NH_3 may be present as an additive to aid removal of the sulphur and nitrogen oxides. Radiation energy is absorbed by gas components in proportion to their mass fraction in the mixture. The fast electrons slow down, and secondary electrons are formed which play important role in overall energy transfer.

After irradiation, fast electrons interact with gas creating various ions and radicals. Primary species formed include e-, N^{2+}, N^+, O^{2+}, O^+, H_2O^+, OH^+, H^+, CO_2^+, CO^+, N_2^*, O_2^*, N, O, H, OH, and CO. In the case of high water vapour concentration the oxidizing radicals $OH^\cdot$ and $HO_2^\cdot$ and excited ions as O (3P) are the most important products. These species take part in a variety of ion-molecule reactions, neutralization reactions, and dimerization [7].

The SO_2, NO, NO_2, and NH_3 present cannot compete with the reactions because of very low concentrations, but react with N, O, OH, and HO_2 radicals. After humidification and lowering its temperature, flue gases are guided to reaction chamber, where irradiation by electron beam takes place. Ammonia is injected upstream the irradiation chamber. There are several known pathways of NO oxidation. In the case of electron beam treatment the most commons are [8]:

$$NO + O(^3P) + M \rightarrow NO_2 + M$$

$$O(^3P) + O_2 + M \rightarrow O_3 + M$$
$$NO + O_3 + M \rightarrow NO_2 + O_2 + M$$
$$NO + HO_2^{\cdot} + M \rightarrow NO_2 + {}^{\cdot}OH + M$$

After the oxidation NO_2 is converted to nitric acid in the reaction with $OH^{\cdot}$ according to the reaction:

$$NO_2 + {}^{\cdot}OH + M \rightarrow HNO_3 + M$$

HNO_3 aerosol reacts with NH_3 giving ammonium nitrate that can be written:

$$HNO_3 + NH_3 \rightarrow NH_4NO_3$$

Partly NO is reduced to atmospheric nitrogen.

There can be also several pathways of SO_2 oxidation depending on the conditions. In the electron beam treatment the most important pathways are radio-thermal and thermal reactions [9].

Radio-thermal reactions proceed through radical oxidation of SO_2 in the reaction:

$$SO_2 + {}^{\cdot}OH + M \rightarrow HSO_3 + M$$

Then HSO_3 creates ammonium sulphate in the following steps:

$$HSO_3 + O_2 \rightarrow SO_3 + HO_2$$
$$SO_3 + H_2O \rightarrow H_2SO_4$$
$$H_2SO_4 + 2NH_3 \rightarrow (NH_4)_2SO_4$$

The thermal reaction is based on the following process:

$$SO_2 + 2NH_3 \rightarrow (NH_3)2SO_2$$

$$(NH_3)2SO_2 \rightarrow (O_2, H_20) \rightarrow (NH_4)2SO_4$$

The total yield of SO_2 removal consists of the yield of thermal and radio-thermal reactions that can be written [10, 11]:

$$\eta SO_2 = \eta 1(\phi, T) + \eta 2(D, \alpha NH_3, T)$$

The yield of the thermal reaction depends on the temperature and humidity. It decreases with the increase of the temperature. The yield of radio-thermal reaction depends on the dose, temperature and ammonia stoichiometry. The main parameter in NOx removal is the dose. The rest of parameters play minor role in the process. Nevertheless, in real industrial process, dose distribution and gas flow conditions are important from the technological point of view [12]. To achieve reduction of energy consumption, combined EB/MW process has been investigated [13].

2.2. Radiation treatment of SO_2 and NOx

Japanese scientists demonstrated in 1970 -1971 the removal of SO_2 using an electron from a linear accelerator (2-12 MeV, 1.2 kW). A dose of 50 kGy at 100^0C led the conversion of SO_2 to an aerosol of sulphuric acid droplets, which were easily removed [14].

Ebara Co. used an electron accelerator (0.75 MeV, 45 kW) to convert SO_2 and NO_x into a dry product containing $(NH_4)_2SO_4$ and $(NH_4)_2SO_4$,.NH4NO$_3$ which could be used as a fertilizer. Using the "Ebara process", two larger scale plants were constructed in Indianapolis, USA [15] and Karlsruhe, Germany [16]. The Indianapolis plant was equipped with two electron beam accelerators (0.8 MeV, 160 kW) and had a capacity of 1.6 - $3.2 \cdot 10^4$ m^3/h with gas containing 1000 ppm SO_2 and 400 ppm NO_x. In Karlsruhe, two electron accelerators (0.3 MeV, total power 180 kW) were used to treat 1 - $2 \cdot 10^4$ m3/h flue gas containing 50 - 500 ppm SO_2 and 300 - 500 ppm NO_x.

However, the final engineering design technology for industrial applications was achieved at the pilot plants being operated in Nagoya, Japan [17] and Kaweczyn, Poland [18]. In the case of the last, new engineering solutions were applied; double – longitudinal gas irradiation, air curtain separating secondary window from corrosive flue gases and modifications of humidification/ammonia system (high enthalpy water or steam injection, ammonia water injection) and others. The obtained results (Fig.1) have confirmed physico-chemistry of the process, which was discussed earlier. A high dose is required for NO_x removal, while SO_x is removed in proper conditions, at low energy consumption.

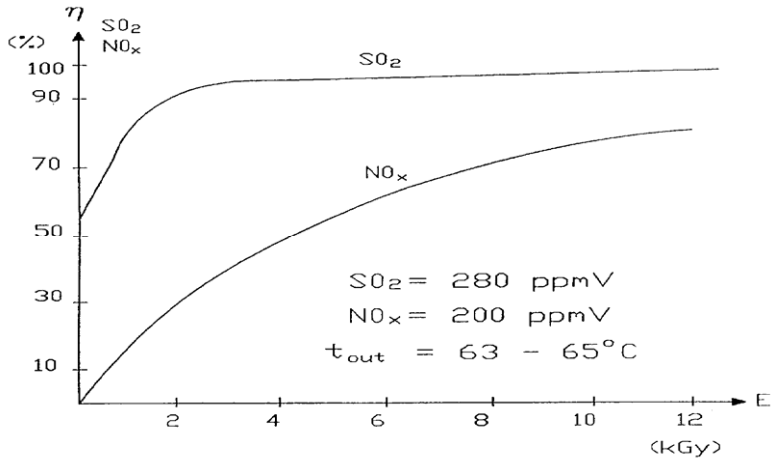

FIG. 1. SO_2 and NO_x removal efficiency vs. dose. The results obtained by the pilot plant experiments and theoretical calculations.

These new solutions led to economical and technical feasibility improvement and final industrial scale plant construction. Ebara Corporation has constructed full scale plant in Chengdu (Fig. 2), China mostly for SO_x removal, therefore the power of accelerators applied is 320 kW for treatment of 270 000 cubic meters per hour of the flue gas. Reported efficiency is 80% for SO_x and 20% for NO_x [19].

FIG. 2. Flue gas cleaning plant in Chengdu, China.

The flue gas treatment industrial installation is located in EPS Pomorzany in Szczecin (Fig.3) in the north of Poland [20]. The installation purifies flue gases from two Benson boilers of 65 MWe and 100 MWth each. The maximum flow rate of the gases is 270 000 Nm3/h and the total beam power exceeds 1MW. There are two reaction chambers with nominal flow gas rate of 135 000 Nm^3/h. Each chamber is irradiated by two accelerators (260 kW, 700 keV), which are installed in series. The applied dose is in the range of 7-12 kGy. The removal of SO_2 approaches 80 - 90% in this dose range, and that of NOx is 50-60%. The by-product is collected by the electrostatic precipitator and is shipped to the fertilizer plant.

FIG. 3. General view of EPS Pomorzany.

The installation consists of four main parts:

- flue gas conditioning unit
- ammonia storage and dosage unit
- reaction chambers
- by-product collecting and storage unit.

As it was previously mentioned the removal efficiency depends strongly in the process conditions. The highest obtained efficiency for SO_2 reaches 95%, while for NOx it reaches 70% (Fig. 4). The obtained results may be compared with previously reported, based on the pilot plant experiments and theoretical calculations, presented in Fig. 1. The very good agreement between results obtained may be noticed.

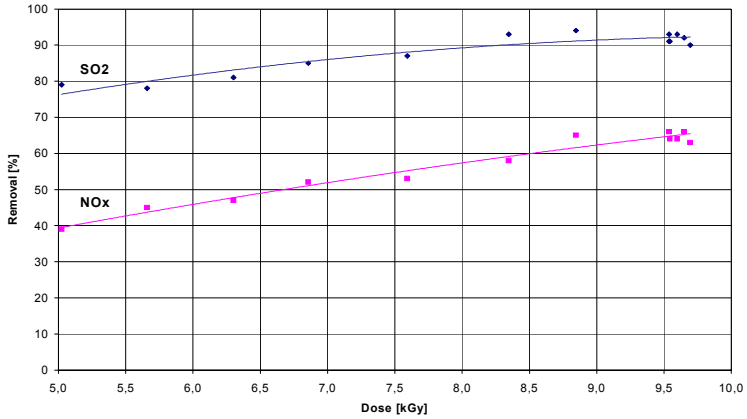

FIG. 4. SO_2 and NO_x removal efficiency vs. dose. The results obtained at the industrial installation. (SO_2 inlet conc. 1500 – 1630 mg/Nm^3, NO_x inlet conc. 470 – 540 mg/Nm^3).

The data obtained during the operation of the installation confirmed the previously taken theses on the impact of the process parameters on the removal effectiveness. In the case of NOx removal the most important parameter is the dose. The inlet concentration of NOx is the second parameter. The impact on this pollutant removal was observed. The correlation is linear and total removal (taken in mg/Nm^3) increases with the inlet concentration of NOx, while the relative removal (in %) decreases with the increase of parameter. The ammonia stoichiometry factor has a very little impact on the NOx removal.

In the case of SO_2 removal there are more parameters, which affect this pollutant removal efficiency. The most important among them is temperature of the gas down stream humidification tower due to the thermal reactions contribution. Afterwards, the dose should be mentioned. Although the humidity seems to have the major impact on the process efficiency, it is hard to prove it with no doubt because of the strong correlation between the humidity and the temperature of the process (dew point temperature is a factor of both parameters: humidity and temperature). During the water evaporation process the temperature decreased and humidity increased. High influence of ammonia stoichiometry ratio on the SO_2 removal efficiency has been observed. The other factors as flue gas flow rate and inlet concentration have much less impact on the removal efficiency. During the experiments one more parameter having impact on the whole process has been detected: the ammonia injection mode. It was observed that the injection of part of ammonia water directly to humidification tower increased the SO_2 removal efficiency. This phenomenon is under investigation now.

The pilot plant for high sulfur coal lignite fired boiler off-gases treatment has been constructed in the Maritsa 2 East Thermal Power Station (Fig.5) in Bulgaria. For high SOx and humidity flue gases, high efficiencies for SOx and NOx removal have been reported.

FIG. 5. General View of Maritsa – East 2 TPP PLC.

2.3. Radiation induced VOC removal from off gases

In the case of VOCs decomposition the process itself is based on similar principles as primary reactions concerning SO_2 and NOx removal i.e. free radicals attack on organic compounds chains or rings causing VOCs decomposition [21].

For chlorinated aliphatic hydrocarbons' decomposition (e.g. chloroethylene), Cl-dissociated secondary electron – attachment and Cl, OH radicals reaction with VOCs plays very important role for VOCs decomposition.

For aromatic hydrocarbons, VOCs decomposition will mainly go through:

1. *Positive ions' charge transfer reactions*

$$M+ + RH\ (RH=VOC,\ e.g.\ Benzene\ or\ PAHs) = M + RH+$$

Because RH has lower ionisation energy (IE) (Benzene: IE = 9.24 eV; PAHs: IE <10 eV) than most primary positive ions (IE > 11 eV) formed above, part of VOC will be decomposed by rapid charge transfer reactions.

2. *Radical – neutral particles reactions*

OH radical plays very important role for VOC decomposition, especially when water concentration is 10 %. $^{\cdot}OH$ radicals react with VOC in two ways:

- $^{\cdot}OH$ radical addition to the aromatic ring (e.g. toluene)
- $^{\cdot}OH + C_6H_5CH_3 = R1^{\cdot}$

and H atom abstraction (for the alkyl-substituted aromatic compounds) or H atom elimination (for benzene, naphthalene and the higher polycyclic aromatic hydrocarbons)

- $C_6H_5CH_3 + {}^{\cdot}OH = R2^{\cdot} + H_2O$ (H atom abstraction)
- $C_6H_6 + {}^{\cdot}OH = C_6H_5OH + H$ (H atom elimination)

Radicals ($R1^{\cdot}$, $R2^{\cdot}$) formed above go though very complex reactions: O_2 addition, O atom release, aromatic –CHO (-dehydes), -OH (-ol) compounds formed or ring cleavage products:

$$R^{\cdot}\ (R1^{\cdot}, R2^{\cdot}) + O_2 = RO_2^{\cdot}$$
$$2\ RO2^{\cdot} = 2RO^{\cdot} + O_2$$
$$RO2^{\cdot} + NO = RO^{\cdot} + NO_2$$
$$RO^{\cdot} + O_2 = HO_2^{\cdot} + products\ (aromatic\text{-}CHO,\ \text{-}O)$$
$$RO^{\cdot} \rightarrow aliphatic\ products$$

The possibility of process application for dioxins removal from off gases has been studied [22, 23] and recent pilot studies demonstrated that process is technically and economically feasible [24].

3. RADIATION PROCESSING OF AQUEOUS SYSTEM

3.1. Irradiation of water

Irradiation of water with ionising radiation produces ionised and excited water molecules and free electrons. The excited water molecules quickly return to the ground state. Ionised molecules react in liquid water to form hydroxyl radicals, OH,

$$H_2O^+ + H_2O \rightarrow H_3O^+ + OH$$

The free electrons become hydrated

$$e^- + nH_2O \rightarrow e^-_{aq}$$

The radicals react together or with hydrogen ion (H_3O^+) to form molecules H, HO and H. The yield of radicals and molecular products depends on pH. At low pH, hydrated electron react with hydrogen ion (H_3O^+ or H^+) to form hydrogen atom.

$$e^-_{aq} + H_3O^+ \rightarrow H_2O + H$$

Radical products are highly reactive and are responsible for most of the chemical reactions when aqueous solutions are irradiated. The scheme of the process is given in Fig. 6 [25].

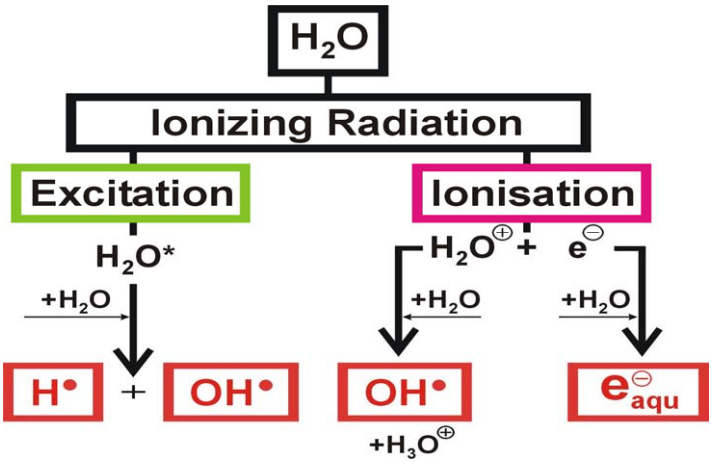

FIG. 6. Water radiolysis: formation of free radical species in water by means of ionizing radiation.

The comparison of processes involved by UV and electron beam irradiation is presented in Table I.

TABLE I. THE FUNDAMENTAL REACTIONS FOR OH RADICAL GENERATION IN DIFFERENT ADVANCED OXIDATION PROCESSES

UV-irradiation	Electron beam irradiation
of aqueous solutions	
$O_3 \xrightarrow{h\nu} O + O_2$ $O + H_2O \longrightarrow H_2O_2$ $H_2O_2 \rightleftharpoons HO_2^{\ominus} + H^{\oplus}$ $O_3 + HO_2^{\ominus} \rightsquigarrow OH$ $H_2O_2 \xrightarrow{h\nu} 2\,OH$	$H_2O \xrightarrow{e^{\ominus}} \begin{cases} H_3O^{\oplus};\ OH^{\ominus} \\ \boxed{OH\ ;\ e^{\ominus}_{aqu}\ ;\ H} \\ H_2\ ;\ O_2\ ;\ H_2O_2 \end{cases}$ $O_3 + \begin{cases} e^{\ominus}_{aqu} \\ H \\ H_2O_2 \end{cases} \rightsquigarrow OH$
Radiation is absorbed by solutes not by water! Always just *one* source for OH (O_3 and H_2O_2, resp.)	Radiation is absorbed by the water not by solutes. Two sources for OH (water radiolysis and O_3 decomposition)

3.2. Radiation purification of waste water

Contamination of surface water and groundwater from industrial waste and anthropogenic activities is a serious problem in many countries. The wide application of fertilizers, pesticides, fungicides can lead to ground water pollution and consequent contamination of drinking water. Population growth and declining fresh water supplies a need for clean water and is one of the critical challenges for the 21st century. Because of the increasing levels and complexity of polluted effluents from municipalities and industry, current wastewater treatment technologies are often not successful for the remediation of polluted waters and disinfection.

The development and implementation of alternative technologies for the clean up of industrial wastewater, municipal water, groundwater and drinking water is critical to the sustainability of many countries. Among the possible water treatment alternatives radiation processing, a very effective form of energy use, can degrade toxic organic compounds and biological contaminants. Prof. Pikaev [26] was a pioneer in development of this technology. Furthermore important research has been performed at Miami pilot facility [27].

Aqueous effluents that have been treated by irradiation include polluted drinking water, liquid industrial and agricultural wastes. However, attention must be paid to the toxicity of the by-products formed in the process what is the main limitation of its implementation. This is the major factor, which has to be carefully studied during the implementation of all Advanced Oxidation Technologies (ozone, ozone+TiO_2, UV) [28]. The differences and similarities of UV and EB water treatment mechanisms are given in Table 1 [29]. The industrial effluents contain variety of pollutants, higher concentrations, and substances that are toxic or difficult to destroy such as salts of mercury and bismuth, cyanides, phenols, and dyes. High doses are generally required to remove such pollutants by irradiation treatment and combined processes, which have been developed in combination with conventional processes such as chemical, biological, or thermal treatment, floatation, and others. Only few full-scale applications are available.

When water containing humic substance is treated with chlorine, carcinogenic chlorinated organic compounds are formed. Studies suggest that comparatively low dose of 1 kGy will bring about decolourisation, deodorization, and disinfection of natural water and dechlorination of organic chlorocompounds present in low concentrations [30].

Electron accelerator (0.7 – 1 MeV, 50 keV) was applied at Voronezh rubber plant in Russia to convert the non-biodegradable emulsifier Nekal in the plant waste into biodegradable form. The dose required to decompose 10^{-3} mol/dm^3 nekal in aqueous solution was 300 kGy. The plant had two production lines and could treat up to $2 \cdot 10^3$ m^3 of effluent per day [31].

The most promising achievements were achieved recently in Republic of Korea, where a pilot plant is in operation [32].

The pilot plant (output 1000 m^{3}day^{-1}) with ELV electron accelerator (energy 1 MeV, beam power 40 kW) is in operation from October 1998. Combined electron-beam and biological treatment was used for purification of dyeing complex wastewater under continuous flow conditions. The main results of pilot-scale experiments consisted in the fact that decrease in total content of pollutants after biological treatment was substantially influenced by preliminary electron-beam treatment (mainly, because of radiolytic conversions of terephthalic acid being a main pollutant of the wastewater). The reduction of non-biodegradable COD into biodegradable BOD compounds was achieved. Equal purification degree corresponded to 17 hours of bio-treatment without preliminary irradiation and about 8 hours of bio-treatment with preliminary electron-beam treatment at absorbed dose 1-2 kGy.

An industrial plant is going to be constructed. Based on the data obtained in the laboratory and pilot plant experiments, the suitable doses are determined as around 0.2 kGy for the flow rate of 10 000 m^3 effluent per day. Therefore, accelerator with the power of 400 kW is applied for economies and compactness of the plant. Cost for high power accelerator is around $US 2.0 ~ 2.5 M and building, piping, other equipment and construction works could be estimated $US 1.0 ~ 1.5 M. Even by considering the additional cost for tax, insurance and documentation (which is $US 0.5 M), the overall cost for plant construction is stipulated to be approximately $US 4.0 ~ 4.5. This sum doesn't include cost for land, R & D and the authority approval. Expected construction period includes 11 months in civil and installation works and 3 months for trial operation. To estimate the operation cost, the electricity consumption is estimated for accelerator with 500 kW (80% efficiency) and other equipment in additional 300 kW to the total of 800 kW. Based on the year round operation (8400 hr/yr), it costs 336 000 $US/yr when the cost of electricity (kWh) was assumed to be $US 0.05.

The labour cost is calculated 3-shift with one additional operator and is approximately 100 000 \$US/yr. Thus, the actual operation cost for 100 000 m^3/day plants is 430 000 \$US/yr and if we consider the interest and depreciation of investment, the cost comes up to around 1 \$USM/yr. It is approximately 0.12 \$US/m^3 for construction and 0.03 \$US/m^3/yr for operation, so is inexpensive compared to other advanced oxidation techniques such as ozonation, UV techniques etc.

3.3. Radiation induced removal of heavy-metal ions from water

The toxic metals from industrial effluent streams include heavy metals such as lead, mercury, cadmium, nickel, silver, zinc, and chromium. These heavy metals are accumulated in soil and eventually are transferred into human food chain [33]. Ionising radiation of aqueous solutions generates free radicals, radical ions and stable products:

$$H_2O \rightarrow e^-_{aq}, H; OH, H_2O_2, H_2, H_3O^+$$

with yields (G value) of 0.28(e^-_{aq}), 0.062(H); 0.28(OH), 0.072(H_2O_2), 0.047(H_2) in units of μmol/J

The hydrated electron e^-_{aq} is the strongest reducing agent.

$$e^-_{aq} + H_3O^+ \rightarrow H^\bullet + H_2O$$

$$Cr(VI) + H^\bullet \rightarrow Cr(V)$$

Cr(V) is unstable and is further reduced to the stable Cr^{3+} ions.

$$Pb + e^-_{aq} \rightarrow Pb^+$$
$$2Pb^+ \rightarrow Pb + Pb^{2+}$$

Lead can also be reduced by $H^\bullet$ atoms

$$H^\bullet + Pb^+ \rightarrow PbH^+$$
PbH^+ decays to produce Pb
$$2PbH^+ \rightarrow H_2 + Pb^{2+} + Pb$$
$$HgCl_2 + e^-_{aq} \rightarrow HgCl + Cl^-$$
$$HgCl_2 + H^\bullet \rightarrow HgCl + Cl^- + H^+$$

HgCl is not stable and dimerizes to Hg_2Cl_2 as a final insoluble products

$$2HgCl \rightarrow Hg_2Cl_2$$

The hydroxyl radical ($^\bullet$OH) is one of the powerful oxidizing species, which lead to transformation of metal ions to the higher valence states [34]. However, due to the fact that normally concentrations of heavy metals in wastewater are very low (ppms), the process seams to be no technically feasible, since trace quantities of reduced metals have to be separated on mechanical way from the wastewater. For higher concentrations chemical (precipitation, ion exchange) or physical methods (membranes, electrolysis) are more feasible from economical or technical points of view.

4. RADIATION PROCESSING OF SOLID STATE SYSTEMS AND SLUDGE

4.1. Municipal sewage and sludge

Electron beam irradiation is a practical and economic method for disinfecting liquid municipal wastes and sludge. Deer Island Electron Research Facility in Boston found a dose of 0.5 kGy was sufficient to disinfect municipal wastewater effluent and also to decompose organic pollutants. Takasaki Radiation Chemistry Research Establishment found that a dose of about 0.4 kGy was required in order to disinfect raw wastewater prepared by mixing primary and secondary sewage effluents [35].

Research has shown that sewage sludge can be disinfected successfully by exposure to high-energy radiation. At a plant near Munich doses of 2-3 kGy destroy more than 99.9% of bacteria present in sewage sludge, and at a plant near Boston a slightly higher dose (4 kGy) was used. Higher doses (up to 10 kGy) are required to inactivate more radiation resistant organisms at plants in Albuquerque and Ukraine. Both gamma sources (Co-60, Cs-137) and electron accelerators can be used for irradiation of sewage sludge. Gamma sources have better penetration allowing thicker layers of sludge to be irradiated [36], although they are less powerful and take longer irradiation time than electron sources [37].

The pilot plant using gamma source is operated in India. The irradiator system can be easily integrated with conventional treatment plant with flexibility of operation. Various dose treatments can be imparted to sludge with addition of sensitising agents such as oxygen, air, ozone etc. The radioactive source loading, unloading or transport is very easy and very safe. It can be accomplished in a day. After augmentation of source strength in early 2001, 12 cubic meter of sludge is irradiated in one shift (yielding 5 ton sludge per month). About 3 kGy of absorbed dose in sewage sludge removes 99.99% of pathogenic bacteria consistently and reliably in a simple fashion.

The process of hygienisation of sewage sludge using radiation is very simple. The incoming sludge is taken to an underground reservoir. It is then fed to irradiation vessel of 3 m3 capacity and circulated continuously in a loop for a pre-determined period. After the radiation exposure the treated sludge is withdrawn from irradiation vessel and pumped out to drying sand beds where the water evaporates yielding pathogen free dried sludge. The irradiated sludge being pathogen free can be beneficially used as manure in the agricultural fields as it is rich in nutrients required for the soil. When performed in villages of Baroda city initial field trials of sludge as manure in agriculture fields in winter wheat crops as well as in summer green gram crops have been very encouraging and have prompted farmers for putting increasing demands. Since the irradiated sludge is free from bacteria, this can also be used as a medium for growing soil useful bacteria like rhizobium and azetobactor to produce bio-fertilizers, which can be used to enhance the crop yields.

4.2. Soil remediation

US Environmental Protection Agency (EPA) has determined that polychlorinated dibenzodioxins (PCDDs) and polychlorinated dibenzofurans (PCDFs) present serious public health risk and set limits on storage, transport, and disposal of waste materials containing dioxins. A limit of 1 ppb has been established for 2,3,7,8-tetrachlorodibenzo-p-dioxin (TCDD), which is the most toxic member of this family of compounds. Studies have demonstrated that TCDD can be converted to products of negligible toxicity by radiolysis with gamma rays from cobalt-60. Destruction of greater than 98% was achieved with a dose of 800 kGy in a soil contaminated with 100 ppb of TCDD. Addition of contaminants such as dichlorobenzene and hexachlorobenzene did not affect the result. The addition of 25% water and 2.5% nonionic surfactant was beneficial with the model soil [37].

4.3. Mail decontamination

Anthrax that was sent in the mail in October 2001 caused several deaths and big economic losses in the USA. The radiation proved to be very effective for mail decontamination. About 4000 tons of letter mail and 200 tons of parcels had been sanitized by the end of 2003 [38, 39].

5. CONCLUSIONS

(1) Electron beam flue gas treatment (deSOx and deNOx) has been implemented in industrial scale in Poland and China.
(2) Electron beam flue gas treatment has been proved to be effective for VOC and PAH removal. The technology has been investigated in industrial conditions for flue gases from coal fired boiler and municipal solid wastes incinerator plant. Toxicity reduction is the efficiency measure to different byproducts formed.
(3) Regarding the treatment of organic pollutants in the wastewater, similar to other AOT, byproducts formed, have to be considered and toxicity tests are the best parameter of the process efficiency. Combined eb/biological process has been studied in pilot scale in Korea.
(4) Biological contamination of the secondary effluents seems to be the most promising application at the moment and an industrial plant applying the process is constructed in Korea.
(5) Pilot plant for gamma rays sludge hygenization has been in operation in India for several years. The technology proved its effectiveness and the product is a fertilizer of a good quality.
(6) New application of the technology based on the accelerator has been mail decontamination against bio-terrorist agents.

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ECONOMIC EVALUATION OF ELECTRON BEAM FLUE GAS TREATMENT

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Abstract

The industrial scale electron beam flue gas treatment plant for simultaneous removal of sulphur and nitrogen oxides from flue gases has been built in electric power station Pomorzany in Szczecin, Poland. The plant may purify up to 270 000 Nm^3/h of flue gases with the efficiency as high as 95% for SO_2 and 70% for NO_x. The total power of four installed accelerators reaches 1.04 MW that makes it the biggest radiation processing facility in the world. After the completion of the investment the operational and economic data have been obtained. Basing on the gathered data the possible costs of EBFGT installations have been evaluated.

1. INTRODUCTION

The impact of the pollutants being emitted to atmosphere during fossil fuels combustion (mainly SO_2 and NO_x) on environment and humans is well known [1]. The biggest sources of the pollutants' emission of human origin are thermal power plants. Therefore present and foreseen law regulations impose the application of emission control technologies in all kinds of such plants. Among the number of flue gas desulphurisation (FGD) methods the most common used are wet FGD technologies [2]. Also NO_x removal processes are realised by selective catalytic and non-catalytic reduction (SCR and SNCR) methods [3, 4]. Although these solutions are well known they are still two separate facilities with numerous disadvantages as: high complication level and space requirements, solid and liquid waste generation or high cost.

Therefore apart of the combined (SO_2 + NO_x) systems, the technologies for simultaneous removal of both pollutants were sought. Many different options have been investigated, but according to our knowledge, only one was implemented in the industrial scale – the electron beam process [5]. The solution saves both energy and space that is especially important in the case of retrofitting. Moreover the technology is dry and with the agricultural use of byproduct, which is a mixture of ammonium salts, neither solid nor liquid waste is generated. The technology is simple in operation, control and other technical issues. According to recent research there is also the possibility to remove volatile organic hydrocarbons (VOC's) in this process [6]. All this advantages make it competitive to conventional technologies not only from technical and operational point of view, but also economically.

The technology is quite a new one. There are only three industrial installations in the world: two in China and one in Poland. At present the NO_x emission in China is not limited, so these two installations were designed for SO_2 removal with low NO_x removal efficiency. Opposite to this the Polish installation was designed for high efficiency of removal of both pollutants. The installation was constructed and proved its advantages in normal operation. After that the summary of the investment was possible. This paper present the technical and economical aspects of electron beam technology based on the experiences gathered during the construction and operation of Polish plant.

2. PLANT DESCRIPTION

2.1. Plant design

The installation consists of four main units clearly distinguished both from constructional and functional point of view. Each of the units may be realised in various ways. Some of the solutions are presented below.

- cooling and humidification of flue gases – unit responsible for the adjustment of the desired temperature and humidity of flue gas;
- ammonia supply system – unit responsible for dosing of exact amount of main process reagent and for the proper way of dosing;
- reaction unit – the main unit of the installation, where the process of pollutants removal and byproduct creation occurs;
- Byproduct filtration and storage – the unit for separation of byproduct particles from flue gas.

2.2. Cooling and humidification unit

The first operation unit is cooling and humidification. The flue gases from combustion process usually have to high temperature and too low humidity for optimal reduction of SO_2 and NO_x. The proper change of both parameters my be obtained by evaporation of water in inlet stream of flue gas. The optimal temperature after cooling should be from 60 to 75°C and optimal humidity 12 to 14% vol. The optimal temperature and humidity of flue gas by evaporation of water is possible for high values of inlet temperatures (T_0) of gas of order 170°C to 190°C. It depends on humidity of flue gas, w_0. If humidity is higher the temperature T_0 may be lower. Evaporation of water in flue gas may be done in different ways:

- cooling column with dry bottom
- cooling column with circulation of water
- cooling in ducts

Cooling column with dry bottom is simplest solution but has also some disadvantages. In dry bottom column water should be sprayed into very fine droplets with diameter less then 100 μm. It means that should be applied two medium air - water nozzles. Such nozzles need a lot of compressed air. If temperature of inlet flue gas is lower than 180°C humidity of flue gas after cooling is too low and should be increased by introduction of steam, which increase operational costs. Good operating cooling column with dry bottom have also dry walls and the corrosion, problems are not significant.

Cooling column with circulation of water was tested in Kaweczyn, Nagoya and Maritza II e-b pilot plants and applied in Chengdu industrial plant. It is good when plant is designed mainly for removal SO_2 and low temperatures after humidification are provided. If temperature of inlet flue gas is low there might be too low humidity after cooling and the circulation water should be heated. It introduce additional significant stream of heat for evaporation of more water. The temperature of outlet gas T is of order 55 to 65°C. If it is to low temperature for the process, part of flue gas can be by-passed the column.

Advantages of this process are:

- water droplets can be much bigger and can be installed one medium water nozzles. This reduces costs of compressed air.
- this type of column reduces significantly dust content in flue gas.

Disadvantages are:

- more serious corrosion problems
- generation of some waste water containing collected dust and absorbed HCl and SO_2.

The third possibility is spraying of water and its evaporation in flue gas ducts. Of course it is possible in big and long ducts. It is possible fine pulverisation of water and its full evaporation and system with circulation of water. According to our knowledge such solution was not yet tested in e-b technology. That solution can reduce investment costs from 5 to 7%.

2.3. Ammonia supply system

Usually ammonia is supplied and stored in form of liquid ammonia in pressurized tanks. Before injection into flue gas, liquid ammonia is evaporated in a small evaporator. Ammonia in contact with SO_2 forms solid salts which settle near ammonia nozzles. A special devices near the spray nozzles should be installed for prevent of plugging the nozzle orifices by deposit. Some amounts of salts also settle on the walls of ducts after ammonia introduction point. Another system of introduction of ammonia was elaborated by EBARA Co. In this system ammonia is mixed with hot air and sprayed in gas - water nozzles at inlet to reactor. Water dissolved ammonia salts and prevents formation of deposits. This system is good for low temperature process, in which mainly SO_2 is removed.

Other option is using ammonia water as a source of ammonia. There was no experience in pilot scale with such solution before. There may provide two versions of ammonia introduction. In the first version gaseous ammonia is separated from ammonia water in distillation column and introduced into flue gas ducts. In the second version ammonia water is sprayed and evaporated in cooling column. Spraying of ammonia water needs separate system of nozzles. After the preliminary tests it was found that the best is mixed version in which the part of ammonia is sprayed in cooling column in form of ammonia water and the rest is separated in distillation column and introduced as a gas. Application of ammonia water increases the operation and investments costs. Also ammonia dissolved in ammonia water costs more then pure liquid ammonia. In this way if the safety conditions allow application of liquid ammonia, such solution should be applied. Mixing of ammonia in flue gas stream usually doesn't make a problem if ammonia is injected in many points because flue of gas stream is turbulent and good mixing conditions occur in the ducts.

2.4. Reaction unit

Reaction unit consist of accelerators with:

- power suppliers
- water cooling system
- windows cooling system
- windows
- reaction chamber
- X-radiation shielding with ventilation system.

Accelerators are the most important and the most technological advanced apparatus in e-b technology. Only a few companies in the world produce high power electron accelerators, which can be applied in this technology. The price of accelerator depends on its power. The power of accelerator is proportional to mass flow of flue gas and the dose that is necessary for removal of SO_2 and NOx. Generally for efficient reduction of SO_2 the enough dose is 4 to 5 kGy. Reduction of NOx usually needs doses of 8 to 12 kGy. Than it should be taken into consideration combined processes for example installation of low NOx burners that allows using of accelerators with lower power and lower cost. For this technology there were constructed the biggest accelerators in the world and its technology is still improved. The main parameters of accelerator are energy of electrons and beam current. For the electron beam technology there are applied accelerators with energy of electrons up to 800 keV and beam current up to 500 mA. That values are limited by the power of power suppliers.

Up to now accelerators are the most sensitive part of this technology. Producers provide time of operation up to 6500 hours per year while other installations in power plants work about 8000 hr. per year. Also costs of spare parts and maintenance by manufacturer are very high. We hope that progress in technology, challenge between producers as well as more orders from industry applied e-b flue gas treatment technology will cause significant decrease of the accelerators' cost and will reduce the cost of their maintenance.

The auxiliary installations like water cooling system, air cooling of windows usually work without problems. The windows, particularly with the secondary windows located at reactor need more careful construction. First this window needs good sealing. The leak causes cooling of window and formation of deposits of salts which causes corrosion of titanium foil. Also the construction of the window should be adjusted to avoid vibrations of the foil that can lead to their destruction. It seems that there should be developed easy exchangeable frames with windows allows in short time replace broken foil by new one.

Flue gas is irradiated in reaction chamber. It is empty tube with circular or rectangular cross section. Diameter of the tube should be adjusted to penetration range of electrons in irradiated gas. The dose deposition in the cross section of the reactor is not uniform. The theoretical studies carried out in Institute of Nuclear Chemistry and Technology showed that using not uniform flow that means increasing the gas velocity in the space, where higher energy is deposed may improve the removal efficiency 10 – 14%. Air cooling the windows is strongly irradiated and it generates some ozone. Up to now it is removed into atmosphere by ventilation system. It seems that that air can be used in the process or in combustion chamber of the boiler.

2.5. Filtration unit

After irradiation and the radical and chemical reactions the ammonium salts condense in very fine submicron aerosols. It should be separated from stream of flue gas because their emission would make big damages in environment and because it is valuable product. There were a few kinds of filters tested in pilot plants:

- electrostatic precipitator
- bag filter
- gravel bed filter
- Venturi scrubber.

The two first filters are dry and the last two are wet filters. The wet methods of filtration are very complicated and generate some amount of wastewater that needs further treatment. They are not recommended in the case of dry technology.

Bag filter however has very good efficiency and gives additional removal of SO_2 in layer of filter cake was not applied in industrial plant because of not sufficient cleaning of filter bags from filter cake. The product is too sticky and used in tests pulse-jet method of cleaning is not enough good. It seems that should be tested other methods of bag cleaning.

The electrostatic precipitator was used in industrial scale plants and proved their usability for this technology. Product precipitated in ESP is hygroscopic and if it is wet the corrosion may occur. For protection from the condensation of water the electrically heated bottom of ESP is used.

2.6. Description of the Pomorzany electron beam flue gas treatment plant

Pomorzany electron beam flue gas treatment plant is rather small and retrofit installation. The station purifies up to 270 000 Nm^3/h of flue gas from two hard coal fired Benson boilers of 65 MW_e each. The temperature of flue gas is up to 140°C and inlet concentration of pollutants about 2000 mg/Nm^3 of SO_2 and 400 – 600 mg/Nm^3 of NO_x with low dust concentration (after electrostatic precipitators). The installation purifies about half of the total amount of flue gases, while the rest is bypassed and mixed with the treated part before stack. In this way the temperature in the stack is above 110°C that reduces the problems with stack corrosion. The scheme of the installation is presented in figure 1.

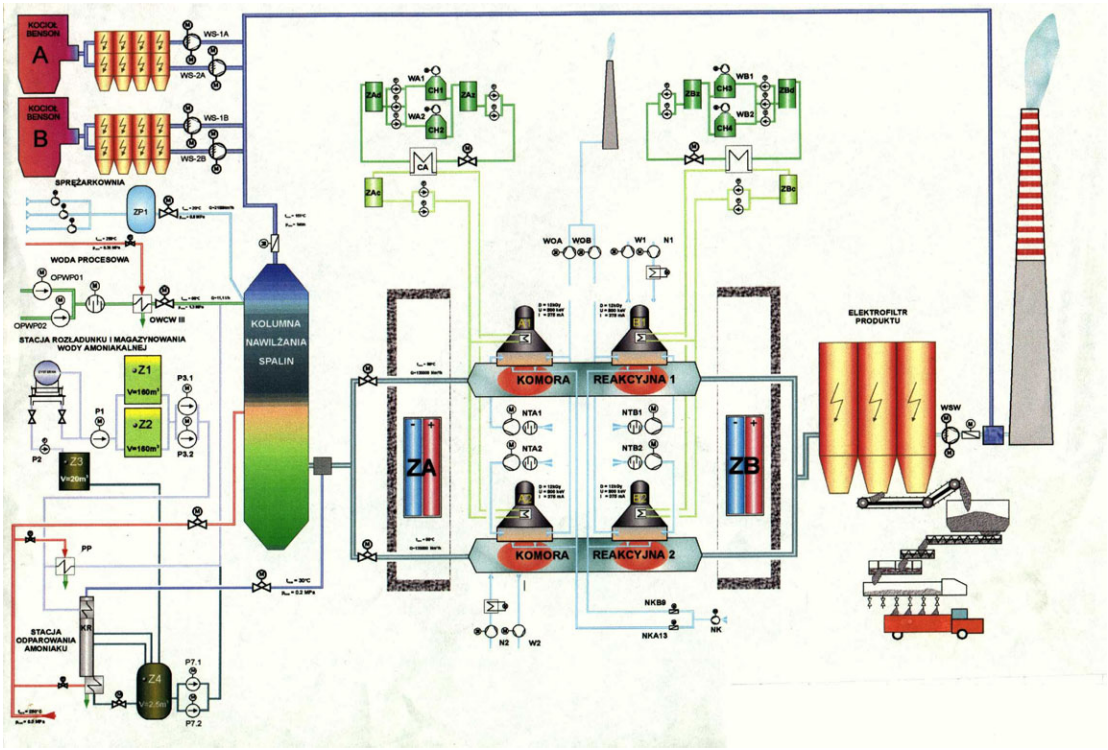

FIG. 1. The scheme of the Pomorzany electron beam flue gas treatment plant

For the above described conditions the dry bottom cooling tower has been chosen. This solution is appropriate for medium inlet temperatures and low dust concentration. Although it requires very careful water atomisation it is simple and very useful construction.

The local conditions (electric power station is situated near the living area) and local law regulations ammonia water was allowed as a source of ammonia. In this way the double system of ammonia dosage was applied. Part of ammonia is evaporated and injected into the flue gas before the reaction chambers, while the rest is sprayed straight to cooling tower. As it was noticed during the operational tests the ratio between ammonia water and gaseous ammonia has great impact on the SO_2 removal efficiency [7]. The reaction unit was implemented as a system of two parallel, cylindrical reaction chambers. The flue gas in each reactor is irradiated by two accelerators (700 keV, 260 kW) installed in series. Such a solution provides the proper irradiation of flue gas. The obtained byproduct is collected in electrostatic precipitator and granulated before storage. The granulation reduces the dusting of the byproduct and increases its bulk density. After that the byproduct is sold to fertilizer producing company as a substrate for NPK mixtures.

3. OPERATIONAL RESULTS

The efficiency of pollutants removal that can be achieved by electron beam flue gas treatment installation is comparable with the conventional ones. As was reported in Pomorzany electron beam flue gas treatment plant the SO_2 removal efficiency exceeds 95% while NO_x removal efficiency reaches 70% . The experiences of Pomorzany plant operation are described more detailed elsewhere [7, 8], but it is worth to underline the most important factors that have impact on the process. From the SO_2 removal point of view the most important factors are temperature and humidity of the process and ammonia stoichiometry due to the thermal way of this pollutant removal. On the other hand the impact of the ammonia dosing way on the SO_2 removal efficiency was observed. Dosing ammonia in the form of ammonia water straight to the humidification tower improves the process efficiency comparing with the gaseous ammonia dosing upstream the reaction chamber [8].

The removal of NO_x first at all depends on the dose that means on the energy deposed in the flue gas. The energy is transferred to flue gas by accelerators and this factor is responsible for large part of total energy consumption in the installation. Other parameters important in this process are inlet NO_x concentration and to a lesser degree ammonia stoichiometry. From the chemical point of view NO_x removal process is simpler than in the case of SO_2 and requires less parameters to sufficient description. The full understanding of electron beam flue gas treatment process will allow for better optimization of the process and lowering the operational costs.

The electron beam flue gas treatment plant shows great flexibility according to inlet parameters change. The Pomorzany installation was tested for flue gas flow rate within the range of 100 000 – 270 000 Nm^3/h without any significant efficiency loss. This is almost impossible to achieve in conventional installations. It is also easy to follow the change of other inlet parameters as temperature, inlet flue gas composition or humidity. As was proved by the recent research other pollutants as VOC [6] or dioxins [9] may be removes in this process. The above points, that this method may be used for purification of flue gases from different, not only coal combustion processes.

The byproduct output is in the range of 200 – 300 kg/h. It contains mostly ammonium sulphate (45 – 60%), but due to high efficiency of NO_x removal process it is enriched in ammonium nitrate (22 – 30%). The most important impurity is ammonium chloride (10 – 20%) being formed from coal contained chlorine. The insoluble matter content is in the range 0.5 – 2% and it consists mostly of residuals of fly ash and some rust, so the heavy metals may occur only in traces. The byproduct is a valuable high nitrogen content fertilizer that may be used as a source of nitrogen, while pure ammonium nitrate is explosive. The total nitrogen content is up to 27%, comparing with 21% in ammonium sulphate. The byproduct is sold to the fertilizer factory that lowers the operational costs of the plant.

4. ECONOMIC EVALUATION OF THE INVESTMENT

4.1. Cost estimation

The Pomorzany electron beam flue gas treatment plant is in fact the prototype installation and some solutions were improved during realisation of the investment. It is also important from both technical and economical point of view that this is retrofit installation and local conditions in electric power plant have impacted on the plant construction. The gathered experience as well as economic data of the installation will allow designing of other facilities. Basing on the Pomorzany EB flue gas treatment plant case, the technical and economical characteristic of the plant may be evaluated.

The following economic evaluation was made, after Pomorzany case, for the retrofit electron beam flue gas treatment installation for boiler size about 130 MW_e with similar configuration (i.e. for two reaction chambers). The main technical data are presented below:

- Flue gas flow rate: 300 000 Nm^3/h
- Fuel: hard coal (0.8 – 1% S)
- Inlet flue gas temperature 130 – 150°C
- Inlet flue gas composition:
    - SO_2 1000 – 1500 mg/Nm^3
    - NO_x 400 – 600 mg/Nm^3
    - CO_2 8% vol.
    - O_2 7 – 8% vol.
    - H_2O 9 – 10% vol.
    - N_2 to the balance
    - Fly ash < 50 mg/Nm^3

Basic operational data:

- SO_2 removal efficiency 90%
- NO_x removal efficiency 70%
- Total power of accelerators 1 MW
- Ammonia consumption 100 - 150 kg/h
- Byproduct output 200 – 300 kg/h

4.2. Investment costs of the plant

The foreseen investment costs of concerned installation are assumed for $US 21 million. The main parts of this calculation are presented below (in $US thousand):

- Preliminary works 700
- Licenses and documentation 1 700
- Spray cooler 500
- Reaction chambers 400
- Accelerators' system 5 000
- Media supply system 400
- Ammonia storage and dosing system 500
- Flue gas ducts 800
- Electrostatic precipitator 2 000
- Auxiliary fan 200
- Byproduct handling and storage system 4 000
- Electric power supply 1 800
- Control and monitoring system 800
- Training, supervision, putting in motion 800
- Final works, builder's yard and others 400

The above calculation gives the unit investment costs at a level of 160 US$/$kW_e$. Realising that these costs were evaluated for retrofit installation and further reduction is possible. First at all the retrofit installations cost even up to two times more, then new built ones. This calculation assumed, after Pomorzany case, use of two parallel reaction chambers that needs four accelerators. The use of single reaction chamber (if possible) or smaller accelerators (for less NO_x removal efficiency) may considerably reduce the total cost of the plant. Also the cost of all support systems (as flue gas ducts, media supply systems, civil works etc.) depends on the local conditions. So the total investment cost of other electron beam flue gas treatment plant of similar size should not exceed the amount given above.

On the other hand the unit cost of the EBFGT plant decreases with the facility size increase. As was calculated by Jackowski et al. [10] the unit cost of the small installation, about 35 MW_e, is as high as 300 $US/$kW_e$, while for 200 MW_e it is only 150 $US/$kW_e$. For 300 MW_e and higher plants this cost is about 140 $US/$kW_e$. In this way larger installations although more expensive, becomes more economically competitive.

4.3. Operational costs of the plant

The operational costs of the plant were calculated also for the above assumed operational data. It is worth to notice, that the operational costs of real installations depends on the process carrying way as well as the host pollutant removal needs. Assuming the same depreciation period and interest for better illustration fixed costs has been omitted.

Annual operating costs were assumed for US$ 1125 thousand. The following are important parts of this amount:

- Basic raw materials 140 thousand $US

- Energy consumption 620 thousand $US
- Auxiliary raw materials 45 thousand $US
- Maintenance and spare parts 200 thousand $US
- Direct labour 120 thousand $US

Opposite to most conventional installations the byproduct is fully sellable as a fertilizer. Although it doesn't cover the costs of plant operation (all the flue gas treatment facilities are non profit) it may lower these costs. In addition, the plant may save money reducing the emission penalties. In that way the incomes were calculated for 170 thousand $US:

- Byproduct selling 35 thousand $US
- Saved penalties for emission 135 thousand $US

The total annual cost of plant operating as a difference between expenditures and incomes is 955 thousand US$. The most important part of this sum is energy and maintenance and spare parts – both connected with accelerators. The energy cost may be lowered with lowering the NO_x removal efficiency, while maintenance cost is caused by uniqueness of such large accelerators. According to the warranty rules titanium foils from the scanner windows and accelerators cathodes are to be exchanged every 2000 hours and all the maintenance works are to be done by manufacturer. From our experience these elements should last for longer time, so after the warranty period the maintenance cost might be considerably lowered. Also the other accelerator manufacturer may offer better terms. The lowered number was taken for these calculations.

The unit costs of plant operation may be related either to amount of removed pollutants or to the size of electric power station (the amount of electricity generated). In the first case the operational costs related to the SO_2 removal are $US 1061 per one ton of removed SO_2. But in fact the electron beam installation removes simultaneously both sulphur and nitrogen oxides, so the costs cannot be divided for the costs for removal of SO_2 and NO_x. In this way the operational costs related to both removed pollutants lowers to $US 806 per one ton. Regarding the second case the operational costs of the plant related to boiler size are $US 7346 per one MW_e of installed power.

4.4. The comparison of EB and conventional method costs

Although several attempts have been made the electron beam flue gas treatment method is the first that was applied for simultaneous removal of sulphur and nitrogen oxides in the industrial scale. The most often a combination of wet flue gas desulphurisation and selective catalytic reduction systems is used.

The investment costs of retrofit wet FGD installations are usually 80 – 120 $US/$kW_e$ depending on size and local conditions. It is worth to notice, that this kind of pollution control facility is installed mostly in large power plants of size above 500 MW_e. So in the case of smaller installations (about 120 – 250 MW_e) the investment cost should arise.

On the other hand the retrofit SCR installations' investment costs are 59 – 112 $US/$kW_e$ depending on the plant size and difficulty and scope of retrofit [11]. For new facilities such costs are 45 – 60 $US/$kW_e$.

The investment costs of these two emission control methods seem to be lower, then for electron beam plant. But the cost of both installations taking together is 140 – 230 $US/$kW_e$. For small plants of boiler size 120 – 250 MW_e this cost will be about 200 – 230 $US/$kW_e$, that is considerably more than 160 $US/$kW_e$ for electron beam technology. The cost of combined wet FGD and SCR system is estimated for 270 – 474 $US/$kW_e$ for the units of 300 – 50 MW_e.

In the case of annual operational costs the wet FGD methods cover about 2500 – 3000 \$US/MW_e, while SCR methods cover 3800 – 4600 \$US/MW_e [12]. That means, that removal of both pollutants by a conventional methods costs annually 6300 – 7600 \$US/MW_e.

The comparison of the costs of various emission control methods for a 120 MW_e unit is presented in table I.

TABLE I. THE COSTS OF VARIOUS EMISSION CONTROL METHODS FOR A RETROFIT 120 MW_E UNIT

Emission control method	Investment cost (\$US/kW_e)	Annual operational cost (\$US/MW_e)
Wet flue gas desulphurisation	120	3000
Selective catalytic reduction	110	4600
Wet FGD + SCR	230	7600
Electron beam FGT	160	7350

5. CONCLUSIONS

(1) Three industrial EB plants have proven the ability of the technology for efficient removal of SO_2 and NO_x from flue gases from coal combustion processes.
(2) The EB technology is a new one, but with a great potential spectrum of applies and still needs developing works.
(3) During the erection and operation of Pomorzany EB plant a lot of experience has been collected, that should be used during the design and construction of new facilities. Also the staff taking part in construction of the plant has been specialized in that kind of investments.
(4) Although the costs of this prototype and retrofit installation are relatively high, they are comparable with conventional technologies.
(5) Further development of EB technology can significantly reduce both the investment and operational costs of the plant. Particularly we expect significant reducing of accelerator cost connected with increase of their production and progress in their technology.

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RESULTS OF THE STARTUP OPERATION OF A PILOT INSTALLATION ELECTRON BEAM FLUE GASES TREATMENT IN THE MARITSA EAST 2 THERMAL POWER PLANT

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Abstract

The electron beam flue gases treatment technology has been developed first in Japan later in USA, Germany and Poland. According tree party contract which was sign between IAEA, JAERI and NEK, has been constructed Pilot Plant for electron beam flues gases treatment in Bulgaria. The flow rate of 10000 nm³/h are irradiated by three high energy accelerators of 800 keV and 35 kW beam power each. The plant has been operated since January 2004. The removal efficiency is from 90 -99 % for SOx and 85 -90 % for NOx.. The quality of the coals are with high ash content up to 45%, high moisture up to 57% , the low calorific value from 1196 kcal/kg up to 1603 kcal/kg and high concentration of sulfur made the Bulgarian lignite coals unique in their usage as fuel for the thermal power plants in Maritsa East side.

1. INTRODUCTION

The conference entitled Environment for Europe (Sofia, October 1995) defined as one of the most important initiatives the one to decrease the air pollution with sulfur and nitrogen oxides. These emissions are the main pollutants of the air in Bulgaria, where the real load of 150-160 kg of sulphur oxides per person annually defines the country as one of the “hot spots” on the continent. The international commitments undertaken by Bulgaria for decreasing the harmful emissions envisage reduction of the sulphur emissions with 33% by 2000 and with 45% by 2010, compared to1980.

There are different methods and technologies worldwide for industrial flue gases cleaning. The choice of the best and most efficient method and technology depend on a long series of criteria, part of which are about the simultaneous purification of sulfur and nitrogen oxides, the presence of a secondary pollution, the type of the by-products, the possibility for partial investments return, the raw materials supply etc.

The electron beam technology is a dry method for simultaneous purification of sulfur and nitrogen oxides in flue gases, without any waste products or waters. The mix of nitrogen sulfate and nitrogen nitrate obtained as a by-product can be used directly as fertilizer or as component for mixed fertilizers used in agriculture. This technology has shown very good results when treating gases with high concentration of SO_2, as flue gases in the thermal power plants at the Maritsa East site are (Fig.1).

2. CONSTRUCTION CONTRACT

In November 1997, during the official visit of the President of the Republic of Bulgaria Mr. Peter Stoyanov in Japan, a tripartite contract was signed between the International Atomic Energy Agency (IAEA) in Vienna - a United Nations agency, the Japanese Atomic Energy Research Institute (JAERI) and the Bulgarian Government represented by NEK EAD, for joint financing of the construction of a Pilot Installation for Flue Gases Treatment at the Maritsa East 2 Thermal Power Plant. The Contract has been extended for 3 years more. The term of this extension expired at the end of 2003. The installation uses the Electron Beam Technology, developed jointly by JAERI and the Japanese company Ebara. IAEA participated in the project financing for part of the main equipment supply and for personnel training at the Pilot Installation. JAERI donated through IAEA to NEK EAD three accelerators and the entire electrical DC supply with them.

According to the above mentioned Contract, the Bulgarian Party - respectively NEK EAD has provided for the financing of:

- the operational design,
- the elaboration of the reactor and the spray cooler, the local production equipment supply,
- the Pilot installation construction, erection and commissioning,
- the elaboration of a research study and report about the possibilities for applying in Bulgaria the electron beam technology for flue gases treatment.

The project is of major importance from environmental point of view and it is monitored by IAEA in Vienna, the Bulgarian authorities, namely the Ministry of Environment and Waters, the Nuclear Regulatory Agency and the Ministry of Energy and Energy Resources. The operational design was fulfilled by Energoproekt EAD. Construction, erection and commissioning works were carried out by the Chimremontstroy company.

The Maritsa East 2 TPP provided for part of the equipment of Bulgarian make and carried out the investments monitoring. It is responsible also for supplying the ammonia needed for the technology process, for the operation and for the maintenance of the installation and for the realization of the by-product, which is a mix of ammonia sulfate (about 97 ÷ 98 %) and ammonia nitrate (2÷3%).

The construction and erection works on the installation were completed in December 2003 and 72-hours tests were carried out in January 2004. The installation was commissioned with a Permit for Usage issued by the State Agency for Construction Control with the Ministry of Regional Development and Infrastructure Nº ST-12-177 on May 14, 2004. The company of the Maritsa East 2 TPP has obtained for the operation of the accelerators a five years License, issued from the Nuclear Regulatory Agency (NRA), concerning the industrial usage of Ionizing Radiation Sources (IRS).

3. TECHNOLOGY DESCRIPTION

Flue gases of a flow rate of 10 000 Nm³/h are aspirated from the gas ducts of boilers 1 and 7. The electron beam method is a dry method for simultaneous removal of sulfur and nitrogen oxides from flue gases.

The process mechanism is the following:

Flue gases are cooled down to a temperature of 65-70º C and after that they pass through a reactor vessel, where they are exposed to radiation by a beam of high-energy electrons from three electron accelerators at a voltage level of 800 keV. This high voltage is generated in the electrons energy and it is absorbed mainly by the molecules of N_2, O_2, H_2O, CO_2 contained in the flue gases. By interaction of these molecules with the ionizing radiation, positive ions are obtained (N_2+ O_2+ H_2O+) and excited molecules as well (N_2* O_2* H_2O* CO_2*), which in a time of several microseconds react with the water steam and oxygen and produce free radicals (OH, H_2O, N, O).

$$e^- \quad N_2; O_2; H_2O \longrightarrow OH; O; H_2O$$

These radicals interact with NOx, SO_2 and SO_3 until nitric acid (HNO_3) and sulfuric acid (H_2SO_4) are obtained as intermediate products.

$$OH;\ O \quad OH;\ O \quad SO_2 \longrightarrow H_2SO_4 \qquad NOx \longrightarrow NHO_3 \qquad H_2O \qquad H_2O$$

Gasiform ammonia and chemically treated water are injected in the reactor vessel close to the stoichiometric amount.

The sulfuric acid and the nitric acid react with the ammonia to produce aerosol ammonium sulfate $(NH_4)_2SO_4$ and ammonium nitrate NH_4NO_3, which are separated from the outlet gas in the electrostatic precipitator.

$$H_2SO_4 + 2NH_3 \longrightarrow (NH_4)_2SO_4$$

$$HNO_3 + NH_3 \longrightarrow NH_4NO_3$$

$$SO_2 + 2NH_3 + H_2O + \tfrac{1}{2}O_2 \longrightarrow (NH_4)2SO_4$$

The reactor is separated from the accelerators by two layers of titanium foil, through which the high-energy electrons pass. The depositions on the reactor bottom are washed with water, which together with the condensate of the outlet protection labyrinth is taken to a draining basin and from there goes to the water treatment system for purification. When high-energy electrons react with the substance, a secondary X radiation is produced. Its intensity is 175 Sv/h. In order to provide the necessary radiation protection, the reactor is installed in a room with reinforced concrete walls 1.3 m thick, and at the inlet and the outlet metal labyrinths are erected for reducing the radiation down to the admissible rates. In consequence of the radiation, ozone is produced in the room, so it is then taken away by an inflow and suction ventilation fans. The secondary product obtained is captured in an electrostatic precipitator and is transferred for packing.

The flue gases fan overcomes the aerodynamic resistance in the installation. The purified flue gases are taken back in the stack. The ammonia necessary for the process is supplied by tank trucks and is unloaded at a constant storage, from where a feeding pump takes it to the evaporator.

FIG. 1. Electron beam plant

4. ANALYSIS OF THE RESULTS FROM THE ELECTRON BEAM TREATMENT INSTALLATION OPERATION

The direct goal of the constructed pilot installation at the Maritsa East 2 Thermal Power Plant for reducing SO_2 and NO_x in the flue gases was to prove the applicability of the electron beam technology at power plants on Bulgarian lignite coal, which quality parameters are given in Table I. The Table shows the high ash content at air-dried base A^d - from 30% to 45% and high moisture W_t^r as well - from 57.2% to 52.5%.

The low calorific value Q_i^r as of 1196 kcal/kg to 1603 kcal/kg and the high concentration of sulfur make the Bulgarian lignite coal unique in their usage as fuel for the thermal power plants at the Maritsa East Site.

TABLE I. QUALITY PARAMETERS OF BULGARIAN LIGNITE COAL

N	Indicator	Design	Unit	I Compo-sition	II Compo-sition	III Compo-sition	IV Compo-sition
				Guaran-teed	max.W_t^r	max. A^d	max. Q_i^r
Maritsa East 2 TPP							
1	Ash, air-dried base	A^d	%	34.0	30.0	45.0	30.0
2	Moisture as received	W_t^r	%	55.5	57.5	52.5	55.0
3	Ash as received	A^r	%	15.2	12.8	21.5	13.5
4	Carbon as received	C^r	%	18.7	19.1	15.7	20.0
5	Hydrogen as received	H^r	%	1.5	1.6	1.3	1.7
6	Oxygen as received	O^r	%	5.0	5.2	4.2	5.5
7	Nitrogen as received	N^r	%	0.3	0.3	0.2	0.4
8	Combustible sulphur as received	S_r^r	%	2.4	2.3	2.6	2.5
9	Volatiles	M^r	%	1.4	1.2	2.0	1.4
10	Calorific value (LHV)	Q_i^r	kcal/kg	1446	1480	1196	1603

5. INCOMING FLUE GASES CONTENTS AT THE MARITSA EAST 2 TPP INSTALLATION

The tests were carried out at the Pilot Electron Beam Installation together with control tests for the trustworthiness of the physical and chemical measurements and their calibration, as well as their impact on the computer information system for operational management of the processes in the installation.

The tests carried out by now give reasons to make the following conclusions and analyses:

- The tested lignite coal have a quality contents and a lower fire heat as shown in Table II, while the gases composition is represented in Table III for the Maritsa East 2 TPP.
- The temperature after the cooling tower is kept in the range of 62÷65 ^oC , so there is no corrosion of the surfaces;
- The measured dew point of the flue gases is 58 ^oC.
- The measured humidity of the incoming flue gases is not 21 %, it is 16÷18 % depending on the fuel burnt and the content of additional air (O_2) in the flue gases. This indicator is necessary for the stoichiometric calculations.

Flue gases from the process of burning coal with the qualities described in Table II are typical with the prevailing content of SO_2 compared to NOx and with the small change in these two components and the relative stability of their average value. One of the very important information from Table I is high concentration of sulfur dioxide SO_2 in the incoming Flue Gases from 16095 up to 16 151 mg/nm^3 . The nitrogen oxide NOx is from 112 up to 390 ppm.

Detailed studies are done with respect to the effect of the following parameters: temperature at the reactor inlet, dose rate, effect of the nozzles water sprinkling, effect of the ammonia needed for the loads as of 5000 up to 10000 nm3/h.

TABLE II. FLUE GASES FROM THE PROCESS OF BURNING COAL

Indicator	Desig-nation	Unit	Design parameters	W_t^r=54% A^d=33.5 Q_i^r=1495 kcal/kg	W_t^r=51.9%A^d=37.7 Q_i^r=1440 kcal/kg
Oxygen	O_2	%	7.5	12.0	12.5
Carbon dioxide	CO_2	% vol.	12.0	6.5	6.2
Water vapours	H_2O	% vol.	22.0	15.7	15.9
Sulfur dioxide	SO_2	Ppm	5600	3287	3092
Sulfur dioxide	SO_2	mg/nm^3, O_2=6%	18 230	16 151	16 095
Sulfur trioxide	SO_3	Ppm	140	-	-
Nitrogen oxides	NO_x	Ppm	390	95	112
Nitrogen oxides	NO_x	mg/nm^3, O_2=6%	-	217	248
Hydrogen chloride	HCl	mg/nm^3	10 – 30	-	-
Hydrogen fluoride	HF	mg/nm^3	21 – 40	-	-
Flying ash		mg/nm^3, O_2=6%	200	35	47
Temperature	$t_{дг}$	^oC	170	155	158
Rarefaction	S	mm H_2O	80	25	40

In our opinion, on the basis of the studies made, the best operating parameters are as follows:

- cooling tower temperature tk 62÷65 oC;
- stoichiometric ammonia quantity α= 0.85÷0.95;
- stoichiometric water quantity β=0.6÷0.9;
- stoichiometric air quantity γ =1.0;
- dose rate about 4 kGy.

The main results of the studies are shown in the Table III below:

TABLE III. MAIN RESULTS OF THE STUDIES

N	Parameter	Unit	Results					
1	Flue gases consumption	Nm^3/h	5 000	6 000	7 000	8 000	9 000	10 000
2	Temperature before the reactor	^oC	68.3	65.9	65.9	61.0	63.9	65.7
3	Stoichiometric ammonia consumption α	-	0.90	0.90	0.75	0.85	0.90	0.85
4	Stoichiometric water consump. B	-	0.20	0.90	0.90	0.60	0.90	0.85
5	Stoichiometric air consump. γ	=	1.00	1.00	1.00	1.00	1.00	1.00
6	Ammonia consumption	Kg/h	16.3	21.41	24.6	26.82	33.4	31.79
7	Dose rate	KGy	9.29	6.93	4.98	3.96	3.95	3.94
8	Efficiency in cleaning of SO_x	%	99.1	99.6	92.9	93.4	98.2	97.8
9	Efficiency in cleaning of NO_x	%	90.7	85.8	80.4	89.1	84.4	86.3

The analysis of the results obtained so far gives grounds for the following conclusions:

1. The constructed installation successfully catches both sulfur and nitrogen oxides.
2. The effect of the dose rate on the cleaning of SO2 in the flue gases is minimal.
3. The dose rate has a dominating role in the cleaning of NOx in the flue gases.(Fig.2.)
4. The influence of the dose rate magnitude on desulphurization is relatively week.
5. The quantity of the ammonia sprinkled in the reactor has the decisive role for SO_2 removal, while it has a much less effect in NOx purification (Fig. 2).

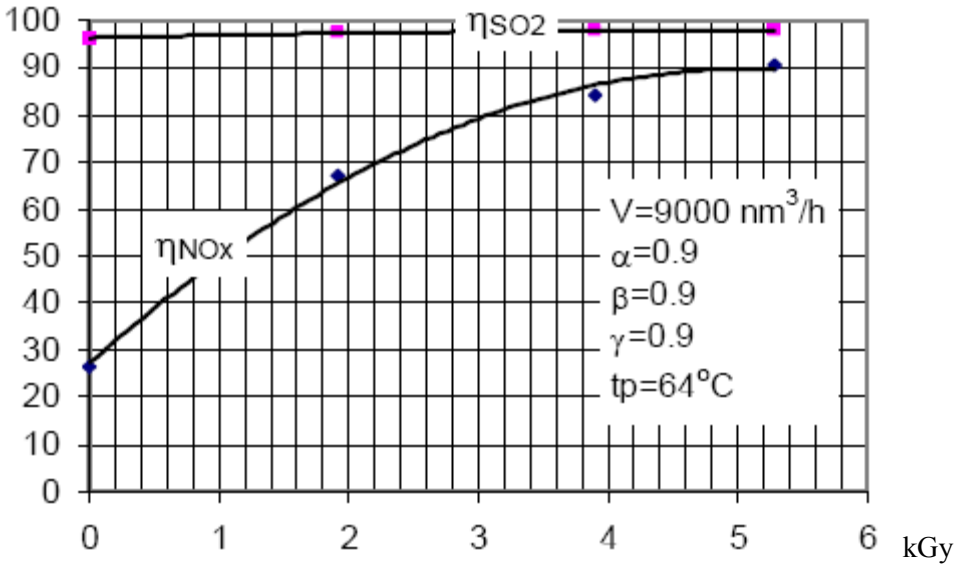

FIG. 2. Efficiency of removing SO_2 and NO_x versus dose rate

The figure 3 illustrates the relation between stoichiometry of ammonia quantity and efficiency of SO_2 and NO_x removal when basic parameters are stabile as below.

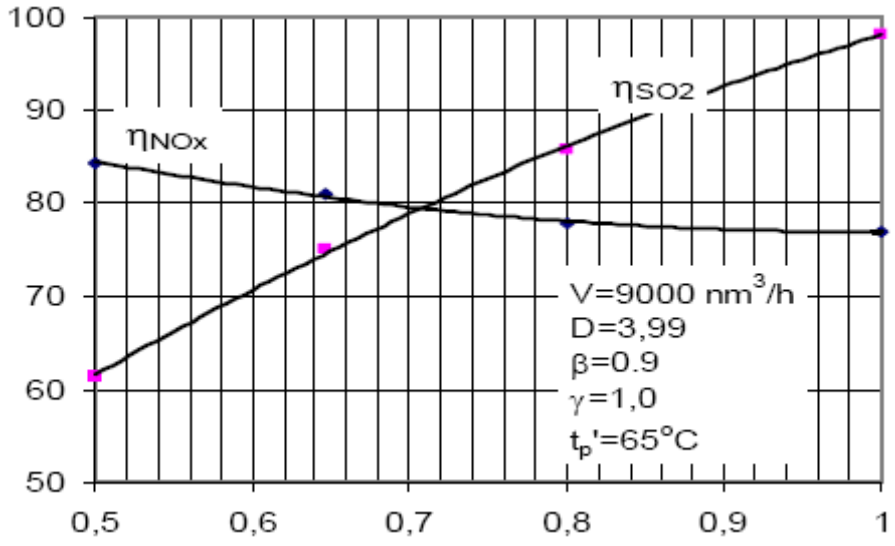

FIG. 3. Efficiency of removal of SO_2 and NO_x versus stoichiometry of ammonia

The flow rate of flue gases under stabilized process parameters contributed of the revealing of this influence. The quantity of the flow rate flue gases has greater influence on removal efficiency of SO_2 and NOx between 5000 up to 7500 nm³/h and lower from 8000 to 10 000 nm³/h.(Fig. 4).

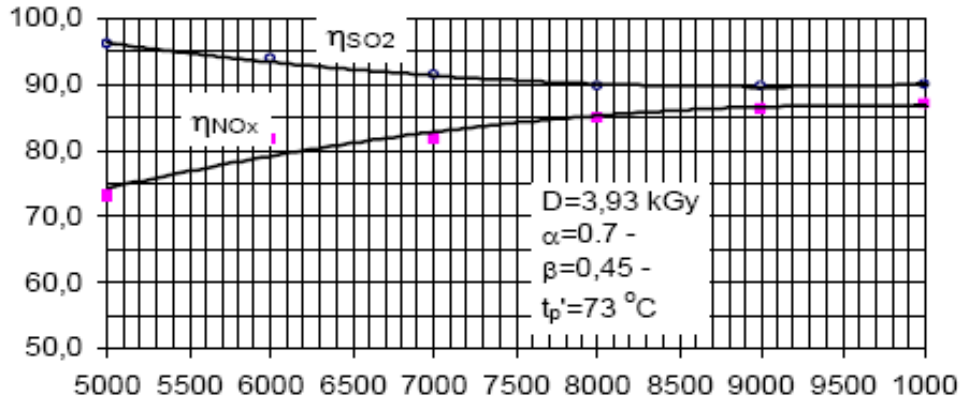

FIG. 4. Removal efficiency of SO_2 and NOx versus the flow rate of flue gases (nm^3/h).

- Very high efficiency can be achieved with respect to desulphurization ($DeSO_2$) - up to 99 % and with respect to de-nitrification (DeNOx) - up to 90%.
- The electricity costs needed for the purification of the gases generated by the boilers at the Maritsa East Site, with prevailing pollution of sulphur oxides, are considerably lower than the expected cost
- The study of the by-product shows that this product can be successfully used both separately and as an appropriate component in the production of a large range of mixed fertilizers.

Figure 5 illustrates the derived relation between of the ammonia quantity in kg/h and the effectiveness of the desulphurization and denitrification, when accelerators are off.

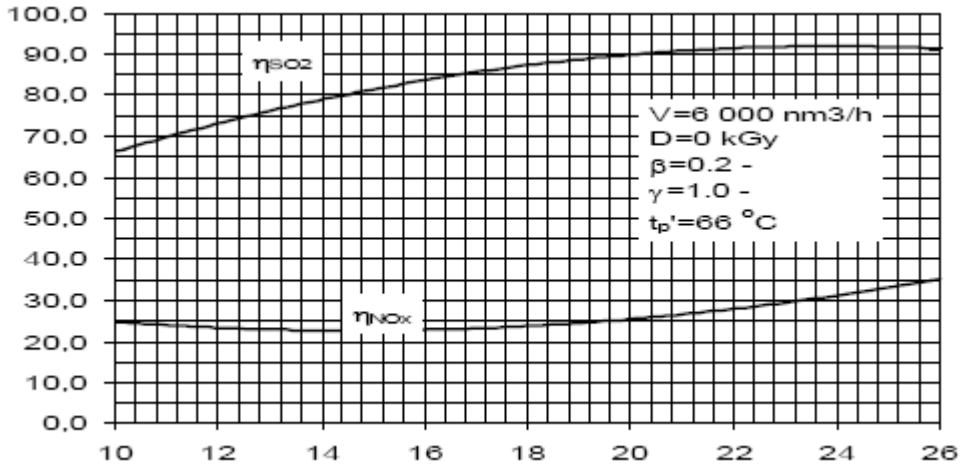

FIG. 5. Removal efficiency versus the quantity of ammonia (kg/h) and stoichiometry, when accelerators are off.

6. GENERAL DESIGN OF THE ELECTRON BEAM INSTALLATION

The main parameter of flue gases – inlet and outlet are:

- The inlet flue gases content SO_2 – 17190 mg/Nm³ and NOx -217 mg/Nm³.
- The removal efficiency of SO_2 is 99.4% and NOx is 82.9%, when the stoichiometry ammonia quantity α is 0.70, stoichiometry water quantity β is 0.8, stoichiometry air quantity γ is 1.0, dose rate is 3.93 kGy.

The efficiency in purifying SO_2 and NO_X depends mainly on the flue gases temperature, on the absorbed dose, the stoichiometric ammonia rate and the humidity. The experimental efficiencies are higher than 95 % for Sox and 70 ÷ 85 % for NOx.

The pilot tests have shown that for higher SO_2 concentration and moisture the efficiency of purification increases. The results of the tests have provided a strong argument in favour of competitiveness of the radiation processing to the FGD plants technology.

The by-product from pilot plant is valuable either as granulose fertilizer or as appropriate component for further production of fertilizers. Commercialization of the byproduct is in process.

7. CONCLUSION

The main goal of the pilot installation was to obtain experience in research and in operation during the process of treating flue gases, and experimental production of a useful by-product with a clear market value. This goal is extended for investigating the possibility for improving the technology as applied to fuel conditions in Bulgaria and in South East Europe.

A technically viable installation has been established; the pilot design has been operating by specialists, who have given proofs of high professional skills.

The research should continue after the completion of the tasks for which the installation was constructed. We consider necessary to carry out studies for other types of fuels; simulation of diverse flue gas compositions can be performed.

After discussion and evaluation of the obtained results a decision for construction of an industrial electron beam installation would be taken; in this case the research and tests performed on the pilot installation provide useful experience for scaling it up.

A study of the process with flue gases of high concentration of NOx, as the case of Bulgarian thermal power plants in Russe with heavy oil, would be also indispensable.

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PROCESS OF FLUE GAS DESULPHURATION WITH ELECTRON BEAM IRRADIATION IN CHINA

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Abstract

This paper gives a brief review of flue gas desulphurisation and de-nitrification with electron beam irradiation (hereafter EA-FGD) process in China. Several installations have been introduced from bench scale to industrial demonstration project, including the state-of-the-art of EA-FGD process, the economy based on local prices and demonstration project, etc. At the end, some proposals of accelerating the industrial application of EA-FGD process are brought forward.

1. INTRODUCTION

Flue gas desulphuration and denitration with electron beam irradiation (EA-FGD) is a kind of reclamation technology for which can simultaneously remove SO_2 and NOx from flue gas without secondary pollutants production. EA-FGD gets its name from its features in electron beam flue gas desulphuration process with ammonia as its reagent. With its basic idea of taking the pollutants SO_2 and NO_X in flue gas as a kind of natural resources and transforming them into a kind of fertilizer nourishing elements of nitrogen and sulphur, purification of coal-fired flue gas and reconnection of the circulation chain of sulfur element from natural coal in a "green" way, which is broken by coal combustion could be achieved. The advantages of EA-FGD were recognized gradually home and abroad, thus rapid progress was made in past decade.

History of the process can be traced back to 1970 in Japan. So far, there are 5 industrial scale demonstration installations completed or under construction all over the world, including 3 in China, one in Japan and another in Poland. Among those three installations in China, the process of the two of them comes from Japanese Ebara, another one from Chinese Entech (Sichuan Entech Environment Technology Co. Ltd.). Entech is an engineering construction company of environmental pollution control that controlled by the Institute of Environmental Protection Engineering (IEPE), which is a division of China Academy of Engineering Physics.

The research on EA-FGD process in China started from the middle of the1980s. Shanghai Institute of Nuclear Research (SINR), a division of Chinese Academy of Science (CAS), firstly started bench scale research. IEPE started to develop EA-FGD process in the middle of 1990s and completed a pilot plant in 1999. At present, Entech is constructing an industrial scale demonstration installation at a power plant in Beijing suburb. At the end of 2000 another pilot plant was completed by the Institute of Nuclear and New Energy Technology (INET), which is a division of Tsinghua University. Chinese also devote themselves to high power electron beam accelerators while developing process, which is a kind of key equipment for the industrial application of EA-FGD process. Now in China, IEPE, SINR and Institute of Modern Physics (IMP), which is located in Lanzhou as a division of CAS, are developing such kind electron beam accelerators.

2. BENCH SCALE RESEARCH IN SINR

During the period of 1987 to1990, SINR started bench scale research with a based on a bench scale installation constructed, of which the maximum simulated flue gas flow was 25 Nm^3/h. Main influential elements for removal of SO_2 and NO_x such as electron beam dose, flue gas temperature, as well as ammonia stoichiometry were investigated.

It finds that the removal efficiency of SO_2 and NO_x obtained are respectively over 92% and 77.5% at flue gas temperature of 70℃ and electron beam dose of 17 kGy, the powder by-product mainly consists of $(NH_4)_2SO_4$ and $(NH_4)_2SO_4 \cdot 2NH_4NO_3$ [1].

3. EA-FGD PROCESS IN IEPE AND ENTECH OF CAEP

3.1. Mianyang pilot plant

IEPE started EA-FGD process research in 1995. After a series of laboratory experiments and theory analysis, a pilot plant was built at Science Town Thermal Power Station in Mianyang in the end of 1999. Its main technical parameters and flow diagram are illustrated in Table I and figure 1 respectively. The process flow is similar to the conventional one except for such features as: 1) the inlet SO_2 and NO_x in flue gas can be simulated by injecting pure gaseous SO_2 and NO; 2) flue gas to be treated can be extracted from the outlet and (or) inlet of the water membrane particulate collector of the power plant, through which particulate concentration of the flue gas can be regulated to meet the demands of investigation; 3) the reaction vessel is shaped in cylindrical with 3 meters in diameter and horizontal lay-down. To avoiding of insufficient turbulence, the flue gas flows into the reaction vessel are induced and flow along the tangent of the vessel inner wall; 4) the electron beam accelerator was manufactured by NIIFA Russia, resonance transformer type, 35mA×800kV.

TABLE I. MAIN PARAMETERS OF MIANYANG PILOT PLANT

Parameters	Design Values
Flue gas flow	3000~12,000Nm^3/h
Inlet SO_2 concentration	400～3000ppmv
Removal efficiency of SO_2	≥90%
Inlet NO_x concentration	200~800ppmv
Removal efficiency of NO_x	≥50%
NH_3 exhaust concentration	≤50ppmv
Electron beam energy	800~1000keV

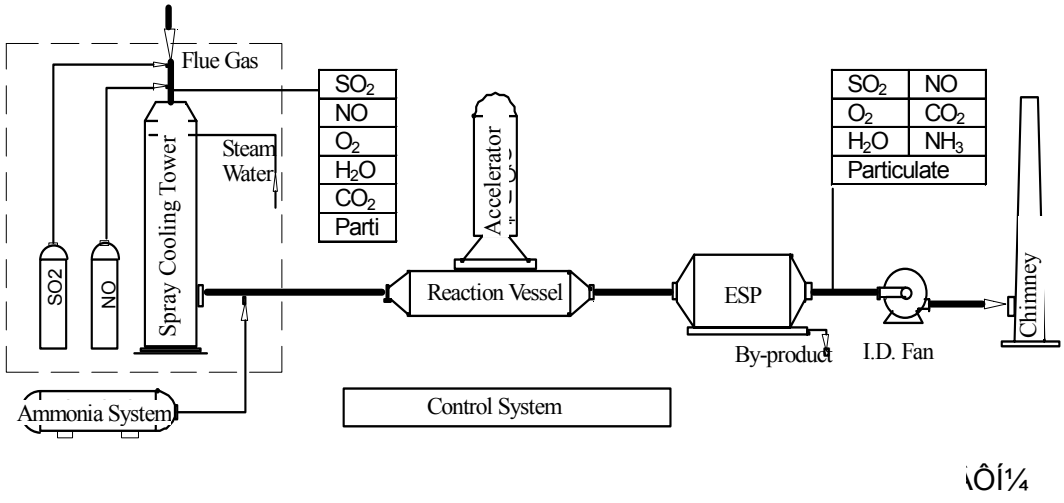

FIG. 1. Flow diagram of Mianyang pilot plant

Based on the pilot plant, the process investigation and equipment research has been performing. The removal efficiency of SO_2 and NO_x can respectively reach up to 97% and 75% at electron beam dose of 3kGy~4kGy, inlet SO_2 of about 2000ppmv, less than 50ppm of outlet ammonia concentration can also be achieved and by-product of ammonium sulphate and ammonium nitrate obtained can meet the requirements of Chinese national standards.

3.2. Beijing Jingfeng industrial demonstration plant

The industrial demonstration plant that is under construction is located at Jingfeng Thermal Power Plant at southwest Beijing and will be completed in the middle of 2005. It can treat flue gas of 630 000 Nm^3/h, corresponding to 50 MW and 100 MW coal-fired units. The main technical parameters and the flow diagram are illustrated in Table II and Fig. 2 respectively.

TABLE II. MAIN TECHNICAL PARAMETERS OF BEIJING JINGFENG INDUSTRIAL DEMONSTRATION PLANT

Parameters	Design Values
Flue gas flow	630,000Nm^3/h
Inlet flue gas temperature	146^{0}C
Inlet SO_2 concentration	4200mg/Nm^3
SO_2 removal efficiency	90%
Inlet NOx concentration	1200 mg/Nm^3
NOx removal efficiency	20%
Outlet particulate concentration	≤200mg/Nm^3
Outlet ammonia concentration	< 40 mg/Nm^3
Electron accelerators	1000kV/500mA×2 1000kV/300mA
Total power consumption	≤2850 kW

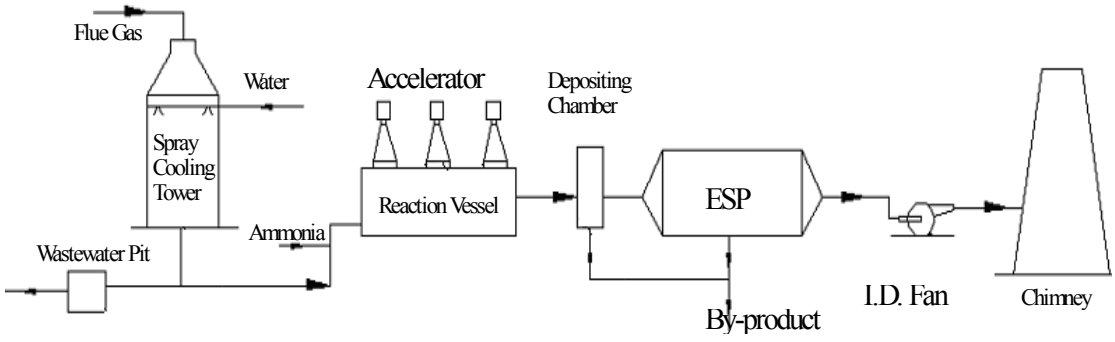

FIG. 2. Flow diagram of Jingfeng industrial demonstration plant

In the demonstration plant, the cooling tower is a hollow cylinder, in which spraying water cools flue gas down. A little wastewater that comes from the cooling tower is collected by a pit designed beneath the end of the cooling tower, and then discharged to neutralize power plant's acid wastewater.

Three accelerators are installed in series on the up surface of the reaction vessel, which is a horizontal rectangle box. The connection set of connecting accelerator and reaction vessel is elaborately designed, in which cooling air firstly cools down the titanium foil of electron beam extracting window on accelerator's scanning box, then the outer and inner surface of titanium foil of reaction vessel inlet window successively. The byproduct is collected mainly by a special developed ESP. A depositing chamber is set between ESP and reaction vessel, which is used for collecting part of by-product and conveniently connecting reaction vessel and ESP.

Economic benefit of EA-FGD is always concerned by customers and will severely influence the competitive nature to others processes. Based on local Beijing price and Jingfeng demonstration plant, the economy of EA-FGD process is evaluated, as illustrated in Table III. The evaluation is based on three modes of sulphur content in coal and the price of by-product is estimated as 60 $US/ton. Comparing to conventional process in local China, such as WFGD plant of 300 MW, the operation cost of EA-FGD process is about half of WFGD, the capital cost is about twice of WFGD, which fluctuates with the sulphur content in coal.

TABLE III. ECONOMIC EVALUTATION AND COMPARISION OF BEIJING JINGFENG INDUSTRIAL DEMONSTRATION PLANT

Cost	EA-FGD Process (Jingfeng plant, 150 MW)			WFGD Process (300MW)
	S, 2%	S, 1.5%	S, 1.1%	
Operation cost ($/yr.)	518,600	569,900	591,300	----------
$DeSO_2$ cost ($/ton)	38.11	55.84	78.99	100~120
Capital cost ($/kw)	74.5			40

During past ten years, IEPE has been devoting to develop EA-FGD process and equipment technology. Except for above two plants, the main work focuses also on 1) process of flue gas desulphuration and denitration; 2) some key equipments and engineering techniques, such as high power electron beam accelerator, by-product collecting technology, electron beam transportation apparatus, etc.; 3) other application researches of EA-FGD in metallurgy industry, VOC purification, etc. Now, above research and development work are been carrying through.

4. EA-FGD PROCESS IN TSINGHUA UNIVERSITY

In December 2000, INET of Tsinghua University constructed a pilot plant with a maximum flue gas flow of 10 000 Nm^3/h [2]. The main technological parameters are listed in Table IV and the process flow diagram of the plant is illustrated in Figure 3.

Wet electrostatic precipitator is applied to collect by-product, in which ammonium sulphate and ammonium nitrate are dissolved in water. The water solution contented ammonium sulphate and ammonium nitrate is then injected into spraying dryer to lower temperature and increase humidity of flue gas, meanwhile to crystallize ammonium sulphate and ammonium nitrate. Notice that the ideal removal efficiency of SO_2 and NO_x can be achieved at lower electron beam radiation dose (～1kGy), thus energy consumption of the EA-FGD installation is reduced greatly and system reliability gets improved.

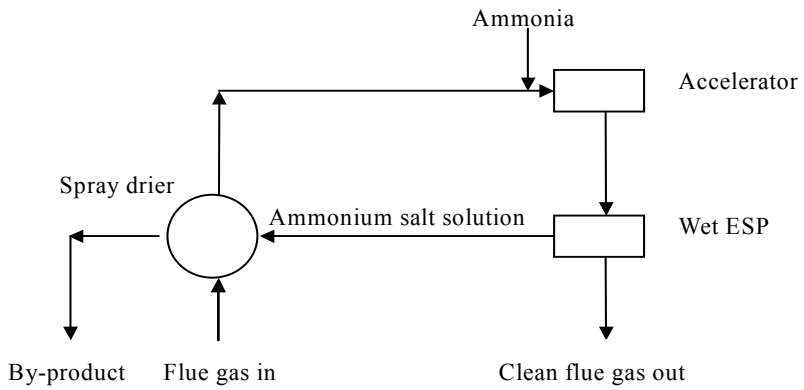

FIG. 3. Flow diagram of Tsinghua pilot plant

TABLE IV. MAIN TECHNOLOGICAL PARAMATERS OF TSINGHUA PILOT PLANT

Items	Values
Flue gas flow	10,000Nm^3/h
Inlet Flue Gas Temperature	140□
Inlet SO_2 Concentration	1.5×10^{-3}～1.6×10^{-3}
SO_2 Removal Efficiency	92%～94%
Inlet NO_x concentration	4.0×10^{-4}～4.5×10^{-4}
NO_x Removal Efficiency	30%～33%
Outlet Ammonia Concentration	$<1\times10^{-6}$
Electron Beam Dose	1kGy

In January 2003, another pilot plant was set up at Beijing Kaituo Thermal Power Center. It was set up to purify a 35 t/h boiler's flue gas of 43000 Nm^3/h. It should be mentioned that the flue gas coming for the boiler is absorbed by NH_4SO_3 solution in an absorber, and then exhausted to a stack directly. The NH_4SO_3 solution in the absorber is fed to a chamber for irradiated by electron beam. After irradiation, the solution contented NH_4SO_4 coming form irradiation chamber is vaporized, dry and crystal NH_4SO_4 is obtained.

5. EBARA'S PROCESS IN CHINA

There are two industrial scale plants which adopting Ebara's process in China. One (Chengdu EBA installation) is located at Huaneng Chengdu Thermal Power Plant, another (Hangzhou EBA installation) at Hangzhou Xielian Thermal Power Plant. The main technical parameters of above two installations are illustrated in Table V.

Chengdu EBA installation is the first industrial scale EA-FGD installation in the world [3]. The construction began in March of 1996, completed in July of1997, and successfully passed acceptance

test organized by China State Power Company in May 1998. During actual operation, SO_2 removal efficiency reaches and exceeds 80%, NOx removal reaches 18%. The main problems which had ever met in operation are on the ESP used for collecting by-product and corrosion of metal materials. The accelerator had also met problem and been transported back to Japan for repair. Two sets of electron accelerators applied in this installation made by Nishing High Voltage Company, which is a Japan Company. The total capital cost is about $US 11.4 million, which is about corresponding to 126.5 $US/kW. The operation cost is about 120$US/ton SO_2.

Xielian EBA installation is the second industrial scale EA-FGD installation in China, which is the third same kind installation in the world, and passed 168 hours acceptance test in the end of 2002. The actual SO_2 removal efficiency reaches 90%. During operation, it does not meet obvious problems up to now, especially as which Chengdu EBA installation met.

It should be mentioned that 1) accelerator applied made by Ebara itself in which the transformer is insulated with SF_6 other than oil applied in Chengdu EBA installation; 2) the cooling tower is replaced by a water spraying cabin other than Chengdu EBA installation' spray tower, and cooling water is used in circulation; 3) a mist eliminator is set at the outlet of the water washing cabin.

TABLE V. MAIN PARAMATERS OF TWO EBARA INSTALLATIONS IN CHINA

Items	Design Values	
	Chengdu Installation	Hangzhou Installation
Flue gas flow	300,000Nm^3/h	305,400Nm^3/h
Inlet flue gas temperature	132^oC	150^oC
Inlet SO_2 concentration	1800ppm	2767.6mg/Nm^3
SO_2 removal efficiency	80%	85%
Inlet NO_x concentration	400ppm	200ppm
NO_x removal efficiency	10%	55%
Outlet particulate concentration	≤200mg/Nm^3	≤200mg/Nm^3
Electron accelerator	800kV/400mA×2	800kV/400mA×2
Total power consumption	≤1900kW	≤1896Kw

6. CONCLUSIONS

Since two years ago, Chinese coal fired power plant construction began to develop rapidly and will reach a boom from now on and will last 10 years or so by forecasting. It is estimated that new constructed power plant will reach 0.5 billion kilowatts by the year of 2020. Except for new power plants, the constructed power plants have been more than 0.4 billion kilowatts in May 2004, of which only several percent has constructed and under construction FGD installations. So huge FGD market will not be met by WFGD and needs new processes, not only for Chinese huge natural gypsum reserves, but also for Chinese extensive territory and different local situation where power plants located. All above gives EA-FGD an opportunity to develop.

EA-FGD is concerned intensively by Chinese government and customers, but there are few customers that would like to try this process of their own accord. The main reasons are that 1) the capital cost is higher, about twice of WFGD at present; 2) the process is not matured enough, especially the stability which can not meet the requirement of power plant customers, normally customers require a FGD installation operating at the rate of 95% of a power plant operation hours; 3) power consumption occupied nearly 2% of power plant capacity, which is higher than WFGD of about 1.5% or less. Based on above situation, we are going to push EA-FGD going forward as follows:

(1) Decrease the power consumption

By developing process, optimizing engineering design and equipment selection, we believe that the power consumption is possibly decreased 0.2% or more.

(2) Lower the capital cost

The accelerator applied in the process is almost the only main equipment needing to import and its capital cost occupied about 30% of present total capital cost at local China. According to our estimation, the accelerator capital cost can bee decreased to 15% of total capital cost when it is manufactured in local China. With the process applied in larger capacity power plant, such as 300MW or more, the capital cost will be decreased more.

Our target is to decrease the capital cost to 55$/kW to 60$/kW. Considering the denitration, the EA-FGD process will be competitive compared with present WFGD and SCR process of denitration.

(3) Optimize the engineering technology and equipment to improve installation's stability

The main works are focus on ESP, anti-corrosion and adhesion of by-product on reaction vessel, duct and electrodes of ESP, etc.

(4) Enhance customer's confidence

At present, by constructing more and more industrial scale demonstration plants with good quality to enhance customer's confidence and win more development opportunity for EA-FGD. This needs extensive help and support of international organization and local government. A 300MW scale industrial demonstration installation is been planning, which will be supported by China government.

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ANALYTICAL METHODS AND MONITORING SYSTEM FOR INDUSTRIAL PLANT FOR ELECTRON BEAM SIMULTANEOUS SO_2 AND NO_X REMOVAL FROM FLUE GASES.

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Abstract

The reliable and precise measurements of gas parameters in different points of industrial plant are necessary for its proper operation and control. Natural flue gases there are only at the inlet. At other points of plant gas parameters are strongly modified by process control system. The principal role of process monitoring system is to provide the Computer System for Monitoring and Control and the operator with instantaneous values of alarm states, media consumption and continuous recording and controlling of process parameters. The structure of the process control system is based on algorithms describing functional dependence of SO_2 and NO_x removal efficiencies. The best available techniques should be used for measurements of flue gases parameters at critical points of installation.

1. INTRODUCTION

The electron beam flue gas treatment technology was developed in 1972 in Japan [1]. Research on the process has been carried out in laboratory and pilot plants in Japan [2], USA [3], Germany [4] and Poland [5]. It should be noted that IAEA has also played an important role in the development of this technology. The experiences gained during these studies have allowed the preparation of full-scale industrial plant designs. The first such installation was built at EPS Chengdu in China at 1997 by Ebara Co. and State Power Cooperation. The plant has a capacity of 300 000 Nm^3/h and is equipped with two accelerators of 800 kVx400 mA. SO_2 removal rate is more than 80 % and is obtained by irradiation with dose 3.2 kGy of flue gases contain SO_2 of 500 to 2400 ppm. In Poland, the industrial plant was built at EPS Pomorzany (Dolna Odra Power Cooperation) in Szczecin. Its maximal treatment capacity is 270 000 Nm^3/h flue gases emitted from two coal-fired Benson boilers. Parameters of the electron beam process are chosen so as to guarantee the efficiency of NO_x removal up to 80% and SO_2 removal above 70% in a continuous operation of the installation.

Monitoring and control system for industrial plant as well as applicable analytical methods are the goal of the paper. This paper is based on the designs of these systems prepared for Polish industrial plant. The designs were presented during the special Meeting IAEA in Vienna and then implemented in the constructed plant. During the three-year operation of the plant these systems were verified. The paper presents final version of the requirements for these systems.

2. MONITORING SYSTEM

The monitoring and control system is extremely important for achieving a successful operation of the plant. The principal role of process monitoring system is to provide the Computer System for Monitoring and Control and the operator with instantaneous values of alarm states, media consumption and continuous controlling and recording of process parameters. The industrial plant is designed for continuous operation with small number of staff.

The general tasks of the monitoring system are:

- continuous, reliable and precise measurements of flue gases and electron beams parameters having direct influence on the efficiency of SO2 and NOx removal as well as on the operation of the whole industrial plant,
- acquisition and visualization of process parameters. Visualization of flow charts on high-resolution monitor including visualization of regular updated measurement data,
- supervision and control of process parameters under normal, transient, alarm and breakdown condition,
- storage, visualization and evaluation of process parameters. Preparation of reports (obligatory or on request),
- on-line adjustment of control parameters.

The presentation of all data should take place in the control room of the plant on high-resolution monitors and on video screen. Furthermore presentation takes place in the operator station for boilers and in the ecological department of EPS. Fig. 1 presents the scheme of the process monitoring system of the industrial plant.

The electron beam flue gas treatment (EBFGT) process is realized in the following technological steps:

- inlet where initial parameters of flue gases should be determined,
- spray cooler where humidification and cooling the gases is performed,
- ammonia dosage to flue gases,
- irradiation of flue gases in the process vessel by high-energy electron beams from two accelerator,
- precipitation of by-product using electrostatic precipitator,
- outlet where final parameters of purified flue gases should be determined.

In each of the above steps the monitoring and control system realizes specific tasks. The actual values of process parameters, determined by monitoring system, are employed for the technological modification realized by control system.

In the inlet the following gas parameters should be measured: flow rate, temperature, pressure, humidity, particulate and concentration of SO_2, NO/NOx and O_2. Gas composition strongly depends on the combustion conditions in boiler and quality of used fuels. It is recommended that the fly ash content in the flue gases at inlet to plant should be lower than 50 mg/Nm^3. High particulate concentration will complicate plant operation as well as decrease the quality of obtained by-product. Normally the SO_2 and NOx concentrations are high and exceed the allowed emission level.

Electron beam flue gas treatment process is applied for simultaneous removal of SO_2 and NOx from flue gases. The gas humidity and temperature should be modified in spray cooler to achieve optimal conditions for their irradiation in the process vessel. In the spray cooler atomized water is injected into flue gases by means of dual fluid air-water nozzles. The parameters of water and compressed air supplied nozzles and obtained gas temperature and humidity at outlet of spray cooler should be recorded. If the obtained gas humidity is lower than the required optimal value then steam should be additionally injected to spray cooler and its parameters should be recorded. Near stoichiometric amount ammonia is injected to the humidified flue gases before their inlet to process vessel. Such gas mixture is irradiated in the process vessel.

Two-stage irradiation enhances NOx removal from flue gases. The flow rate of flue gases at the inlet to process vessel and applied electron beams current are necessary for calculation of irradiation dose. The flue gas leaving the process vessel contains a mixture of ammonium sulfate and ammonium nitrate. The particles are generally small and hygroscopic. It was found in the Indianapolis and Nagoya Pilot Plants that an electrostatic precipitator (ESP) is the most effective collector of this by-product.

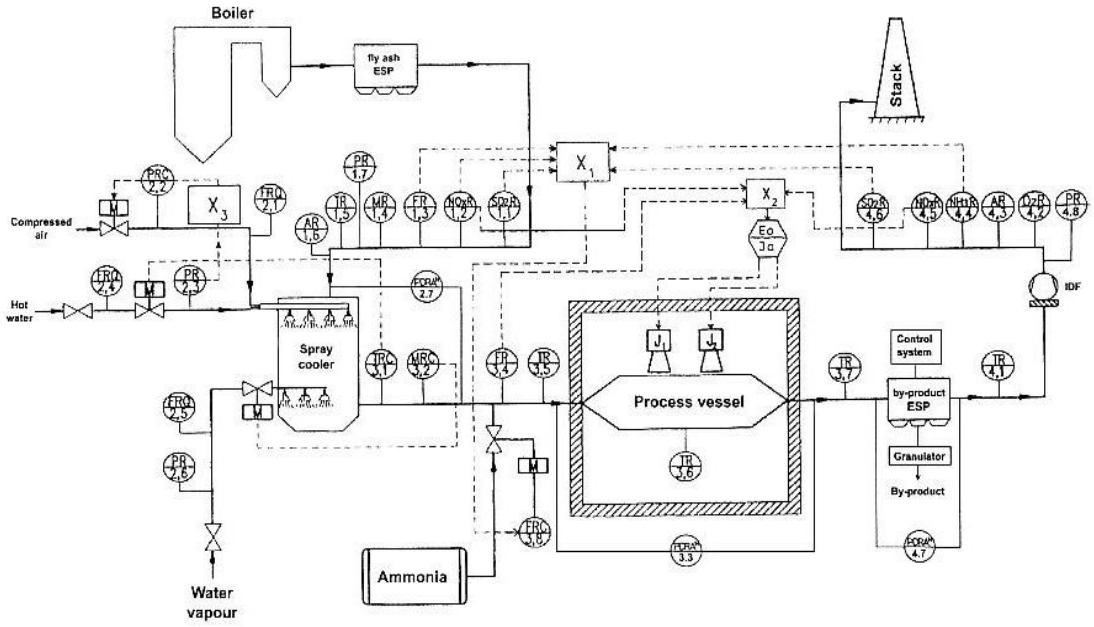

FIG. 1. Scheme of the EBFGT process control system

The ESP is equipped with its own control system tied in to the main system and an energy saving operating system. After the by-product is collected it will go to a granulator for processing and ultimately to storage and shipment to users. The final parameters of purified flue gases are determined at the plant outlet. The concentrations of SO_2, NO/NOx, O_2, NH_3 and fly ash content are important process parameters. These values are used for determination of the obtained SO_2 and NOx removal efficiencies and for control plant operation. Additionally these values are used for preparation of the obligatory ecological report delivered by modem to provincial environmental agency.

3. CONTROL SYSTEM

All individual sub-systems of the plant are tied together and are controlled as a unit to ensure successful operation of the plant and to meet environmental standards and regulations. The structure of the process control system is based on algorithms describing functional dependence of SO2 and NOx removal efficiencies.

3.1. Control of spray cooler operation

The flue gases leaving the coal-fired boiler and its ESP have high temperature (usually above 120^0C) and low humidity (4% to 6% (V) by volume). Fig. 2 presents the dependence of SO_2 and NO_x removal efficiencies on gas humidity.

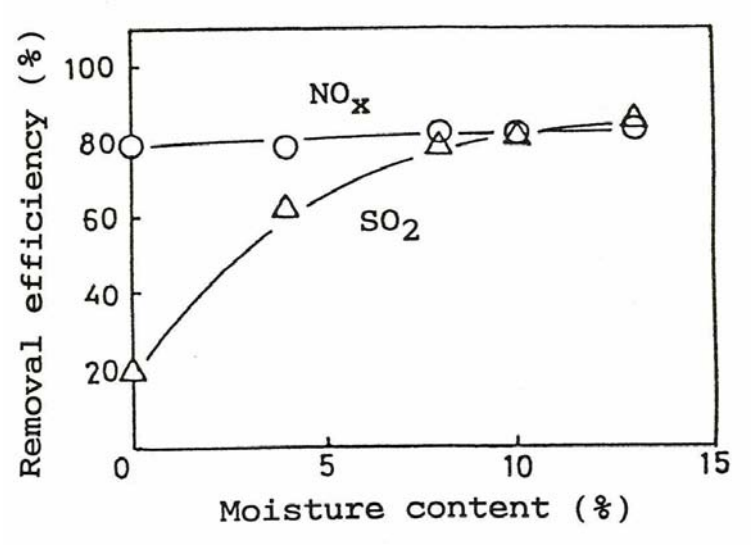

FIG. 2. Effect of moisture on NO_x and SO_2 removal efficiencies [6].

The optimal removal efficiencies of both pollutants are obtained for gas humidity greater than 11% (V). More complicated are dependencies of SO2 and NOx removal on gas temperature (Fig.3 [6]). SO_2 removal efficiency strongly increases with lowering gas temperature. This contrasts with NOx removal which increases with the increase of gas temperature. Then the gas temperature at the outlet of spray cooler should be chosen depending on requested SO_2 and NOx removal efficiency from flue gases. Humidification and cooling of the flue gases is performed in a spray cooler. Atomized water is injected into flue gases stream by means of dual fluid air-water nozzles. The spray cooler is operated with a dry bottom, i.e. all of the water injected into the flue gases is evaporated. The amount of sprayed water (FRQ-2.4) is controlled by the gas temperature at the outlet of spay cooler (TRC-3.1). The pressure of compressed air (PRC-2.2) should be greater than the pressure of hot water (PR-2.3) delivered to nozzles. The nozzle producer determines the relationship between these pressures. This relationship should take into account the controller X4 that controls the pressure of air. The amount of sprayed water is sufficient for the reduction of flue gases temperature to requested value but may not be enough to increase the humidity to 11% vol.

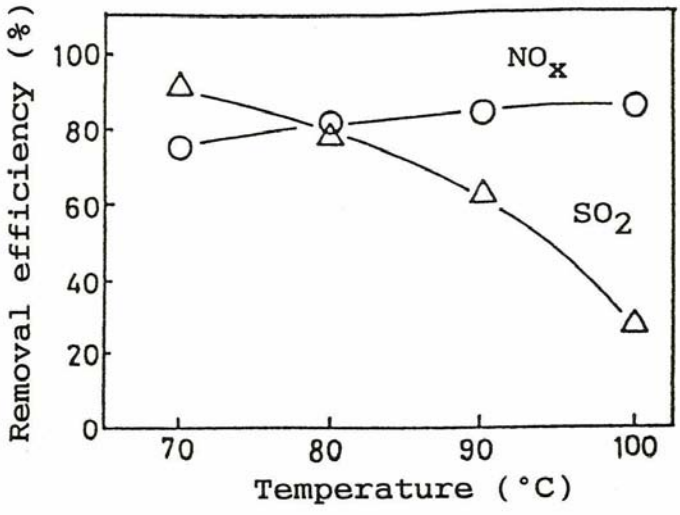

FIG. 3. Effect of gas temperature on NO_x and SO_2 removal efficiencies [6].

Therefore in the bottom part of the spray cooler, the steam is added. The amount of added steam is controlled by humidity control system (MRC-3.2). The measurement of the pressure drop between the inlet and the outlet of spay cooler is used for control operation of the whole spray cooler. At the bottom of the cooler may collect some water (in case of non proper dispersion of water by nozzles) and fly ash carried by flue gases. These deposits may increase the measured pressure drop. If this value is higher than permissible one, then alarm signal arrives in Computer Control System.

3.2. Control of ammonia dosage to flue gases.

Figure 4 presents dependence of SO_2 and NO_x removal efficiencies on ammonia stoichiometry.

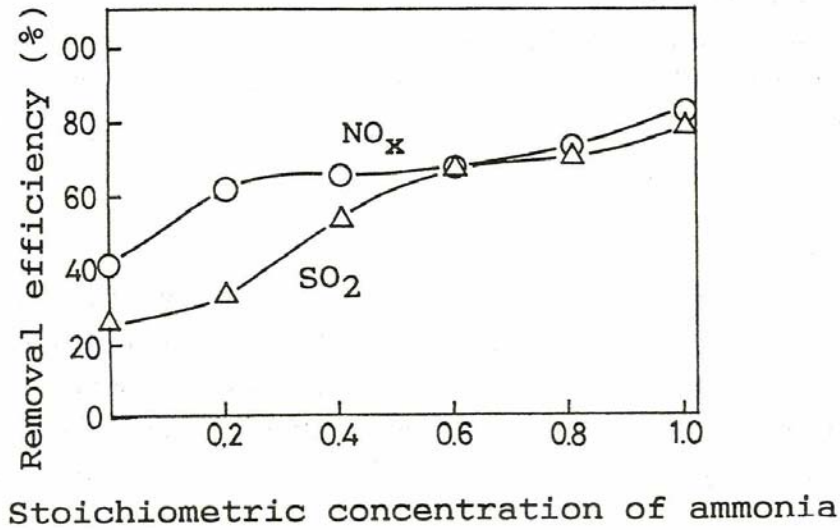

FIG. 4. Effect of ammonia on removal of NO_x and SO_2 [6].

The optimal removal efficiency of both pollutant are obtained for ammonia stoichiometry greater than 0.9. In general two types of process control are employed for control of ammonia dosage system: feed forward and feed back. In feed forward a quantity of injected ammonia is based on the inlet SO_2 and NOx concentration, the flue gases flow and the required NH_3 stoichiometry. Due to the fact that the SO_2 and NOx removal efficiency is never 100% (due to imperfect mixing of the NH3 with the NOx and SO_2 for example), a fraction of the NH_3 remains unreacted and exits in the plant outlet (the so-called ammonia slip). In practice it is desirable to keep the ammonia slip as low as possible due to the environmentally harmful effect of ammonia. In feed back system the ammonia dosage is controlled on the basis of SO_2, NOx and NH_3 concentrations measured on-line at the outlet of installation. The ammonia dosage to the flue gases is controlled by controller X1 employed combined system: a feed forward control ensures a fast system response, while the $NH_3/(NOx+SO_2)$ ratio is automatically fine-tuned by means of the measured outlet NH_3, NOx and SO_2 concentrations (feed back).

3.3. Control of electron beam flue gases irradiation in the process vessel.

Flue gases are irradiated in process vessel by high-energy electron beam from two accelerators. It is necessary to apply a high dose to obtain a high NOx removal. NOx removal is a radiation-induced process. The SO_2 removal is based on two different pathways: thermochemical oxidation and a radiation-induced process. At zero doses, the SO_2 removal efficiency is caused by a thermal reaction of SO_2 and NH_3 in the presence of moisture. These reactions take place in the gas phase as well as in the surface such as those on the filter cake of the baghouse and the collector plates of the ESP. The optimal SO_2 removal efficiencies are obtained for doses smaller than 8 kGy.

The improvement in NOx removal is achieved by multi-stage irradiation and by adequate dose distribution between irradiation stages [7]. In this paper two-stage irradiation was assumed in which optimal dose distribution is following: first-stage - 56% and second-stage – 44% of total dose. Control of the electron beam irradiation process is based on dose dependency of NOx removal efficiency. NOx removal efficiency is strongly effected by inlet NOx concentration. Higher NOx removal is achieved with higher absorbed dose and with lower inlet NOx concentration. Controller X2 is applied for control of irradiation process. The total accelerator electric beam current is determined by the: inlet NOx concentration, required NOx removal efficiency and flow rate of flue gases at inlet to process vessel. This is feed forward process control. In feed back control, the actual outlet NOx concentration is compared with requested NOx concentration and adjusted.

The electron beams with the moderate electron energy and high beam power should be applied for irradiation of flue gases. In the Chengdu Industrial Plant, two accelerators of 800 kV x 400mA with one power supply from Nisshin High Voltage Co. (Japan) are installed. Such system is delivered with own computerized control system.

4. ANALYTICAL METHODS.

For monitoring and controlling of the electron beam (e-b) process, the reliable and accurate measurements of flue gases composition at the critical points of e-b installation are indispensable. In the selection of suitable measuring equipment it is necessary to consider the specifics of treatment process. The parameters of flue gases emitted from coal-fired boiler and dedusted by ESP are monitored at the plant inlet. In the spray cooler the humidity of flue gases increases to level 10-12% (V) and gas temperature is lowered. The elevated humidity complicates the gas analysis both at the outlet of humidifier and the subsequent ones. Ammonia is injected to flue gases before their inlet to process vessel. At the process vessel an essential changes of flue gases composition occurs as a result of thermochemical and radiation-induced processes. The gases leaving the irradiation vessel are a multicomponent three–phase system.

The gas phase is characterized by significantly reduced SO_2 and NO concentrations, a slightly increased NO_2, the presence of unreacted NH_3, a small concentration of nitrous oxide N_2O (a gas treatment by–product) and nearly unchanged CO_2, CO, O_2, N_2 and water vapour content. The liquid phase consists of sulphuric and nitric acid aerosols. The solid phase is formed of by-product particulates of ammonium sulphate and nitrate. At the ESP about 99% of by-product and significant amount of liquid phase are removed. Parameters of purified flue gases are monitored at plant outlet.

Measurements of gas composition at this point are extremely difficult for the following reasons: low gas temperature and its high humidity, the presence of unreacted NH_3 and content of fine particulates of by-product which are hygroscopic and are of submicron size [8]. For minimalization of these negative influences, the extractive gas analysis system should be equipped with the following special components:

- sample probe should be equipped with heated gas filter with proper pore size. Gas filter should be kept at the temperature above acid dew point of flue gases and it should be regularly cleaned (blowback of dry and hot inert gas).
- hot ammonia scrubber for removing gaseous ammonia from purified flue gases. It should be installed at the outlet of gas filter.
- sample transporting line should be kept at temperature above acid dew point of flue gases.
- in the sample conditioning unit, water vapour should be carefully removed from sample gas and the condensate should be automatically discharged by pumping it off. In the conditioning unit the acid filter should be used.

All measuring devices installed at industrial plant should satisfy the following requirements:

- each component of monitoring system should be selected in order to ensure high precision, selectivity and long-term stability,
- each set-up should adapted for uninterrupted and unattended operation,
- the availability of each measuring set-up higher than 90%,
- analog output – current signal 4÷20 mA,
- simple method of calibration,
- easy access to all serviceable parts,
- manufacturers of installed gas analyzers systems should have at least one of certificates such as ISO, EPA, TUV, MCERTS for offered instrumentations.
- the monitoring system should comply with national environmental standards, especially the measurements of gas composition at the plant outlet which are used by national agencies for analysis of ecological noxiousness of leaving flue gases.

The best available techniques should be used for measurement of flue gas parameters in the industrial plant. From the experiences gained during Kawęczyn pilot plant and Pomorzany industrial plant operation the following measuring devices are recommended for application (Table I).

Periodic verification of continuous measurement of the main pollutants should be performed using Standard Reference Methods (SRM) specific for each pollutant [9]. The SRM are also used for calibration of the in-situ gas analyzers. The monitoring and process control systems, together with own accelerator control system are operated from the computers localized in the control room (Fig.5).

TABLE I. ANALYTICAL METHOD APPLIED FOR APPLICATION IN INDUSTRIAL PLANT.

Flue gases parameter	Measuring method	Measuring device	Special requirements
Flow rate	in-situ	Annubar (multiple-point Pitot tube) or ultrasonic flow meter	correction for flue gases temperature, pressure and humidity, regular blowback of the probe
Dust concentration	in-situ	Double-pass transmissometer for high dust concentration and back scatter device for low-level concentration	
SO_2 concentration	extractive	Pulsed U.V. fluorescence analyzer or NDUV analyzer or NDIR analyzer	
NO/NO_x concentration	extractive	Chemiluminescence $NO-NO_2-NO_x$ analyzer or NDIR analyzer with NO_2-NO converter	
H_2O concentration	in-situ	Diode laser spectrometer in the IR spectrum	application of heated device to avoid water condensation on lenses
NH_3 concentration	in-situ	Diode laser spectrometer in the IR spectrum	application of heated device to avoid water condensation on lenses

FIG. 5. Control room at EPS Pomorzany.

5. CONCLUSIONS

Electron beam flue gas treatment process ensures simultaneous removal of SO_2 and NOx from flue gases. High removal efficiencies of both pollutants are obtained at optimal conditions for process realization. The monitoring and control systems are extremely important for achieving successful operation of the plant. The monitoring system should provide reliable and precise measurements of gas parameters in different points of industrial plant. The best available analytical techniques should be applied. Actual values of process parameters are employed by control system for their technological modification to ensure optimal conditions for electron beam irradiation of flue gases in the process vessel. The above requirements for monitoring and control systems are based on the experiences gained during operation of Kawęczyn pilot plant and Pomorzany industrial plant.

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POLYCYCLIC AROMATIC HYDROCARBONS REMOVAL FROM FLUE GAS BY ELECTRON BEAM TREATMENT — PILOT PLANT TESTS

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Abstract

Volatile organic compounds (VOCs) emitted from coal combustion belong to aliphatic, chlorinated, aromatic hydrocarbons, aldehydes and but as the most dangerous polycyclic aromatic hydrocarbons (PAHs) are considered. Many of them are involved in the formation of photochemical smog and depletion of stratospheric ozone. Some PAHs are mutagenic, carcinogenic or both. Tests at the pilot plant constructed at coal-fired power station were performed to estimate the influence of electron beam on PAHs concentration in flue-gas. The influence of electron beam dose on the global toxicity of flue gas components has been analyzed. The concentrations of PAHs decreased after irradiation.

1. INTRODUCTION

Volatile Organic Compounds (VOC), including Polyaromatic Hydrocarbons (PAH) are emitted in different processes, mostly combustion-based ones applied in power, chemical and metallurgical industries, municipal wastes incineration, etc. They are responsible for ozone layer depletion, ground level and photochemical smog formation, contribute to greenhouse effect, most of them being carcinogenic or/and mutagenic. Some of them are quite quickly destroyed at environment, other persist for a long time.

Persistent Organic Pollutants (POPs) are organic compounds that persist in the environment, are liable to bioaccumulate through the food web, and pose a risk of causing adverse effects to human health and the environment. The potential disorders caused by even relatively low levels of chronic exposure to POPs are thought to include reproductive and immune effects, developmental anomalies, and cancer. Due to resistance to degradation POPs have long environmental half-lives. Successive releases of these chemicals over time result in continued accumulation in the global environment. Most POPs are of anthropogenic origin. Anthropogenic emissions are associated with industrial processes, product use and applications, waste disposal, leaks and spills, combustion of fuels, and waste incineration. Many POPs are relatively volatile, therefore their remobilization and long-distance redistribution through atmospheric pathways often complicates the identification of specific sources. As a result of the tendency of POPs to move from warmer to colder environment even the Arctic ecosystem is exposed to some POPs at levels of concern. International treaties have been signed concerning persistent organic pollutants emission control. The emission of PAHs, due to their hazardous properties, should be controlled.

United Nations Economic Commission for Europe (UNECE) 1998 Protocol to the 197 convention on long-range trans-boundary air pollution on Persistent Organic Pollutants (UNECE POPs) Protocol was signed by 33 European countries, USA and Canada in 1998, 14 countries have already ratified it. PAHs in this protocol are mentioned as the key POPs emitted from stationary source (the coal combustion process is considered as one the biggest). The objective of the POPs Protocol is to control, reduce, or eliminate discharges, emissions, and losses of POPs to the environment (Statutes and Convention Texts of the International Organizations, LRTAP Convention [2]. However even the Protocol is not yet in force, some countries have already applied own PAHs emission limits, e.g.: in many of them benzo[a]pyrene is controlled, additionaly in some fluoranthene emission is limited up to 2[ng/m^3], naphthalene up to 100 [mg/m^3].

Unfortunately, there are not too many technologies available to treat vast quantities of off-gases containing organic pollutants in lean concentrations. Industrial off-gases have been cleaned by several

conventional methods: incineration, adsorption, absorption, and condensation. The methods to control VOCs often have had too low efficiency when considering new emission regulation, production of secondary pollutants and those techniques are quite expensive. New technologies are being developed and electron beam treatment seems to be an alternative technology for reduction of VOCs in air.

2. POLYCYCLIC AROMATIC HYDROCARBONS (PAHS)

The group of polycyclic aromatic hydrocarbons (PAHs) is known as a key pollutant among organic compounds emitted from coal-combustion process. They are also emitted from metallurgical sector. The compounds named PAHs consist of several fused aromatic rings made entirely from carbon and hydrogenfrom 2 (e.g. naphthalene $C_{10}H_8$, acenaphtylene $C_{12}H_8$) up to 6 (e.g. dibenzo[ah]anthracene $C_{22}H_{14}$), some examples of PAHs structures are shown in Fig. 1.

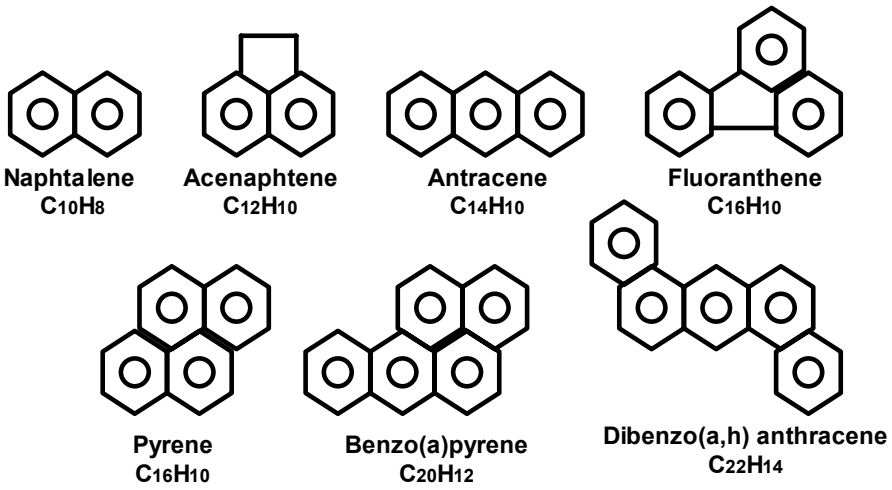

FIG. 1. Polycyclic aromatic hydrocarbons chemical structures.

Their concentration in flue-gas from coal combustion process ranges from few up to several hundreds μg/Nm^3 in the case of Polish coal and boiler type WP-120 [1].Their emission strongly depends on the kind of used fuel and process conditions. PAHs are known as dangerous, toxic and carcirogenic.

The emission of PAHs, due to their hazardous properties, should be controlled. United Nations Economic Commission for Europe (UNECE) 1998 Protocol to the 197 convention on long-range trans-boundary air pollution on Persistent Organic Pollutants (UNECE POPs) Protocol was signed by 33 European countries, USA and Canada in 1998, 14 countries have already ratified it. PAHs in this protocol are mentioned as the key POPs emitted from stationary source (the coal combustion process is considered as one the biggest). The objective of the POPs Protocol is to control, reduce, or eliminate discharges, emissions, and losses of POPs to the environment (Statutes and Convention Texts of the International Organizations) [2]

3. PRELIMINARY TESTS FOR EB PAHS TREATMENT

The preliminary experimental work for electron beam PAHs treatment, carried out at EB pilot plant has shown the positive results. After irradiation the concentrations of many PAHs decreased with the efficiency ranged from 3 up to 98%. The experiments carried out with ammonia added and without

have shown the positive influence of its presence. In the case of ammonia injection, PAH's removal effiencies were higher. The efficiency for benzo[a]pyrene increased from 27% without ammonia up to 43% for its presence, in the case of acenaphtene removal efficiencies values as 8% and 96% were observed respectively [1].

The results of experimental work done at pilot plant for typical DeSOx and DeNOx conditions, with the purpose to confirm the possibility to apply EB treatment as a multicomponent purification technology, covering also PAHs treatment are presented below.

4. PILOT PLANT

The experimental work has been carried out at EB pilot plant, placed at coal-fired electro-power station, installed on a bypass of the main flue-gas stream from boiler WP-120. Through the advancement of experimental work to pilot scale, it is possible to investigate removal of organic compound by electron beam treatment in industrially relevant conditions

The EB pilot plant is equipped with two accelerators (50kW, 500-800 keV each), installed on reaction chamber. The scheme of arrangement is shown in Fig.2.

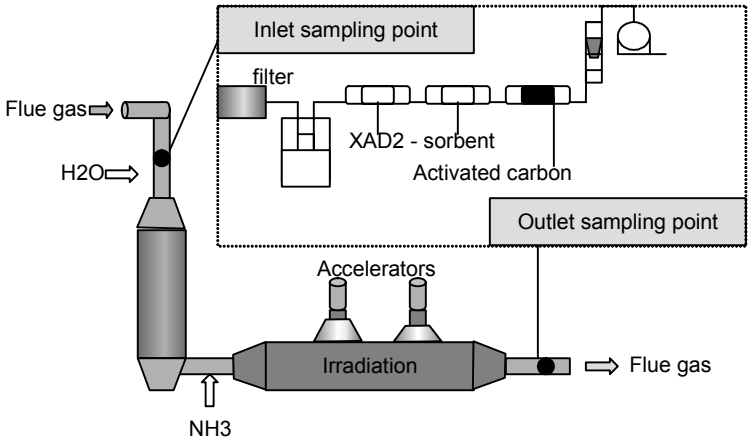

FIG. 2. The arrangement of pilot plant sampling points.

5. PROCESS CONDITIONS

The experimental process conditions were as follow: flue-gas flow 5.200 [Nm3/h], humidity 6% [vol], temperature 650C dose applied 8 [kGy], ammonia stoichiometry 0.8. These conditions are typical optimal for SO2 and NOx removal by electron beam. In the time of carrying out of experiments for volatile organic compounds, high removal efficiencies for SO2 and NOx were reached (85% for SO2 and 70% for NOx).

6. SAMPLING AND ANALYTICAL METHODS

The sample probes, made from stainless steel, fitted with gas filters, were installed at the inlet and the outlet of pilot plant, as it is shown in Fig.2. The inlet and outlet samples were taken simultaneously. In both cases two ceramic coaxial filters were used for removal of particulate matter from the sample. The high - performance filters were heated up to 150^0C to avoid condensation. After filtration the sample gas was transported through the heated stainless steel tube to the gas adsorption tubes (two tubes of XAD-2 resin and one tube of activated carbon) through the dry-ice-cooled condensate trap (to separate a condensate). The gas filters and gas transportation line were kept at the

same temperature, the sampling flow rate was 2 l/min and gas volume sampled was about 300 liters. Then, the condensate trap and the adsorbent tubes were analyzed within 48 hours to prevent degradation of trapped compounds.

Two steps of extraction i.e. with diethyl ether followed by extracting with toluene were used for organic matter desorption [3]. The whole aromatic fractions received was concentrated in the Kuderna-Danish evaporator and analyzed chromatographically using a GC-8160 Fisons gas chromatograph, equipped with FID detector, an on-column injector and a capillary column DB-5 (30 m, i.d. 0.32 mm, thickness 0.25 μm). For quantitative fraction analysis two internal standards (1, 2, 3 - thriphenylmethane and 9, 10 - diphenyloanthrancene) were applied.

7. RESULTS AND DISCUSSION

The results of carried out experiments are summarized in Table I and shown in details in Fig. 2. and Fig. 3. The process conditions in all three series were the same, as described in experimental chapter. In the Table 1 are included inlet ci and outlet co concentrations for 6 PAHs: 3-ringed phenanthrene C14H10 and anthracene C14H10, 4-ringed pyrene C16H10 and chrysene C18H12, 5-ringed benzo[a]pyrene C20H12 and benzo[e]pyrene C20H12. The emission is ranged from 0.105 [μg/Nm3] for pyrene in serie 2, up to 2.30 [μg/Nm3] for chrysene in serie 1.

The removal efficiency, defined as:

$$Ef= (ci-co)/ci,$$

where:

- ci – PAH inlet concentration [μg/Nm3]
- co – PAH outlet concentration [μg/Nm3]

are ranged from 1 up 92%.

In the serie 1 (anthracene) and serie 2 for phenanthrene, pyrene and chrysene the phenomena of negative efficiency removal is observed, the concentrations of these compounds are higher after irradiation. These compounds have in their structures 3 or 4 benzene rings, simultaneously for 5-ringed benzo[a]pyrene and benzo[e]pyrene in all three series high removal efficiency (up to 70%) is reached. Probably, the compounds consisting of less benzene rings are the products of higher-ringed PAHs destruction. In the Table I the toxicity factors TEFEPA for all mentioned PAHs are included [4].

TABLE I. THE CONCENTRATIONS OF PAHS BEFORE AND AFTER IRRADIATION AND PAHS REMOVAL EFFICIENCIES WITH TEF – EPA TOXICITY FACTOR RELATED TO BENZO[A]PYRENE

PAH	TEF_{EPA}	Serie 1			Serie 2			Serie 3		
		c_i	c_o	E	c_i	c_o	E	c_i	c_o	E
Phenanthrene	0.00	0.580	0.242	0.590	0.217	0.220	-0.012	0.230	0.062	0.73
Antracene	0.00	0.170	0.184	- 0,082	0.128	0.120	0.010	0.480	0.040	0.92
Pyrene	0.00	0.240	0.099	0.590	0.105	0.150	-0.429	0.640	0.078	0.88
Chrysene	0.00	2.300	1.087	0.530	0.537	1.520	-1.830	2.030	0.682	0.67
Benzo[a]pyrene	1.00	0.500	0.382	0.250	0.661	0.200	0.069	0.250	0.189	0.26
Benzo[e]pyrene	0.00	0.190	0.176	0.097	0.294	0.250	0.020	0.400	0.124	0.70
	$\Sigma c_* TEF$	0.500	0.382		0.661	0.200		0.250	0.189	

The typical results for two PAHs: anthracene and benzo[a]pyerene are shown in Fig. 3 and Fig.4. Inlet concentration is shown with blue colour, while outlet concentration has blue-gray colour.

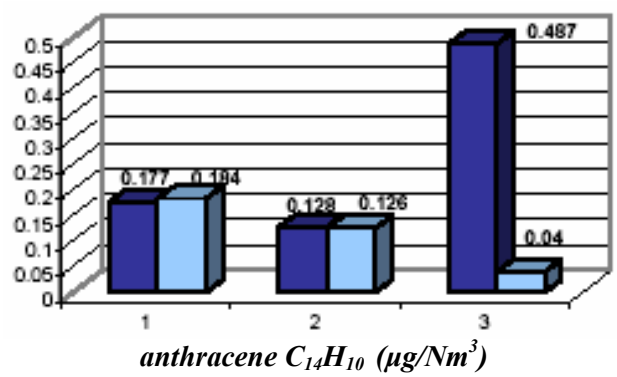

FIG. 3. The anthracene $C_{14}H_{10}$ concentration before and after irradiation

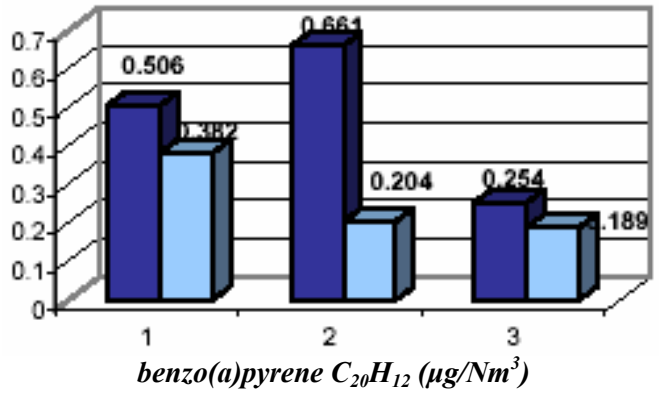

FIG. 4. The benzo[a]pyrene $C_{20}H_{12}$ concentration before and after irradiation.

The overall toxicity $\Sigma c_* TEF$ based on the EPA toxicity factors TEF_{EPA} included into Table 1., together with the efficiency of PAHs removal suggest, that the global toxicity of flue gas after irradiation by electron beam decreased.

8. CONCLUSIONS

The results of experimental work presented above can be concluded in following points:

- Removal efficiency of various PAHs ranged from 1 up to 92% in typical conditions for SO_2 and NO_x removal.
- For some PAHs negative efficiency (anthracene, pyrene, chrysene) – they are produced in EB process. These less-ringed compounds are probably the products of higher-ringed PAHs destruction
- The overall toxicity of flue-gas stream after irradiation decreased.

In the case of pilot plant test this kind of conclusion can be made, because of wide spectra of emitted organic compounds it is not possible to determine the products of single compound radiolysis. For the purpose of detailed study of single PAH behaviour under electron beam, the laboratory tests in flow installation has been carried out.

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ELECTRON BEAM TREATMENT OF INDUSTRIAL OFF-GASES CONTAINING CHLORINATED ORGANIC COMPOUNDS AND DIOXINS

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Abstract

Chlorinated organic compounds and dioxins emission are very harmful to the environment and human's health due to their carcinogenic and mutagenic effect. Decomposition of chlorinated organic compounds and dioxins contained in the industrial off-gases by using electron beam technology is reviewed. General mechanism of Cl-VOCs by using EB technology is outlined. Experiments of Cl-VOCs treatment from laboratory scale to the pilot scale are described. It is drawn a conclusion that EB is a promising technology to remove multiple pollutants from off-gases including chlorinated organic compounds and dioxins.

1. INTRODUCTION

Although most of VOCs are emitted from nature sources, such as forest, but in urban area, VOCs are mainly come from human's activity. For example, VOCs are emitted from industrial solvent applications, automobiles, power stations and waste incineration process.

In Japan, 354 substances are listed as pollutants, the total emission of them is equal to 898 308 t/year and VOC emissions to the atmosphere accounted for more than 80% of this number [1]. Some VOCs emissions are very harmful to the environment and human health due to their carcinogetic and mutagenic effect, PAHs and dioxins are listed as priority pollutants by EPA.

Chlorinated aliphatic and aromatic hydrocarbons, which are emitted from coal fired power stations, waste incinerators and chemical fabrication industries, are very harmful to the environment and human health. Recent studies show that chlorinated aliphatic and aromatic hydrocarbons are suspected to be the precursors of dioxin's formation. High temperature incineration and similar thermal processes lead to the formation of "dioxins" and dibenzofuranes or PCDD/F, in trace amounts. Their toxicity and their low biodegrability require keeping emission as low as possible [2]. This large family of chemical compounds, respectively 75 and 135 congeners of planar chlorinated aromatic compounds. PCDDs and PCDFs are compounds with similar chemical properties. Each compound comprises two benzene rings interconnected by oxygen atoms. In the case of PCDDs, the benzene rings are joined by two oxygen bridges, and in the case of the PCDFs, the benzene rings are connected by a carbon bond and an oxygen bridge. The 17 congeners chlorinated in 2, 3, 7, 8 are toxic.

Polychlorinated dibenzeno-p-dioxins and dibenzenofurans (PCDD/F) gained widespread notoriety when 2,3,7,8-TCDD was found as a contaminant in Agent Orange, a defoliant used in Vietnam. Acute contamination incidents such as those at Times Beach, MO, USA and at Seveso, Italy, helped to further familiarize the public with hazards of these compounds. It was thought that these compounds were formed only as byproducts during chemical manufacturing processes, however twenty years back, it was discovered that PCCD/Fs are being emitted during combustion processes. These are ubiquitous environmental pollutants, which are atmospherically transported from combustion sources to almost all areas of the globe. The global emission of PCDD/F is estimated to be 1800 – 3000 kg/year [3].

In the UK the contribution to the total PCDD and PCDF emissions into atmosphere from combustion sources is estimated to be ca. 25% from domestic coal fires, 10% from industrial coal fired plants and 5% from industrial coal-fired stations. The importance of these coal combustion sources together equals that of municipal waste incineration [4].

Dioxin is one of the most toxic chemicals known. A report released for public comment by the US Environmental Protection Agency clearly describes dioxins as a serious public health treat. The EPA report confirmed that dioxin is a cancer hazard to the people, that the exposure to dioxin can also cause severe reproductive and developmental problems (at levels 100 times lower than those associated with its cancer causing effects); and that dioxin can cause immune system damage and interfere with regulatory hormones. When establishing the toxicological effect of the dioxins present in the exhaust gases, it is common practice to estimate so-called "toxic equivalent" value based on the comparative toxicity of each dioxin congener/isomer present in the gas. It is due to the fact that although 2,3,7,8-TCDD is the most toxic of these compounds, it is generally present in much lower concentrations than the less toxic isomers. For this purpose, a number of equivalence models have been developed. Most often used are those developed by US EPA and WHO [5].

Dioxin's emission into atmosphere will cause severe environmental problem by environment contamination and its emission has to be strictly controlled. The Government of Japan established new emission standards for dioxin in 1999, in which the existing Municipal Waste Incinerators (MWIs) having incineration capacity over 4 t/h must reduce the emission lower than 1.0 ng-TEQ/N cubic meter of flue gases by December 2002. An emission limit of 0.1 ng-TEQ was set for newly constructed incinerators [6].

Unfortunately, there are not too many technologies available to treat vast quantities of off-gases containing organic pollutants in lean concentrations. Industrial off-gases have been cleaned by several conventional methods: incineration, adsorption, absorption, and condensation. The methods to control VOCs often have had too low efficiency when considering new emission regulations, production of secondary pollutants and those techniques are quite expensive. New technologies are being developed and electron beam treatment seems to be an alternative technology for reduction of VOCs in air. Some tests performed in different countries have shown that electron beam technology can be a promising technique in these applications.

Some special extremely toxic groups of pollutants are chlorinated hydrocarbons, dioxins among them. Chlorinated volatile organic compounds (Cl-VOC) degradation has been broadly studied in the recent years. Different technologies, such as bioreactors, catalytic oxidation, photoinduced decomposition, thermal plasma process, and nonthermal plasma processes, have been applied. Electron beam (EB) treatment is a promising technology for removal of low concentration Cl-VOC contained in flue gases. This is because chloroethylenes have low ionization potential and high electron capture cross section. Chlorinated methane and ethylene have been successfully studied by using this technology. Good removing efficiency for chlorohydrocarbons, dioxins and PAH by using EB process has been studied in lab scale and pilot scale [7-11], promising results were achieved.

2. PROCESS DESCRIPTION

In the EB process, when fast electrons from beams are absorbed in the carrier gas, they cause ionization and excitation processes of nitrogen, H_2O and oxygen molecules in the carrier gas. Primary species and secondary electrons are formed. The secondary electrons are thermalized very fast within 1 ns in air at the pressure of 1 atmosphere. These active species and secondary electrons react with VOCs to cause its degradation.

For chlorinated aliphatic hydrocarbons' decomposition (e.g. Chloromethane, chloroethylene, chloroethane and others.), Cl- dissociated secondary electron –attachment, Cl atoms addition reactions (for chlorinated ethylene) followed by peroxy radical reaction and OH radicals reaction with VOCs play very important roles for VOCs decomposition. Example is given in Fig.1 [12]

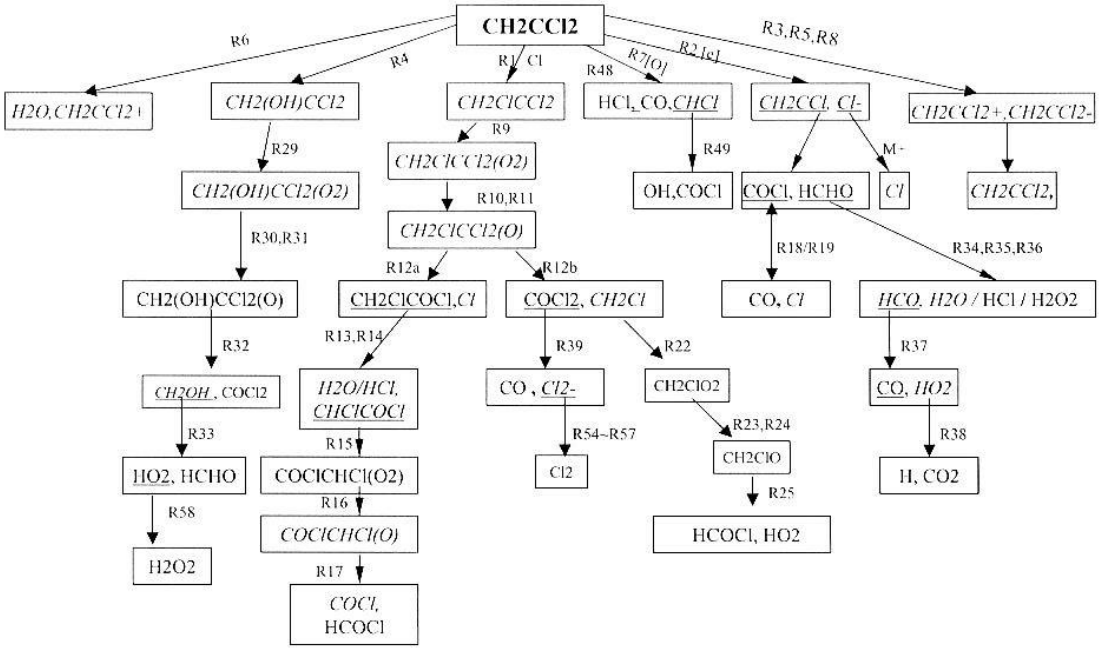

FIG. 1. Schema of reaction pathways of 1,1-DCE decomposition and organic products formation.

General mechanism of chlorinated aliphatic hydrocarbons can be written as follows:

- CxHyClz + e $\rightarrow$ Cl- + R•.
- R• + O2 = RO2•
- 2RO2• = 2RO• + O2
- RO• = products + Cl•
- RO• = products + R1•

For chlorinated aromatic hydrocarbons, VOCs decomposition will mainly go through:

1. Positive ions' charge transfer reactions

M+ + RH (RH= VOC, eg. 1,4-dichlorobenzene or PAHs) = M + RH+

Because RH has lower ionisation energy (IE) (Benzene: IE = 9.24 eV; PAHs: IE <10 eV) than most primary positive ions (IE > 11 eV), such as N2+, O2+ formed above, part of VOC will be decomposed by rapid charge transfer reactions.

2. Radical – neutral particles reactions

- •OH radical plays very important role for VOC decomposition, especially when water concentration is 10 %. •OH radicals react with VOC in two ways:
- •OH radical addition to the aromatic ring (e.g. toluene)
- •OH + $C_6H_5CH_3$ = R1•

- $C_6H_5CH_3$ + •OH = R2• + H2O (H atom abstraction)
- C_6H_6 + •OH = C_6H_5OH + H (H atom elimination)

Radicals (R1•, R2•) formed above go though very complex reactions: O_2 addition, O atom release, aromatic –CHO (-dehydes), -OH (-OCl) compounds formed or ring cleavage products:

- R• (R1•, R2) + O_2 = RO_2•

- $2\ RO_2\bullet = 2RO\bullet + O_2$
- $RO_2\bullet + NO = RO\bullet + NO_2$
- $RO\bullet + O_2 = HO_2\bullet$ + products (aromatic-CHO, -OH)
- $RO\bullet \rightarrow$ aliphatic products

3. INDUSTRIAL PILOT INSTALLATIONS

3.1. Pilot plant investigations

Pilot plant investigations for VOCs treatment have been carried out in Japan, Germany, USA. The pilot plants are listed in table II.

TABLE II. PILOT PANT FOR VOCS TREATMENT BY USING EB TECHNOLOGY.

	Operator	Pollutants	Applications	Flow rates [m^3h^{-1}]	Operation
AGATE-M	FZK and IOM (Germany)	VOC, PCDD/F, CHC	Solvent emissions, waste incinerator, site remediation	1,000	Since 1994
MINE	IOM(Germany)	CHC	Site remediation	300	Since 1995
Mobile Plant	Zapit echnology Inc. (USA)	CHC	Site remediation		Since 1992
Takahama Clean Center	JAERI(Japan)	PCDD/Fs	Waste incinerator, site remediation	1000	Since 2000

Pilot facilities and remediation efficiency for CHC and VOCs were described in details by Paur [13]. For PCDD/Fs treatment, a flow of 1000 m^3N of flue gas from a waste incinerator was treated by EB technology. It was found that: over 90% PCDD was decomposed at 12 kGy dose for initial concentration of PCDD being 21-110 ng/m3N (AGATE-M plant, Germany) and 0.22-0.88 ng-TEQ/m3N (Takahama Clean center, Japan), respectively. PCDD/Fs decomposition is probably caused by: Cl dissociation, benzene ring cleavage or O- bond cleavage thorough by reactions with actives species. Mechanism of dioxins decomposition is shown in fig.2.

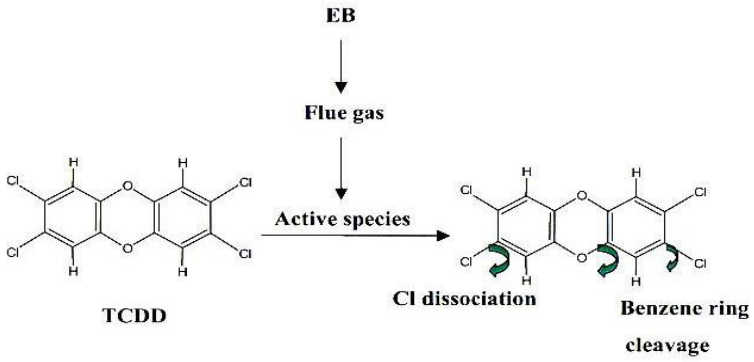

FIG. 2. Possible dioxins decomposition under EB irradiation

3.2. Dioxins treatment from MSWI by using EB technology in Japan - Takahama Clean Center

The electron beam treatment of PCDD/Fs was performed at a site of the Takahama Clean Center, which treats 450 t (150t × 3 furnaces) of solid waste in 1 day [1, 6]. The incineration of 150 t of wastes caused approximately 40 000 m3N/h flue gas. Electric precipitators with the addition of dry activated carbons were used to control the PCDD/F emission from this MSWI. An incinerator flue gas of 1000 m^3N/h for the test facility was obtained at the downstream of one of the precipitators using an induced fan and backed to the main stream in incinerators stuck after an electron-beam treatment.

An accelerator (300 kV * 40 mA) was used to irradiate the incineration flue gas passing through a reactor (120(l) × 45 (w) × 30 (h) cm3). Decomposition of PCDD and PCDF were shown in fig. 3 and 4, respectively.

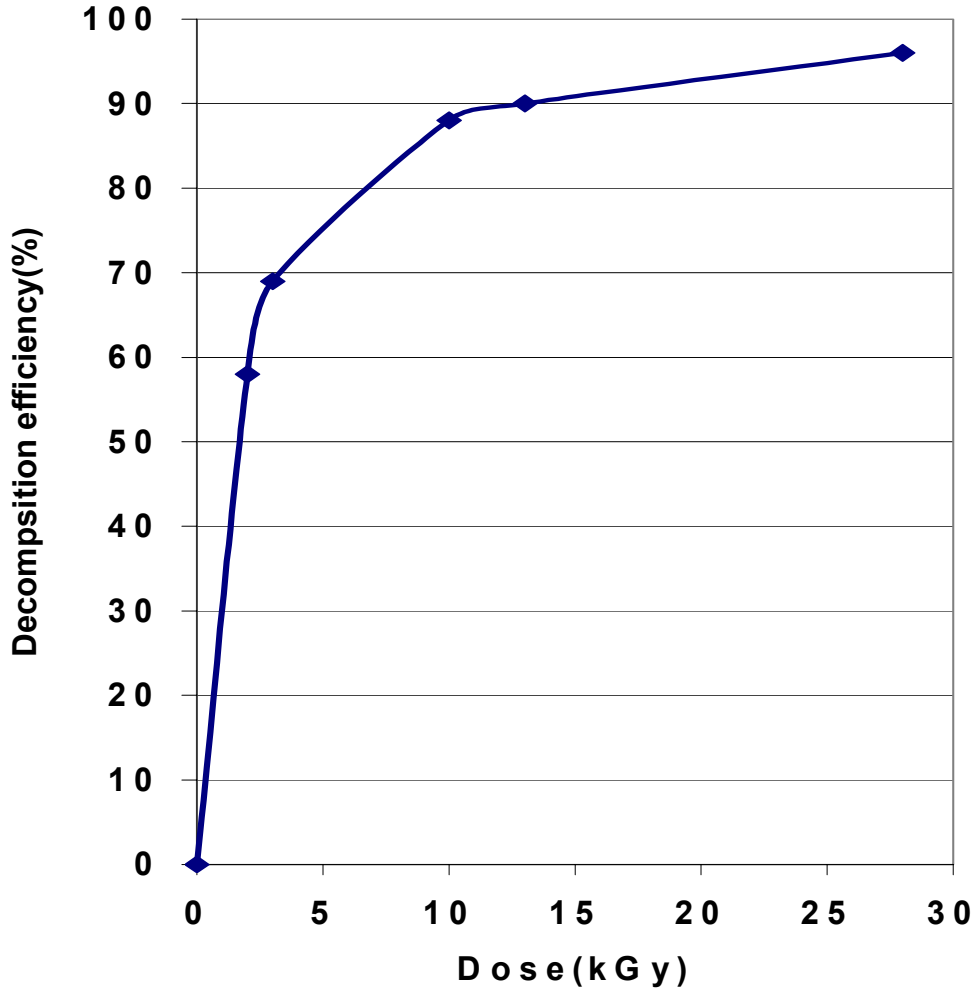

FIG. 3. Decomposition efficiency of PCDD in MSWI flue gases with electron beams. The initial concentration of PCDD is in the range of 0.22-0.88 ng-TEQ/m^3_N.[6].

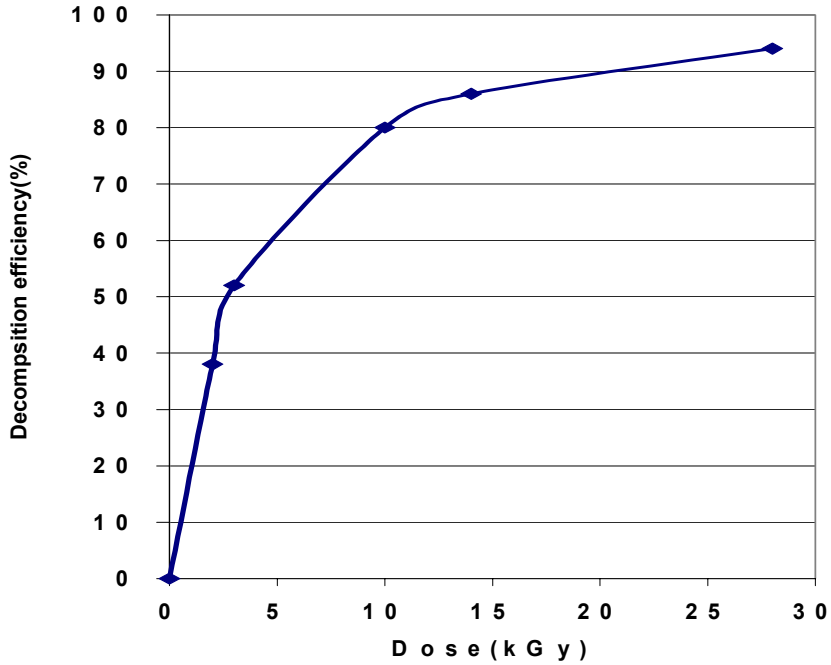

FIG. 4. Decomposition efficiency of PCDF in MSWI flue gases with electron beams. The initial concentration of PCDD is in the range of 0.35-12.4 ng-TEQ/$m^3_{N.}$[6]

3.3.Chlorinated aromatic hydrocarbons treatment by EB irradiation in INCT, Poland.

Chlorinated organic compounds are suspected as precursor of dioxins' formation. Its emission into atmosphere from waste off-gases causes environmental pollution. 1,4-dichlorobenzene (1,4-DCB) degradation under EB irradiation was studied in Institute of Nuclear Chemistry and Technology (INCT) [14]. The results are shown in fig.5. It was found that over 50% 1,4-DCB with the initial concentration 30-90 ppm was decomposed at 20 kGy absorbed dose.

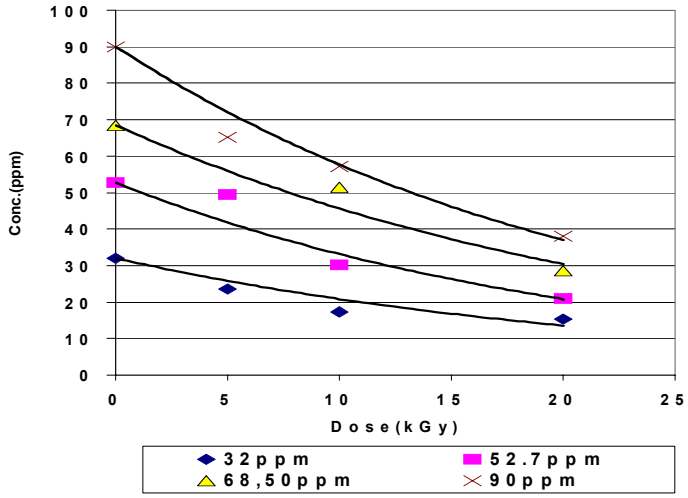

FIG. 5. 1,4-dichlorobenzene/air mixture vs. dose in EB irradiation

4. CONCLUSIONS

Results of laboratory experiments performed in INCT and pilot test in Japan, demonstrated that chlorinated organic compounds and dioxins can be removed by using EB technology. Electron beam irradiation is a promising technology to clean industrial off-gases containing multiple-pollutants.

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RECENT DEVELOPMENTS IN THE U.S.A IN THE APPLICATION OF FGD-BASED TECHNOLOGY FOR SIMULTANEOUS SO_2, NO_X AND MERCURY REMOVAL

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Abstract

A hybrid process design is proposed that will enhance the technical and economic feasibility of use of electron beam technology for flue gas cleaning. A partial chemical oxidation of the flue-gas acid gases is carried out prior to treatment of the gas by ammonization, final evaporative cooling and electron irradiation. Differentiating between low and high sulfur fuel applications, this assessment presents means of substantially improving the cost-effectiveness and practicality in application of the electron beam (E Beam) flue gas treatment process, this through pre-oxidation of the dedusted raw flue gas using selected chemical oxidants. Particularly in low sulfur fuel applications, encountered worldwide, parasitic electric power consumption including electron accelerator energy demand can, importantly, be reduced, e.g. to 2% of the capacity of the generating unit served. This matches that of combined, multi-process, conventional means of removal of SO_2 and NO_x. Optimal selection and dosage of relevant, available, chemical oxidants can advantageously moderate required accelerator capacity and electric energy/usage. This can ensure cost-effectiveness competitive with all other means of removing SO_2 and NO_x in either a simultaneous or combined (multi-step) manner.

1. OVERVIEW: PERTINENT NEW US DEVELOPMENTS/TRENDS

1.1. Existing US facilities for step-wise combined removal of SO_2 and NOx

By the end of year 2004, 45 plants comprising of 68 generating units (of 35 000 MW aggregate capacity) in the US will operate both FGD (flue gas desulphurization) and SCR (selective catalytic reduction) systems for combined removal of SO_2 and NO_x (Table I) [1]. Note that, absent newer more attractive process alternatives, such aggregate capacity is forecast to increase to the range of 117 000 to possibly 160 000 MW by year 2011.

Total cost for SCR typically exceeds $US 1,500 per short ton of NO_x removal. The O&M costs of SCR systems depend on the amount of NO_x at the SCR inlet [14]. For instance O&M costs vary between 160 US$/tone (176 US$/tone), for NO_x emissions of >0.6 lb/million Btu (0.26 kg/GJ) and <0.25 lb/million Btu (0.11 kg/GJ) respectively for a >400 MW power plant. The same range applies for smaller power plants (50-200 MWe). The typical cost for an FGD retrofit lies in the range of 150-270 $/kW. Some designs cost less than this. Recent literature [15], however, reports capital cost for the E Beam Process, providing simultaneous removal of SO_2 and NO_x, at less than $US 200/kW. The most notable progress in the U.S. in simultaneous scrubbing of SO_2 and NO_x is being made by Powerspan's ECO (Electro Catalytic Oxidation) Process [16]. It uses an electrical reactor to oxidize NO_x for removal by a wet ammonia scrubbing system. It is being tested on an 110,000 standard-cubic-feet-per-minute flue gas slipstream (equivalent to a 50 MW plant) at First Energy's R.E. Burger Plant in Shadyside, Ohio.

Test results have indicated SO_2 removal consistently over 99%. Problems with the system's absorber performance have diminished NO_x removal from an expected 90% to 70-80%. In a 1MW slipstream pilot test in 2002 and 2003, the ECO system achieved a 90% mercury removal rate, along with 90% for NO_x and 98% for SO_2. Removal rates for particulate matter, which will be determined through opacity measurements, are due for monitoring in September. According to a cost analysis by Sargent and Lundy, the capital cost of installing the ECO System on a 510MW plant would be $US 120.4 million, about 32% cheaper than conventional controls for NO_x, SO_2 and mercury. Total operation and maintenance costs for ECO would be about $US 2.7/MWh, about 34% cheaper than conventional controls, (including an SCR, limestone scrubber, and activated carbon injection with bag house). Operating costs are offset by revenue from by-product ammonium sulphate.

TABLE I. US PLANTS WITH BOTH SCRUBBERS AND SCR (MW)

State	2004	2011	Difference
Alabama	1,125	6,670	5,545
Arkansas		800	800
Arizona		400	400
Colorado	100	100	
Delaware		786	786
Florida	1,090	1,590	500
Georgia		3,110	3,110
Iowa		790	790
Illinois	800	7,360	6,560
Indiana	5,060	10,540	5,480
Kansas		660	660
Kentucky	5,600	11,900	6,300
Massachusetts		1,450	1,450
Maryland		1,730	1,730
Maine		632	632
Michigan	65	5,130	5,065
Minnesota		598	598
Missouri	540	1,660	1,120
Montana		780	780
North Carolina		6,110	6,110
New Jersey	611	1,600	989
New York	840	1,330	490
Ohio	4,025	11,250	7,225
Pennsylvania	4,850	10,800	5,950
South Carolina	1,550	4,750	3,200
Tennessee	2,600	3,950	1,350
Texas	605	2,600	1,995
Virginia	240	4,270	4,030
Wisconsin		1,840	1,840
West Virginia	5,080	11,500	6,420
Wyoming	90	90	
TOTAL	35,000	117,000	82,000

1.2. US coal use, past, present and future

Like China, North America is heavily dependent on coal as its main energy source, particularly for the generation of power. The U.S. has the world's largest reserves of coal, i.e. 250 g tones (versus approximately 1 000g tones for the entire world.) The U.S. and Canada between them annually mine over 1g tone of coal. The projected electricity generation mix in the U.S. and Canada as related to fuel use is shown in Table II [2].

TABLE II. ELECTRIC POWER GENERATION IN NORTH AMERICA, TWH

	1990	2000	2010	2030
Coal	1,780	2,230	2,240	2,880
Oil	147	140	136	99
Gas	391	664	1,400	2,120
Nuclear	685	873	882	647
Hydro	570	607	644	670
Other renewable	89	98	165	473
Hydrogen fuel cell	0	0	0	122
Total	3,660	4,609	5,460	7,016

1.3. Regulatory measures, including anticipated long term, stack-NO_x emission limit

Federal legislation and enforcement has since 1990 focused on means of abating emissions of SO_2 and NO_x. Transported by prevailing winds from Midwest to northeastern U.S., they act as act as precursors in downwind formation of ozone, acid precipitation, PM-2.5 particulate matter, (i.e. suspended particulates smaller than 2.5 microns,) and visibility-obstructing haze. Electric utility SO_2 emission concentration has been reduced, on average, (in part by retrofitting of FGD, i.e. flue gas desulfurization, but largely by switching to low-sulfur coal), to less than 400 ppm. This has occurred in conjunction with mitigation of acid precipitation and is programmed for a substantially lower level, which will decrease ambient concentration of the ultra-fine particulates, above, much of it sulfate and nitrate solids. Extensive retrofitting of high-dust SCR is taking place to reduce average NO_x emission concentration to 100 ppm, (3% O_2 basis), this to limit ambient ozone concentration, i.e. automotive smog, along the east coast. The U.S. Department of Energy projects stepwise reduction in average NO_x concentration, first to 35 ppm in the short term, then to 6 ppm by year 2015, to achieve the comparatively new fine particulate air quality criteria. Also, major reduction in mercury emission is soon to be mandated amidst much controversy and debate.

1.4. Resulting incentive/need to apportion secondary DeNO_x duty among distinct cleanup steps

The spectacularly low NO_x emission concentrations ultimately targeted can be seen to be beyond the practical capability of high dust SCR technology, in part because of the comparatively low ash content of common US medium-high sulfur bituminous coal and its thus limited ability to beneficially sorb gaseous SO_3 and excess gaseous ammonia reagent, i.e. ammonia slip. Major impairment of Ljungstrom air preheater operation, i.e. corrosion and fouling, is brought about by high residual ammonia concentration following that deNO_x step, which together with flue gas SO_3 and H_2SO_4(v) forms a corrosive adherent sludge in combination with fly ash.

Raw gas fly ash concentration is not sufficiently high to adequately sorb these troublesome gases, ammonia and SO_3. In continued new use of SCR technology its deNO_x performance will no doubt need to be augmented by supplemental downstream process means that most cost effectively reduces a typically modest NO_x concentration (resulting from upstream deNO_x steps,) e.g. 50-70 ppm, to as low as the above mentioned, 6 ppm level. Serving as SO_2 and mercury emission abatement means as well, electron beam technology may be an important candidate process with which to most cost effectively provide this unique and challenging deNO_x duty. All other reasonably practical types of secondary deNO_x may be unable to cope with process design limitations in seeking to do so particularly in conjunction with existing, already built, coal power plants.

1.5. A continuing central focus on massive decrease in national (U.S.) NO_x emission inventory

Pre-1990 total NO_x emissions from electric powerplants in excess of 10 million annual short tons continue to be systematically reduced. It was initially decreased by boiler primary $deNO_x$ measures required by the 1990 Clean Air Act Amendments. This has been followed by widespread retrofit installation of secondary $deNO_x$, principally high-dust SCR, to meet an early, but now superseded, one-hour ambient ozone standard. A new implementation rule, encompassing an 8-hour ozone ambient-air standard, adopted on April 15, 2004, (maximum of 0.085 ppm ozone averaged over 8 hours,) will lead to a requirement for increased electric utility NO_x reductions. In time, with average powerplant NO_x emission decreased to 6 ppm, as indicated above, powerplant plant NO_x emission, as a whole, will be virtually negligible compared to that from mobile sources.

1.6. Advantages and shortcomings of continued dominance of NO_x abatement market by principal use of high dust SCR

1.6.1. Shortcomings of SCR

While SCR retrofitting has become increasingly common and routinely relied upon currently as the principal secondary $deNO_x$ means, major shortcomings have become well known:

- High capital cost, (in excess of US$150 per kW in many retrofit applications), and complexity with uncertainty as to the magnitude of site-specific retrofit factors impacting installed cost
- Major plot space structural complexities in fitting the SCR reactor upstream of the existing Ljungstrom air preheater
- Highly critical need for uniformity of distribution and characteristics of flow across the cross-sectional face of the catalytic reactor inlet, e.g. gas velocity and temperature, ammonia, etc[3].
- High chemical cost, particularly when it is required that urea supply and storage, not that of anhydrous ammonia, will be the basis for ammonia reagent feed to the flue gas
- Adverse impact on catalyst performance of U.S. high-arsenic bituminous and high-calcium subbituminous coals
- Major operational impact of fouling of the downstream air preheater in common high-sulfur, low-ash, U.S. bituminous coal service [6]
- Excessive ammonia slip concentrations, particularly at the end of boiler operational campaign in applications at high, i.e. 90%, NO_x removal efficiency
- Substantial additional operational cost penalty and complexity in the short term when SCR operation becomes mandated on a 12 month a year basis rather than only during the 5-month, warm weather, “ozone” season.

1.6.2. Competitive strengths of high-dust SCR technology

Attractive aspects include:

- Proven technology with more than 200 GW of worldwide capacity
- Technology availability
- Major R & D background and support
- Marketing momentum with continued widespread application in new boiler as well as retrofit service
- Servicing by a large and diverse supporting industry including cognizant catalyst suppliers, architect engineers and system designers.

1.7. Augmentation/substitution for SCR DeNO$_x$ use via utilization of simultaneous multi-pollutant removal employing flue gas pre-oxidation by commercial oxidizing chemicals

Using the E Beam process (or other gas scrubbing means) as the final flue gas cleaning step, acid gases and elemental mercury in the dedusted stack gas can be more readily removed, this accomplished simultaneously, by their upstream gas-phase oxidation by chemical oxidants [4-6].

1.7.1. Selection of chemical for flue gas oxidizing duty

Some of the principal characteristics of major chemicals in commercial use as oxidizing agents are detailed in Tables III and IV.

1) Ozone use for NO_x oxidation without significant SO_3 formation has been commercially developed in the U.S. by BOC Gases America. High unit cost per Table IV may limit its practical use to "trim" applications in which the scrubber, wet or dry, operating as the SO_2 removal means serves downstream of efficient primary or secondary deNO$_x$ facilities, to simultaneously remove only a very small amount of NO_x from a low NO_x concentration inlet gas.

2) A very promising technique in dry scrubbing utilizes ClO_2 as the in-situ formed, oxidizing agent after its in-duct generation from sprayed chemical solution originating from a commonly available supply of sodium chlorate crystals. Commercial suppliers focused on its market include ERCO Worldwide, Toronto, Ontario, Canada. By its nature, ClO_2 gas, formed within a flue gas stream that has been evaporatively cooled to less than 200^{o}F, sequentially oxidizes SO_2 to SO_3 followed by oxidation of NO to NO_2 and with oxidation of NO_2 to N_2O_5 thereafter. In dry scrubbing that uses lime alkali, all pollutant gas species are sorbed:

- SO_3/H_2SO_4(v): removed even more efficiently than is SO_2, resulting in calcium sulfate reaction product
- N_2O_5: sorbed in the same ready manner in which common HCl gas is collected, analogously yielding calcium nitrate and calcium chloride solids
- Chemically reduced ClO_2 gas oxidant generation: yielding calcium chloride solids in the removal step.

3) The U.S. firm, EWT Holdings Corporation, has developed a form of $C_2H_4O_4$, (akin to gluconic acid and additives thereto) used in solution that has a reported oxidation potential far greater than that of any other chemical cited in Table III and is available at comparatively modest cost. Absence of significant hazard in storage or use may lead to attractive commercial use, particularly in dry mode gas cleaning operations.

1.7.2. Technical development of mechanical equipment for effectively contacting flue gas with oxidants solution

Long term development of the advanced, linear, variable gas atomization (VGA) spray nozzle [7] offers advantageous large-capacity means of oxidizing NO_x and elementary mercury in raw flue gas via in-duct injection of solutions of oxidizing chemicals. Dual fluid atomizing combines fine droplet formation with a tailored spray pattern providing rapid and complete intermixing and chemical reaction with the flue gas across the cross section of the raw gas duct minimizing deposition and corrosion.

Optimum atomization is achieved by the formation of a thin liquid sheet (with a thickness of approximately 25 microns) extending over the full length of each nozzle modular section. The liquid sheet flows in the same forward direction as, and issues between, two adjoining, high velocity sheets of atomizing airflow.

The latter simultaneously converge to quickly form a nozzle throat, also in the form of a slit, at the nozzle exit. The liquid atomization occurs primarily within this throat, which is the region of maximum gas mass-velocity (density x velocity) and maximum energy transfer to the two opposite surfaces of the liquid sheet. The fine solution droplets in the expanding flat spray plume discharging from the nozzle intermix with, absorb and react with pollutant gases while cooling the flue gas, rapidly evaporating without reaching inner walls of the duct. By maximizing the transfer of the mechanical energy of the low pressure (40-60 psig) compressed atomizing air to the direct formation of the fine droplet sizes, (mean size less than 25 microns), in use of this nozzle design the required air pressure and parasitic energy consumption is minimized.

Individual nozzles are mounted in-line, end-to-end, within a cylindrical-pipe lance that extends across the flue gas duct. The sprays issue through windows on the downstream side of the lance to form a continuous flat spray plume that induces flue gas sweep around both sides of the lance. The expanding spray plume thus rapidly entrains the flue gas stream to achieve prompt intermixing. Equal spacing of the linear-nozzle/lance assemblies in a cross-sectional plane of the duct provides uniform and complete multi-phase interaction. Coupled with efficient atomization, the design enables the required degree of intra-phase mass transfer and chemical reaction to be achieved with minimum gas residence time.

TABLE III. CHEMICAL PROPERTIES OF OXIDANTS

Oxidant	PH	Oxid. Pot.	Mol. Wt.	[0] Cont (W/W)	Usage lb/lbs
O_2	acid	1.23	32	1.00	0.5
(oxygen)	base	0.40			1.0
O_3	acid	2.07	48	0.33	1.4
(ozone)	base	1.24			1.4
H_2O_2	acid	1.78	34	0.47	1.0
(hydrogen peroxide)	base	0.85			4.0
Cl_2	acid	1.36	71	0.22	4.0
(chlorine hypochlorite)	base	0.90	51	0.21	4.0
ClO_2	acid	1.71	67	0.60	1.0
(chlorine dioxide)	base	1.16			4.0
$KMnO_4$	acid	1.68	158	0.25	3.1
(potassium permanganate)	base	0.58		0.15	12.4
Fe^{+2} (ferrous iron)	All	N/A	56	N/A	1.6

TABLE IV. COSTS OF COMMON OXIDANT SYSTEMS

Oxidant	Moles	Wt. %	Chem. Cost ($US/lb)	Chem. Cost ($US/lb O_2)
O_3 (a)	1	33	1.78	5.36
H_2O_2 (b)	1	47	0.68 to 1.00	1.45 to 2.13
Cl_2 (b)	0.5	23	0.063 to 0.10	0.54 to 0.60
$KmnO_4$ (c)	0.5-2.5	5 to 25	1.21 to 1.41	4.80 to 28.4
ClO_2 (d)	2.5	59	1.58	3.57

Notes: a. Costs include generation equipment.
b. Costs based on chemical cost only.
c. Range includes impact of pH on reaction chemistry. Lowest cost is based on reaction: $MnO_4^- + 8H^+ + 5e \rightarrow Mn^{2+} + 4H_2O$.
d. Based on generation of ClO_2 from chlorate and chlorine.

1.8. Overall inherent superiority of dry/semi-dry scrubbing means in achieving simultaneous multi pollutant removal

1.8.1. Overview

Dry means of simultaneous removal of diverse flue gas pollutants can be seen to be generally superior to the common wet type of gas cleaning, and E Beam Process design can be seen to be the most advanced commercial technology of this former type.

1.8.2. Superiority of dry over wet scrubbing

Lime-based dry flue gas desulfurization [8], (particularly in use of circulating fluid bed reactors), and the E Beam Process have advantages in common as follows in their dry operation:

- Ultra-high efficiency of removal of SO_3 and H_2SO_4(v), virtually reducing the flue gas sulfuric acid dewpoint temperature and acidic corrosion tendency to nil i.e. to less than the water dewpoint temperature
- Low stack discharge opacity free of sulfuric acid mist
- Avoidance of wet, i.e. surface-wetted, operation of the internals of the gas cleaning system as well as the exhaust breeching and stack
- Avoidance of need for high cost, corrosion resistant construction for surfaces in contact with the treated flue gas
- Reliable operation of system booster fan positioned downstream of the gas cleaning system with reduced horsepower demand, i.e. horsepower being directly proportional to the actual flow volume of gas passing through the fan
- Resulting convenient, preferred operation of the gas cleaning system under a negative gas pressure condition
- Avoidance of need for a wet stack operation and its necessarily large diameter, low gas velocity design
- Avoidance of poor dispersion of the stack gas discharge in the atmosphere
- Gas cleaning residue is in a convenient dry, unwetted form
- Water consumption is minimal
- No liquor/liquid effluent
- High particulate removal efficiency is provided by dry past collection of particulate matter.

2. ADVANTAGES/DISADVANTAGES OF E BEAM PROCESS AT BITUMINOUS COAL FIRED PLANTS WITH NO EXISTING SCRUBBERS FOR SO2 REMOVAL

All such existing unscrubbed coal fired generating units are favorable candidates for commercial application of the E Beam process [9].

2.1. Superiority of E Beam technology over other dry process means

The Electron Beam process may be the most attractive of all dry designs because:

- Modest capital cost, $US 200/kW [9]
- Throwaway disposal of collected wastes is not required.
- Major net revenue is gained via conversion of reagent ammonia to high value sulfur/nitrogen, NPKS (nitrogen-phosphorus-potassium-sulphur) fertilizer blending stock comprising ammonium sulphate, nitrate and chloride.
- In the face of rising costs in the U.S. for natural gas, ammonia and urea process economics is enhanced, (the converse being applicable in reagent supply to SCR.)

2.2. Anticipated future SCR application and use

However, designated as an introductory secondary deNO_x means, designed and operated within practical limits of NO_x removal efficiency, ammonia slip, and coal sulfur level, high-dust SCR may continue to provide credible attractiveness and performance as one of the significant steps in achieving stringently low stack NO_x emission concentrations.

2.3. E Beam process attractiveness as relates to coal type fueling the boiler

Operation of boilers in Europe with common lignite firing can be optimized via combustion related measures to maintain NO_x emission levels as low as 100 ppm…as was found in the case of western Germany in the carrying out of the retrofit requirements of its "GFAVO" legislation in the late 1980s. Thus, in some/many instances, in applications with lignite and other low rank coals (where NO_x emission level may already conform to the regulatory limit) competitive attractiveness of the E Beam Process is diminished due to lack of need/demand for the measure of routine NO_x removal it would be expected to provide. The E Beam Process may be an even more evident misapplication in instances where, as in some U.S. services, low fuel sulfur level results, for example, in gross SO_2 concentration less than 1000 ppm, detracting from byproduct yield and net revenue as well as magnifying electron beam power requirement for a prescribed degree, if any, of NO_x removal.

2.4. Oxidation augmentation by chemical oxidant

Particularly in the case of low sulfur service, (wherein the power consumption for a given deNO_x performance is substantially greater than in high-sulfur service), E Beam acceptability may be significantly improved by an optimal degree of pre-oxidation of one or more pollutants by chemical oxidation. See Section 3.3 and 3.4.

2.3. Safety considerations in siting of ammonia storage

In the U.S. and, no doubt, in many other countries of interest, permitting of required high-capacity ammonia supply storage may not be possible at many plant sites. Consideration of public safety may limit E Beam Process applications to comparatively remote powerplant sites in areas of low population density or to merchant generating units to be built at industrial complexes, at major chemical plants or at petroleum coke generating oil refineries, etc. Recent experience in the U.S. speaks to lack of adequate safety in the large-volume storage of ammonia, be it either aqua ammonia or anhydrous ammonia. Wet ammonia scrubbing systems are not being commonly installed. And in many of the numerous utility size SCR installations in recent years permitting has been denied for ammonia storage capacity substantially less than that which would be required for wet ammonia scrubbing or an E Beam Process application of equivalent megawatt capacity. SCR system designs are, instead, frequently based on supply and storage of benign urea solids from which ammonia feed is generated on site and on demand.

3. ADVANTAGES/DISADVANTAGES OF CHEMICAL OXIDIZING TECHNOLOGIES FOR SIMULTANEOUS NO_X REMOVAL AT COAL PLANTS

The most cost-effective chemical oxidants in gaining desired NO/NO_2 and elemental mercury pre-oxidation upstream of scrubbers are seen to also bring about SO_2 oxidation to SO_3 to some degree. Moreover, most US scrubbers are wet type, which have poor efficiency in removing this acid gas. Thus the most promising opportunities for simultaneous removal of SO_2/NO_x and mercury together with SO_3 can be seen to occur with gas cleaning in a dry mode.

3.1. Overview of SO_3 as an operating problem

Sometimes only a minor aspect of sulfur oxides emission, discharge of SO_3 has historically not been a major pollutant in coal firing. In some high sulfur applications, however, it has become a regulatory, stack discharge problem e.g. when stack plume opacity exceeds 10-20%. In wet FGD systems much of the SO_3 and H_2SO_4(v) is converted to H_2SO_4(l), particulate, i.e. visible sulfuric acid mist, an aerosol. Moreover, SO_3 originating in the boiler or brought about by SCR operation can create problems in air preheater operation. Tactically, SO_3/H_2SO_4(v) is most appropriately abated by an adsorption step upstream of the air preheater. But operation of dry FGD will, in all applications, ensure against significant stack opacity caused by acid mist since it will absorb SO_3 more efficiently than SO_2. (An extremely high, prohibitive level of costly flue gas reheat would be necessary, alternatively, in the case of wet scrubbing to vaporize an otherwise significant acid mist concentration).

3.2. Preference for dry process flue gas cleaning

Wet scrubbers cool raw flue gas too rapidly to efficiently remove SO_3 gas and vapor-phase H_2SO_4, i.e. H_2SO_4 (v), the gaseous, molecular form of sulfuric acid created from SO_3 via upstream flue gas cooling, principally that taking place in the air preheater. But, dry FGD use, in and of itself, provides a well-established basis for highly efficient removal of SO_3. By its avoidance of very rapid cooling in its only partial humidification of the flue gas, dry scrubbing provides means of efficiently capturing SO_3 (rather than converting a major portion of it, as do wet scrubbers to a sulfuric-acid-mist aerosol discharged from the stack.) Moreover, except in cold winter operation, dry operation avoids a sometimes objectionable, visible, steam plume at the stack exit.

3.3. Emerging secondary $DeNO_x$ technology utilizing existing scrubbers

Major technological advancements in process design and collection performance of commercial SO_2 removal systems in the last 10+ years provides an advantageous basis for incorporating removal of NO_x therein, simultaneously with the other pollutant species of concern. The several very promising but, to date, not extensively applied such NO_x removal techniques, essentially all of which are tied to need of pre-oxidation of flue gas NO, presently continue to be promoted and examined (for use in wet or dry collection operation [10-12]. Clearly there are major opportunities in the U.S. in the face of pyramiding, restrictive, NO_x regulations for advantageous application of simultaneous, multi-pollutant removal, especially in dry mode operation. Moreover, cost effectiveness of upstream $DeNO_x$ means simpler than SCR can best be realized by employing downstream dry-scrubbing-type chemical process technology that achieves broad simultaneous removal, (e.g. of SO_2, SO_3 and NO_x).

Greatest synergism in performance is achieved in such trim-type $deNO_x$ by flue gas treatment when carried out by dry process means: strictest stack emission limits can be met for PM-2.5 as well as NO_x, Hg, SO_2, SO_3, etc. Stack NO_x emission control may be required to as low as 6 ppm, (0.01 lb/MM Btu.)

3.4. Versatility of pre-oxidation system design

In the case of E Beam Process use, and depending on characteristics of the commercial chemical oxidant chosen to supplement electron accelerator oxidation, low cost, low dosage pre-oxidation may (by choice) preferentially oxidize SO_2 (to SO_3) or, instead, oxidize NO (to NO_2 and then to highly soluble N_2O_5.). With SO_2 entering the E Beam reactor in the form of SO_3 and H_2SO_4(v) instead of SO_2, system performance would be benefited by the opportunity to optimally operate at a higher temperature than “normal” and with an anticipated improvement of cost effectiveness in removal of NO_x.

On the other hand, with pre-oxidation that is instead accompanied by entry of NO_x to the E Beam reactor as N_2O_5 (or a mixture of NO_2 and N_2O_5). E Beam Process design would instead be optimized, temperature and humidity wise, to most cost effectively remove SO_2 (and SO_3).

3.5. Important substitutional or supplementary role of E Beam process serving as the final $DeNO_x$ step

3.5.1. Particulate abatement

In its dry mode operation and serving as the final gas cleaning step with simultaneous removal of SO_2, the E Beam Process provides highly efficient, best available control of stack particulate emission currently required in the U.S. and other countries. This would be particularly so in so far as the Process' flexibility in substituting fabric filtration for electrostatic precipitation. In the 1970s, the U.S. New Source Performance Standard (NSPS), for total particulate in coal boiler flue gas dedusting, tied to common use of electrostatic precipitators, was upgraded from 0.10 lb/MM Btu of boiler heat input, (125 mg/m^3), to 0.03 lb/MM Btu, (37 mg/m^3). This brought about a major reduction in discharge of comparatively coarse particulate by newly built generating units. However, more stringent, recently established legislation to lessen ambient concentration of PM-2.5 (particulate matter of 2.5 micron size or less) now serves as the key criterion for measurement and regulation of particulate in ambient air. Thus, the early, total particulate basis, above, for designating dedusting performance (seen as lenient at this point in time) can be seen to be outdated. Particulate control strategy is further diversified/complicated by the knowledge that PM-2.5 in ambient air is principally comprised of, sulfates and nitrates i.e. atmospheric products of reaction with oxygen, ambient ammonia pollutant and water vapor of stack emissions of precursors, SO_2 and NO_x. Direct PM-2.5 discharge from stacks and other sources is only one constituent of the ground-level atmospheric mix, Ellison, W (1999), but its removal is a difficult challenge in attempts to use dry electrostatic precipitator technology. In now common use of downstream fabric filtration in conjunction with commercial dry scrubbing installations of all kinds, emission of fine particulate is minimized.

3.5.2. Advantages of fabric filtration

Such particulate control capability becomes of critical value in expanding the choice of upstream primary (combustion-related) deNOx means such as economical deNOx by coal reburn. This technology is disadvantageous in its generation of carbonaceous particulate. But it becomes highly attractive, however, given the high fine particle, filtration efficiency in now common application of fabric filters for dedusting. Use of such collectors has resulted in stack emission as low as an order of magnitude below the earlier noted, 0.03 lb/MM Btu, (37 mg/Nm3), particulate emission standard. Apart from any possible upstream, i.e. primary, use of such dedusting equipment for fly ash removal, the increasingly common incorporation of fabric filtration in dry scrubbing system designs ensures achievement of low, complying, stack emission of fine particulate.

4. SUPERIORITY OF HYBRID E BEAM/CHEMICAL OXIDANT DESIGN IN OPTIMIZING EB PROCESS USE FOR COST EFFECTIVE $SO_2/NO_X/HG^O$ REMOVAL

In bituminous coal NO_x concentrations are substantial as compared to those in low-rank coal service because of comparatively low fuel moisture content and resulting high flame temperature. In such instances an optimal manner, means and extent of pre-oxidation of raw flue gas can provide advantages in application of the E Beam process for simultaneous multi-pollutant removal by:

- Decreasing overall capital and operating cost
- Decreasing parasitic power consumption.

4.1. Low-sulphur service

The E Beam Process is at a substantial disadvantage in common low-sulfur coal service in diverse ways:

- Accelerator design capacity and cost together with parasitic electric energy consumption are greater than that required for simultaneous SO_2/NO_x operation in high-sulfur service in achieving the same degree of NO_x removal.
- Parasitic energy consumption may be uneconomically high conventional combined SO_2/NO_x removal nominally consumes 2% of the unit generating capacity, 1.6% for FGD plus 0.4% for SCR.
- High-value byproduct yield and net revenue from its sale/use are less than for a comparable installation in high sulfur service.
- Process economics may thus not be superior to that of other simultaneous or combined methods of multi-pollutant removal.

4.2. Effect of flue gas pre-oxidation

Augmental chemical oxidation upstream of the E Beam Process can be carried out to enhance economics in such applications in four ways:

- Reducing overall parasitic electric energy consumption to a level deemed generally acceptable by industry judgment, e.g. 2% of unit generating capacity
- Reducing capital cost and capacity for required accelerator facilities
- Gaining improved cost-effectiveness at a newly optimal reactor operating temperature
- Reducing total cost per ton of pollutant removal.

5. REMOVAL OF GASBORNE MERCURY AND MANAGEMENT OF MERCURY THEREBY COLLECTED

5.1. Mercury abatement in the U.S.

5.1.1. Overview

Following extensive study in late 2000, EPA announced its decision that mercury emissions from powerplants pose significant public health hazards and that regulation is appropriate and necessary under Section 112 of the 1990 Clean Air Act Amendments. It initially foresaw promulgation of final Hg control rules by December, 2004, with full compliance expected by a 2007 target date. These deadlines have now been extended. In view of imminent need for nationwide control of mercury emissions from coal, diverse technical approaches including the following have been developed:

- Physical cleaning of coal before supply to the boiler
- Converting the gasborne mercury to a solid phase
- Conversion of difficult to collect, elemental mercury into an oxidized, water-soluble form for ease of collection by scrubbing, (dry or wet)
- Adsorption of mercury in elemental form on finely divided, gasborne solids such as carbon, (including activated carbon additive).

5.1.2. Effectiveness of removal in scrubbing systems

As broadly reported, collection of mercury in FGD systems can be maximized by increasing the gasborne $Hg(II)/Hg^o$ concentration-ratio as occurs in the NO pre-oxidation step that is invariably required in flue gas treatment for simultaneous SO_2/NO_x removal. Ability to oxidize Hg^o, as well as attendant cost effectiveness and other considerations, are factors in choice of optimum flue gas NO oxidation means, e.g. from among chemical oxidants such as ozone, chlorine dioxide, ethylene oxide, hydrogen peroxide, etc.

5.2. Mercury control controversy in the U.S.A.

Two cognizant Illinois state government scientists have recently claimed[13] that most of the mercury found in the environment is natural and not of anthropogenic origin. This finding contradicts controversial pending Federal regulations designed to reduce environmental mercury by cutting powerplant emissions. In the background of this new finding is long standing doubt that coal fired powerplant emissions are the leading, or a significant cause, of mercury in fish there is no correlation between powerplant locations and high mercury levels. Against this evidence, the proponents of mercury control, including the U.S. Environmental Protection Agency, contend that mercury circulates nationally (and globally) via a process of general atmospheric deposition. The authors have newly tested this theory by comparing the estimated anthropogenic emission levels against measured levels of mercury in Illinois, U.S., and world soils. The conclusion they offer is that human emissions cannot possibly explain the observed amounts of mercury and that there must exist a natural global mercury flux that is significantly larger than human emissions. Much of it may be water borne or airborne. If so, reducing the estimated 48 annual tons of mercury emitted by U.S. coal fired powerplants might have little or no effect on environmental mercury levels. While it has been reported that most mercury in USA soils is the result of atmospheric Hg deposition and is mostly from anthropogenic sources, the authors have compared the rates of atmospheric Hg deposition to amounts of Hg in Illinois and USA soils. The amounts of Hg in these soils are too great to be attributed principally to anthropogenic Hg deposition. Thus, the scientific foundation for regulating mercury emissions from coal-fired powerplants is significantly challenged.

The Electric Power Research Institute, Palo Alto, California, the research and development organization of the U.S. electric generating industry has in recent days, explained at the MEGA Air Pollution control Conference in Washington, DC, (August 30 to September 2, 2004), that 70% of U.S. mercury emissions impact other countries, not the U.S., impact on the U.S. being primarily from Asian emissions.

5.3. Management of collected mercury

A program in the U.S. for control of mercury emissions will lead to transfer of such toxic waste to residues formed within gas cleaning systems, particularly in the SO_2 removal step. Throwaway solid and liquid wastes from SO_2 scrubbing systems may warrant environmental scrutiny and be seen as waste management problems. On the other hand, commercial experience with production and use of powerplant byproduct ammonium sulfate akin to that from the E Beam Process indicates that the level of mercury is less than that in conventional NPKS, (nitrogen-phosphorus-potassium-sulfur), fertilizer and thus not a new or increased environmental impact or concern.

6. PERMITTABLE AMMONIA-REAGENT STORAGE AND USE FOR MULTI-POLLUTANT CONTROL

Governmental authorities in the U.S. consider, by all comparisons, that the amount of ammonia storage required for an electric utility scale SO_2 removal system is huge and a major public safety consideration. Progress in common, broad application of the E Beam Process will depend on innovative sitting strategies. Prime sales prospects among utility boiler owners include those that:

- Are located in close proximity to available existing chemical-industry ammonia storage facilities thereby affording secure system operation with but minimal local ammonia storage at the boiler site
- Are in isolated areas remote from a local human population
- Are sited in highly industrialized localities in which parallel safety risks are pre-existing
- Are severely space constrained, locally and regionally, so far as achieving ultimate disposal by throwaway of low-value scrubber wastes, solid and liquid

- Are more than approximately 100 miles from an existing gypsum wallboard plant that might otherwise seem to be seen a candidate for large-capacity use of powerplant FGD gypsum from such a potential FGD process
- Plan new coal fired generating facilities to be sited at brownfield sites that are conducive to large scale ammonia storage
- Plan new coal fired generating facilities to be advantageously erected within available plot space at industrial complexes, e.g. a merchant generating facility that can be sited at a petroleum refinery and be fueled by petroleum coke and other refinery waste fuels.

7. TRENDS IN ACHIEVING POWER PLANT BY- PRODUCT GENERATION OF HIGH VALUE

7.1. Substantial net revenue from byproduct generation

Sulphurous byproduct net revenue in high sulfur coal service via generation of a salable yield of ammonium sulfate is a compelling factor in assuring economic feasibility. With bituminous coal sulfur level above approximately 3% no other generally accepted FGD means is as cost effective [12]. Moreover, the high value of this byproduct (in relation to the cost of the ammonia from which it is formed) is seen to be assured in the long term in light of the growing soil sulfur shortage, e.g. in Asia, North America and other regions. The distinct pattern of substantial and increasing soil sulfur shortage in world agricultural operations is due to two principal factors:

- Major technological advancements and chemical industry gains over the late 20^{th} century wherein the proportion of so-called free sulfur (sulfate) in production and supply of NPK fertilizer has been sharply decreased
- Programs, especially those in North America and in Western Europe, beginning in the 1970s for dramatic reduction in national SO_2 emission inventories.

7.2. Emerging market trends

As ammonia FGD and SO_2/NO_x process sales progress in the U.S. [2], major chemical fertilizer firms will play a significant role in cooperation with major flue gas cleaning system supply firms. Specific examples as follows point to the need (in fullest exploitation of the E Beam Process) for the dedicated effort of a major U.S.- based gas cleaning company strategically allied with a principal NPKS fertilizer firm:

- Marsulex (Canada) in its ownership of Marsulex Environmental technological leader in wet ammonia scrubbing technology
- Potash Corporation of Saskatchewan in its management, sale and use of diverse FGD-formed fertilizer blending stacks generated via introduction of a new regenerative sodium scrubbing technology.

7.3. Potential major U.S. players in large-scale marketing of the E Beam process

Based on the nature and complexity of large-scale detailed engineering, this allied with conceptual engineering design by the E Beam technology specialist, a large established U.S. stack gas cleaning company is needed to adequately fulfill the substantial opportunity for its marketing. Aspiring wet ammonia scrubbing system suppliers like Wheelabrator, Alstom, Foster Wheeler and others are logical candidate licensees to carry out such a program. Essential cooperative support in byproduct marketing might well be fulfilled, for example, by Hydro Agri, formerly Norsk Hydro, or any other major chemical fertilizer producer not otherwise affiliated with gas cleaning firms.

8. CONCLUSIONS

8.1. Electron Beam process advantages

Major competitive advantages of the E Beam Process and its simultaneous removal of diverse pollutants in the large and growing market for application of retrofit multi-pollutant control in the U.S. are:

- Unique inherent design capability for simultaneous removal of SO_2 and NO_x, proven in commercial service, and of Hg
- Dry mode operation and its many benefits in comparison to all wet scrubbing processes
- Resulting simplicity of retrofit installation, requiring no replacement of existing stack or breeching
- Absence of solid or liquid throwaway waste disposal and/or of need to manage low-value byproduct generation
- Yield of high-value ammoniated byproduct incorporating all captured acid gases, including hydrogen chloride originating from the coal
- Minimal water consumption, energy requirement and complexity in agglomerating the byproduct (originating as a dry powdered solid yield) for its practical commercial use
- Major net revenue from finished byproduct output in high-sulfur fuel service
- Large and growing market for sale/supply of the byproduct in its large scale use as a sulfur/nitrogen blending stock in manufacture/distribution of NPKS fertilizer products by chemical fertilizer companies
- Greatest cost-effectiveness that can be achieved in control of SO_x and NO_x emissions when applied in medium or high-sulfur service.

8.2. Electron Beam process marketing guidelines

Potential disadvantages or limitations of the E Beam Process calling for technology upgrading or judiciousness in its marketing are:

- Need to gain regulatory permitting of ammonia-supply storage of large capacity or make practical use of existing facilities for ammonia storage, a portion of which is able to be dedicated to E Beam Process use
- Need for special, formal contractual relationships (with U.S. based entities) required in its broad penetration of the U.S. market (these apart from successful sales contracts with boiler/utility owners):
    - Satisfactory and financially advantageous licensing by the E Beam Process technology supplier to a major existing stack gas cleaning system supply firm able to adequately fulfill responsibility for marketing, detailed engineering (based on conceptual design engineering by the technology supplier) as well as installation of the E Beam system
    - Agreement with a suitable major chemical fertilizer manufacturer and marketer of adequate size and effectiveness to take custody of and manage marketing/use of byproduct yield from E Beam Process operations solely
- Need to gain process design innovation to achieve economic feasibility in common low-sulfur coal applications in the face of (and to counter so far as possible):
    - Limited amount of byproduct yield and net revenue
    - Prospect of inherently high, comparative, parasitic electric energy use detracting excessively from the net electric power capacity of the generating unit served
    - Inherently high accelerator capacity and capital and operating cost.

- Cost to the E Beam technology supplier of technical development activity including field pilot-plant testing and demonstration to gain process design upgrading as above, e.g. by establishing a generic or proprietary conceptual design of an optimal means and choice of available chemicals to treat the dedusted raw flue gas by gas phase chemical oxidation so as to cost-effectively and substantially carry out a portion of the flue gas oxidation duty normally fulfilled only by the electron accelerators.
- Substantial nullification of the significance and inherent cost effectiveness of NO_x removal capability of the E Beam Process in the case of low-rank coal, i.e. lignite/brown coal applications, this because of the inherently low NO_x emissions from such fueling, (which, because of high chemically-bound water content, occurs at uniquely low flame temperature.
- Process design reorientation to use of fabric filtration in the final byproduct collection step to gain improved particulate collection efficiency in fully exploiting the dry particulate removal mode of the Process.

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VIEW TO SO_2-EMISSIONS IN THE BALTIC STATES THROUGH REORGANIZATION OF ENERGY INFRASTRUCTURE

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Abstract

The paper deals with predicting sulphur dioxide emissions generated by power production sector in the Baltic States in period up to year 2020. The economies of Lithuania, Latvia and Estonia are rapidly growing therefore forecast of emissions related with this occurrence becomes very important. The Ignalina nuclear power plant (INPP), one of the largest in the world, is situated in the region. Two power production scenarios are modelled to investigate changes in power sector's emissions expected as the consequences of the coming closure of Ignalina NPP. Power market was assumed to be common for all three Baltic countries and was modelled by applying the Balmorel model. The planned closure of Ignalina NPP will bring restructuring of Lithuania power production sector and will change also power transmission between countries. Predictive identified the potential of investments for new modern power generation technologies. At the same time modelling results show in both scenarios SO_2 emissions from power production in the Baltic region will increase. The increment of emissions is discussed in the context of meeting requirements of EC Directives. The SO_2 formation in Lithuania power sector may exceed the limits of the EU Council Directive 2001/80/EB therefore the additional measures to control SO_2 emissions have to be investigated.

1. INTRODUCTION

The change of energy infrastructure is an ordinary occurrence to be investigated by historical, technical and ecological aspects. In course of time the economical activities characteristic of a separate region, grow more or less intensive, become numb, fail or transform to another activity. In the course of history these changes are determined both by natural social-technical evolution and by revolutionary upheavals.

Every change of infrastructure is attended by changes characterized from the point of view of environmental quality evaluation. We can find a lot of unique examples both in the past and at present. These occurrences in the post communist countries including the countries of the Eastern Shores of the Baltic Sea are taking place rather intensively and, we suppose, it will still take place for a long time. Therefore the prognosis of environmental quality related with these occurrences become very important.

After re-establishment of the independence of three Baltic States (Lithuania, Latvia and Estonia) in 1991, the operation of their electric power systems was mainly influenced and determined by a decrease in electricity consumption and existing power generating overcapacity in this region. Therefore no new large scale power plants are developed in the Baltic States in the period after 1991 and thus no significant changes in generating capacities has taken place in this period. The total installed capacity of the Baltic integrated power system is 11000 MW which includes different types of technologies: the nuclear power plant in Ignalina (Lithuania), hydro power plants, condensing power plants, combined heat-power (CHP) plants and a pumped storage power plant, and wind energy is penetrating step-by-step as well. The largest part of this capacity - 55,5% - is installed in Lithuania followed by 24,5% in Estonia and 20% in Latvia. Each of three Baltic States is characterised by different dominating types of power generating capacities (see Figure 1). The next coming years may be characterised as to be crucial turning points for the re-organization of Baltic power structure. It will continue the reconstruction of Estonian power sector, the reconstruction of two Latvian CHP plants is planned.

However the most important consequences will be brought by the closing the Ignalina NPP and potential replacement of it by other power production alternatives. Thus Ignalina NPP is a key-element

for the re-organization of power generation in the Baltic states. Since the closing time of Ignalina NPP approaches the question is: what will be the ecological consequences of this step?

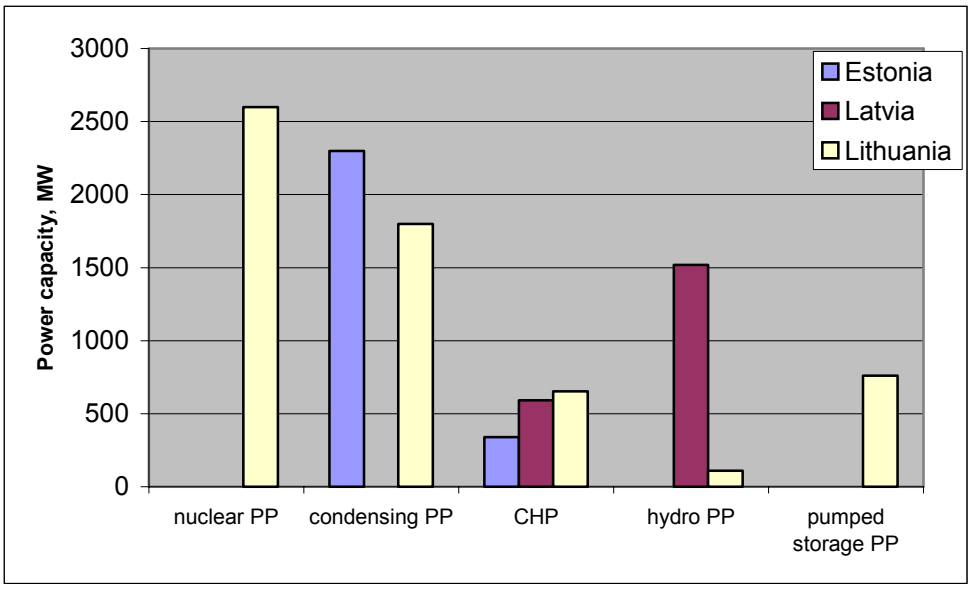

FIG. 1. Installed power capacity in the Baltic states

Ignalina Nuclear Power Plant is one of the largest in the world. Two Russian atomic reactors RBMK-1500 (the thermal power output is 4800 MW, nominal electrical power capacity is 1500 MW, available capacity 1300 MW each) are functioning at present. Over the period of the last five years it generated 80-85% of the total electricity in Lithuania, but in the whole Baltic region this share constitutes approximately half (e.g., in year 2002 Ignalina NPP produced 14143 GWh of power which constituted 47% of total gross power production in three Baltic countries). Due to large capacity of Ignalina NPP the total installed electricity generation capacity in Lithuania twice exceeds the present domestic demand. According to State Nuclear Power Safety Inspectorate (VATESI) in 2002, 12897 GWh was sold to "AB Lietuvos energija", of which 47.5% was consumed in the domestic market, and 52.5% was exported.

When the service time of nuclear fuel channels expires the exploitation of reactors will cease. Evidently, the future development of the whole energy sector is greatly influenced by the choice of operating lifetime for the two units at the Ignalina NPP. There are two possible scenarios for INPP future investigated in the current paper:

1. Scenario 1 - closure date of Block 1 is 2005, closure date of Block 2 is 2011
2. Scenario 2 - closure date of Block 1 is 2005, exploitation of Block 2 is extended at least until 2020 [1].

There is a lot of official debate about the construction of a new modern nuclear reactor at Ignalina. However, the closing of INPP may have negative environmental as well as other consequences. Thus, it is of great importance to evaluate the impact of Ignalina NPP closure on the possibilities of Lithuania and other Baltic states to comply with obligations under international conventions on climate protection and air pollution– i.e. the UNFCC and the UN EEC Geneva convention. At present, Baltic States meet all requirements of international conventions on air pollution and their protocols were ratified in the last years.

However, by 2005 increasing emissions from power generation as a result of the closure of unit 1 at Ignalina NPP will start to be a problem for Lithuania. For Estonia and Latvia the emissions

development is closely linked to the expected growth of electricity consumption. In order to balance demand of energy and environmental issues careful planning and immediate action is necessary. For this reason it is necessary to determine the increased atmospheric pollution levels when the both units of Ignalina NPP will be closed and new generating capacities will be installed according a least cost power sector development plan.

2. INITIAL DATA

Electricity production in Lithuania, Latvia and Estonia according to Scenario 1 is presented in Figure 2 and according to the Scenario 2 - in Figure 3 [2].

According to both power sector development scenarios a small drop (few percent) of electricity production in the Baltic states' region is expected after 2004 due to closing of the 1st unit of Ignalina NPP. This drop will not influence the meeting of growing electricity demand in the region but as a consequence power export to the countries outside region will be reduced. Growth of demand in the region will be met by the new power generation options and in years 2009/10 the power production will reach the level of year 2004 with further increase thereafter. In Scenario 1 due to abandonment of nuclear power in 2010, export of Lithuania electricity to the neighbour countries will be reduced significantly. In 2010 electricity production (scenario 1) in Lithuania will be about 10.2 TWh, in Estonia 9.28 TWh, in Latvia 4.68 TWh. After 2010 electricity production will rise due to the increasing demand.

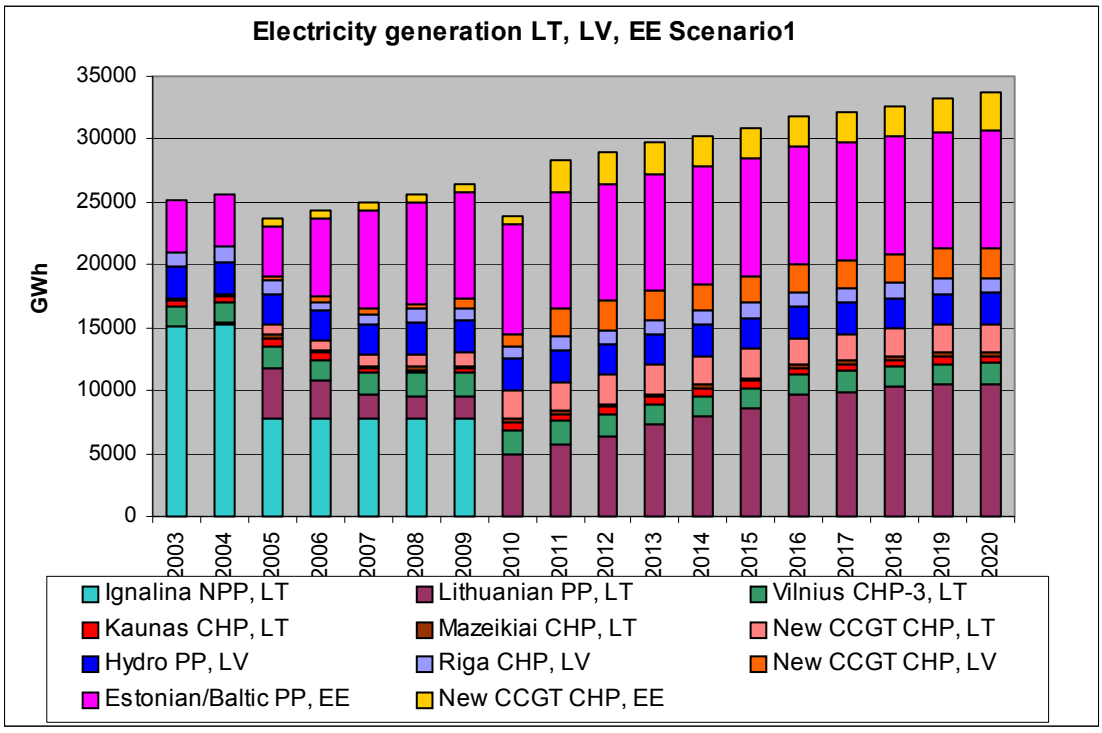

FIG. 2. Electricity generation in Lithuania, Latvia and Estonia according to Scenario 1

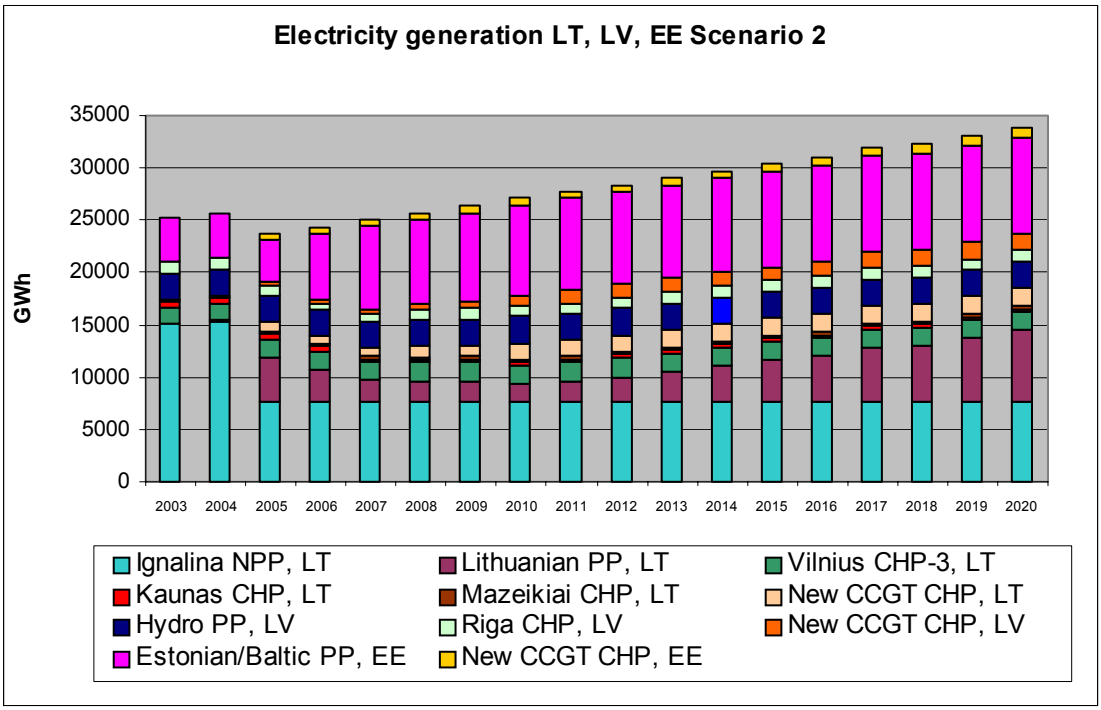

FIG. 3. Electricity generation in Lithuania, Latvia and Estonia according to Scenario 2

For SO_2 formation forecast we used formation (emission) factors taken mainly from literature [3]. Only for shale combustion in Estonian power plants we calculated necessary emission factors according to official information on annual emissions. The used SO_2 formation factors are presented in Table I.

TABLE I. SO_2 FORMATION (EMISSION) FACTORS, KT/PJ, FOR DIFFERENT FUEL TYPES

Fuel type	G_{SO2}
Natural gas	0
Heavy fuel oil	1,463
Heavy fuel oil 40%, natural gas 60%	0,5852
Orimulsion 40%, natural gas 60%	0,77
Shale	0,607

For all power plants and CHP the combination of heavy fuel oil 40% and natural gas fuel 60% was modelled. Only for Mazeikiai CHP is expected to use 100% heavy fuel oil, Lithuanian PP combination of orimulsion 40% and natural gas 60 and Estonian/Baltic PP 100% oil shale.

3. ANALYSIS DATA

3.1. SO_2 formation forecast in the Baltic Region after INPP closure according to Scenario 1

Figure 4 presents the SO_2 formation forecast in Lithuanian, Latvian and Estonian power sector according to Scenario 1

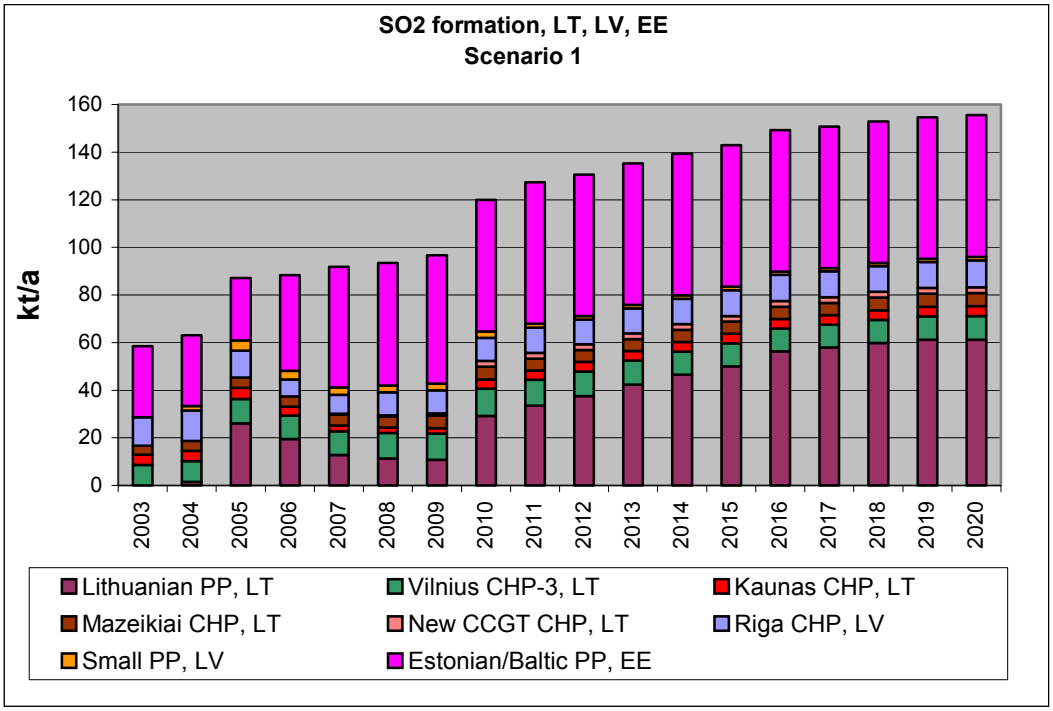

FIG. 4. SO_2 formation forecast in Lithuanian, Latvian and Estonian power sector according to Scenario 1

A SO_2 formation forecast in the power sector of all three Baltic countries shows (Figure 5) that the SO_2 emissions from the Lithuanian power sector will be from to Lithuanian PP, in Latvia – to Riga CHP, in Estonia – to Estonian/Baltic PP.

Modeled scenarios show, after closure of INPP Block 1 in 2005, the formed SO_2 amount in Lithuania will increase almost twice, comparing to the previous year (from 18,6 kt to 45,3 kt). After closure of INPP Block 2 in 2010 SO_2 amount in Lithuania will increase 73% comparing to level of 2009. Later, the SO_2 formation in Lithuania will increase 1.6 times in 2020 comparing to 2010 level and reach 83,2 kt. At the same time in Estonia between 2005 and 2010 formed SO_2 amount will increase twice but graduate. From 2011 this amount will be almost steady [4].

3.2. SO_2 formation forecast in the baltic region after INPP closure according to Scenario 2

After closure of the first INPP-unit, the formed SO2 amount in Lithuania will increase (Figure 5). From 2010 this amount will be almost steady, similarly to Scenario 1.

After closure of the first INPP unit the SO2 formation in Latvian power sector will decrease by use of fuel with lower sulphur content and further on will not fluctuate very much. The situation for Estonia would be not changed from 2009 year.

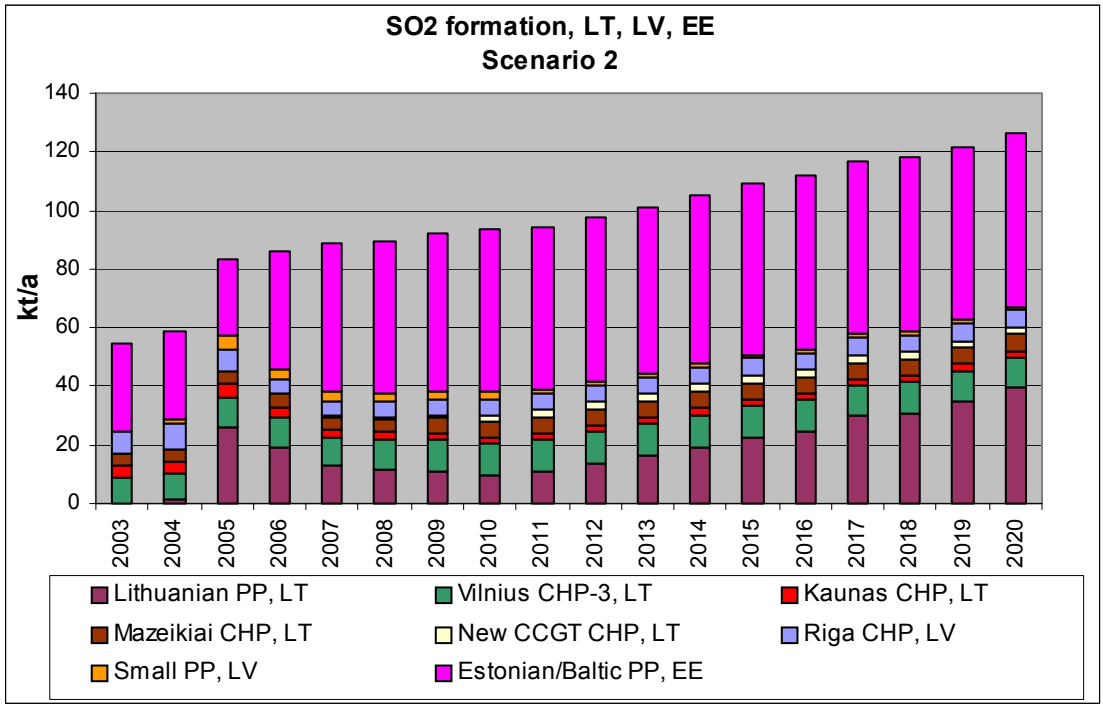

FIG. 5. SO_2 formation forecast in Lithuanian, Latvian and Estonian power sector according to Scenario 2

4. BALTIC STATES AND EU AGREEMENTS

In Annex IX of EU Treaty of Accession for Lithuania (item 10 "Environment" clause D "Industrial pollution control and risk management") stated, that SO_2 emission from Vilnius CHP-3, Kaunas CHP and Mazeikiai CHP could deviate from the requirements of EU Council Directive 2001/80/EB until the 31st of January 2015. The limits of SO_2 emission have to correspond to the following value [5]:

- 2005: 28,3 kt of SO_2 per year;
- 2008: 21,5 kt of SO_2 per year;
- 2010: 30,5 kt of SO_2 per year;
- 2012: 29 kt of SO_2 per year.

In case of Scenario 1 SO_2 emission will strongly exceed the limits. That means that only in case of Scenario 2 in 2010 value of SO_2 emission are close to obligations of the Council Directive 2001/80/EB (see Figure 6). Obviously, desulphurisation should be one of the means of development of environmental acceptable policy on the national and regional level.

Talking through Estonian power sector future engagements the main stress is that they need to achieve 65 % of desulphurisation during the transition period. Estonia struggles to reach a maximum of 25 kt of SO_2 emissions annually up to 2012 [6].

The total level of SO_2 emission from the large combustion plants, operation of which is started till November 27, 2003, is determined by the Latvia's Government (Regulations Nr.379), and is not to exceed 25 kt of SO_2 annually [7]. The modelling shows that this target can be fulfilled.

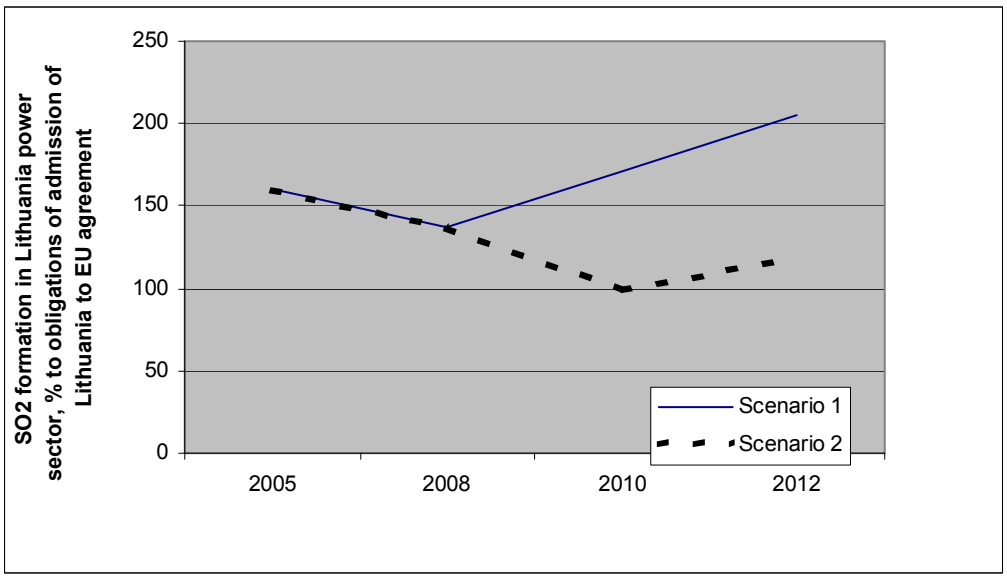

FIG. 6. The part of SO_2 formation according to Scenarios 1,2 by power sector of Lithuania in comparison with obligations of Annex IX of EU accession agreement (item 10 "Environment" clause D "Industrial pollution control and risk management") Lithuania

5. CONCLUSIONS

The impact of Ignalina NPP closure on the SO_2 emissions' development in the whole Baltic states region context is identified. According to two investigated energy sector development scenarios, SO_2 emissions will increase. Increase of emissions depends to a large extent on the operation of Lithuanian PP in the period after closure of one or both units of Ignalina NPP.

SO_2 formation in the region of the Baltic states according to Scenario 1 will change from 58 kt/a in 2003 to 155 kt/a in 2020, but according to Scenario 2 - to 126 kt/a in 2020.

In case of both scenarios SO_2 formation in Lithuanian power sector will exceed the limits of Council Directive 2001/80/EB for SO_2 emissions, therefore additional measures to control SO_2 emissions have to be developed and taken into use.

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ELECTRON BEAM TREATMENT OF TEXTILE DYEING WASTEWATER: OPERATION OF PILOT PLANT AND INDUSTRIAL PLANT CONSTRUCTION

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Abstract

A pilot plant for treating 1,000m^3/day of dyeing wastewater with e-beam has been constructed and operated since 1998 in Daegu, Korea together with the biological treatment facility. The wastewater from various stages of the existing purification process has been treated with electron beam in this plant. Installation of the EB pilot plant resulted in decolorizing and destructive oxidation of organic impurities in wastewater, in reduction of the treatment time, and in increase in flow rate limit of existing facilities by 30-40%. Industrial plant for treating 10000m^3/day each, based upon the pilot experimental result, is under construction and will be finished by 2005. This project is supported by the International Atomic Energy Agency (IAEA) and the Korean Government. A commercial plant for re-circulation of wastewater from Papermill Company is also designed for Pan Asia Paper Co. Cheongwon Mill, and after the successful installation, up to 80% of wastewater could be re-used in paper producing process. The method for the removal of heavy metals from wastewater and other technologies are developed with the joint works with Institute of Physical Chemistry (IPC) of Russian Academy of Sciences.

1. INTRODUCTION

Rapid population growth with industrialization, urbanization and water-intensive lifestyles is resulting in severe problems in wastewater management. In Korea, where the industries are concentrated in urban areas, resulting in severe water pollution problems in most large cities. Major sources of water pollution include chemical-intensive industries such as textiles, metal plating, electronics, papermill and refineries. And hence, the treatment of such industrial wastewater becomes a more important subject in the field of environment engineering.

The treatment of the industrial wastewater containing refractory pollutant with electron beam is actively studied in EB-TECH Co. Electron beam treatment of wastewater leads to their purification from various pollutants. It is caused by the decomposition of pollutants as a result of their reactions with highly reactive species formed from water radiolysis: hydrated electron, OH free radical and H atom [1]. Sometimes, such as reactions are accompanied by other processes, and the synergistic effect upon the use of combined methods such as electron beam with biological treatment, adsorption and others improves the effect of electron beam treatment of the wastewater purification.

In the process of electron-beam treatment of wastewater there are utilized chemical transformations of pollutants induced by ionizing radiation. At sufficiently high absorbed doses these transformations can result in complete decomposition (removal) of the substance. Under real conditions, i.e., at rather high content of pollutants in a wastewater and economically acceptable doses, partial decomposition of pollutant takes place as well as transformations of pollutant molecules that result in improving subsequent purification stages, efficiency of the process being notably influenced by irradiation conditions and wastewater composition [2].

2. WASTEWATER FROM PAPER MILLS

A commercial plant for re-circulation of wastewater with electron beam from Papermill Company is also under planning in Pan Asia Paper Co. Cheongwon Mill and EB TECH Co.. Cheongwon Mill is located from 120 km south of Seoul, and consumes 18,000 m^3 of water per day.

The major products of this company are papers for newsprint (450t/day) and are mainly made of recycled paper (91%) and pulps. For the economical point of view, it is preferable to recycle the treated water to production lines, but now used only 20-30% at total water since the amount of organic impurities after treatment are high and some of them are accumulated during re-circulation.

Purification of wastewater is now performed by 2-stages of chemical and biological treatment facilities (Fig.1). The existing facility for purification of wastewater under consideration consists of the following main stages [3]:

1) Primary chemical coagulation + flocculation;
2) Biological treatment by activated sludge with subsequent sedimentation and filtration through sand filter
3) Secondary chemical coagulation (with the addition of hypochlorite)

The COD value after the first stage gives rise to decrease in COD value to around 150 ppm. The COD value after the third stage is 45-90 ppm. The COD value of finally purified wastewater should be less than 25 ppm. In order to develop the most efficient method for re-circulation of wastewater, the experiments were conducted with samples in various stages of treatment. In the experiments, electron accelerator of 1 MeV, 40 kW with the dose rate of 40 kGy/s is used. In order to carry out the experiments, the laboratory unit schematically shown in Fig. 1 was constructed for irradiation under flow conditions. The initial water is placed in storage vessel, which serves as saturator-equalizer. Air or ozone-air mixture with controlled flow rate up to 40 l/min was fed to the vessel. Wastewater from the vessel is moved with controlled consumption by pump to multi-jet nozzle. Diameter of each jet was equal to 4 min; it is equal to the range of 1 MeV electrons in water. The rate of wastewater moving at the exit of the nozzle was controlled within the range of 2-4 m/s (it corresponded to the rate of wastewater in the industrial plant under design). The wastewater injected directed in parallel each other in horizontal plane; their flight length was equal to ~1.5 m (at the initial rate 3m/s). The wastewater injected along horizontal part of their flight was treated by electron beam. Then irradiated wastewater was collected into the special container.

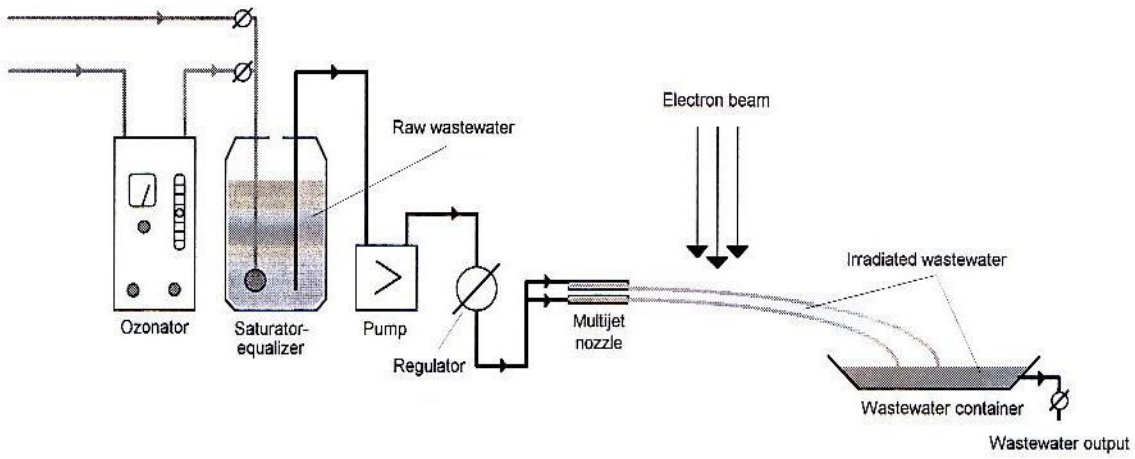

FIG. 1. Laboratory unit used in electron beam treatment experiments.

In order to develop the most efficient combined electron beam method for purification of the wastewater, the experiments were conducted initially with 4 various samples: initial raw wastewater, wastewater after primary coagulation, wastewater after biological treatment and filtration, and finally-purified wastewater. It is shown that the decrease in absorbance is the most for first and third samples.

Because of it the relative changes in COD, BOD_5, TOC and absorbance at 235 nm were measured for raw wastewater and wastewater after the second stage of purification as a result of electron beam treatment at various doses and subsequent coagulation + flocculation. The $Al_2(SO_4)_3$ solution was used as a coagulant. Sometimes the $Al_2(SO_4)_3$ + $Fe_2(SO_4)_3$ solution served as a coagulant ; in this case the better results were obtained. This effect is the most at doses < 3kGy. Note that a small increase in BOD_5 value was observed in initial raw wastewater at doses < 1kGy.

It was found that the positive influence of electron beam treatment is highest for wastewater after second stage of purification. The data obtained allowed to conclude that the most advantageous part of existing technological line for using electron beam treatment is after first coagulation + flocculation and biological treatment. Because of it the treatment of such a partially purified wastewater was studied in detail and under various conditions.

The values of COD_{Cr}, COD_{Mn}, TOC and color were measured. The results obtained are shown in Figure 2. In the figure, the following abbreviations were used: LFS - the treatment by $Fe_2(SO_4)_3$ coagulant and then by polyacrylamide, LAS - the treatment by $Al_2(SO_4)_3$ coagulant and then by polyacrylamide flocculant, LFAS - the treatment by mixed $Fe_2(SO_4)_3$ + $Al_2(SO_4)_3$ (mole ratio 1:1) coagulant and then by polyacryl amide flocculant, Electron beam treatment at maximum dose rate 40 kGy/s, dose 1.3 kGy and rate of water flow 3 m/s. In each figure, solid black line shows the mean value of the respective parameter for the initial wastewater (after primary coagulation + flocculation treatment and biological purification).

The decrease in the initial value of the parameter after any treatment is shown by vertical line. The sequence of treatments is given along vertical line. The vertical line is ended by an arrow which indicates the achieved value of the parameter as a result of the treatment. The best results is irradiation of water after biological treatment combined with coagulation and filtration. Irradiation in this stage, the additional removal of impurities is up to 80% in TOC (Total Organic Carbon) values.

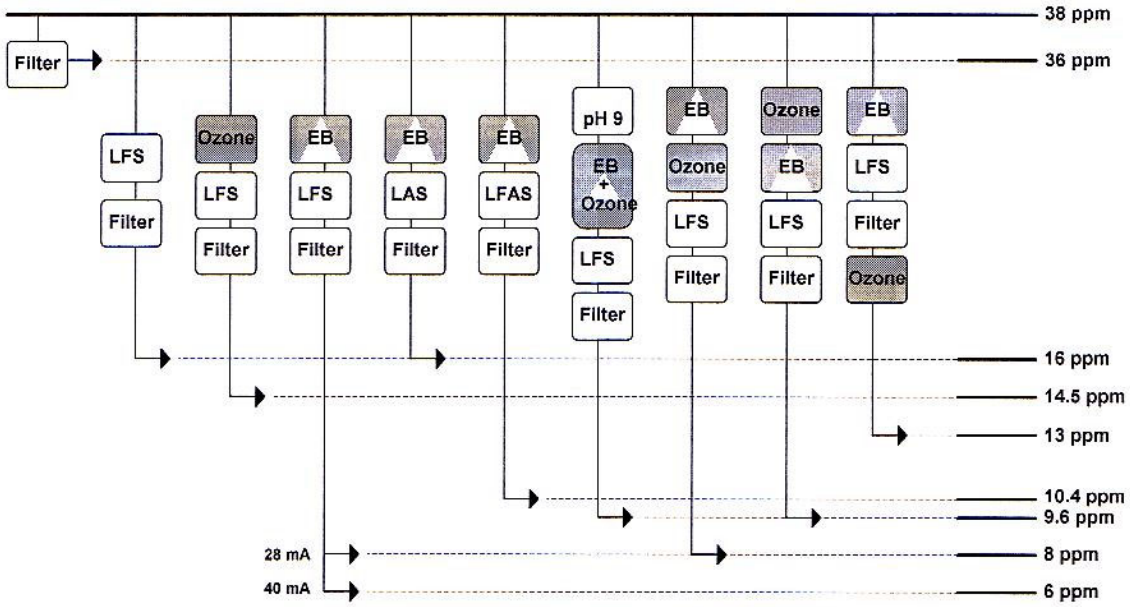

FIG. 2. TOC values of wastewater after various treatment

On the base of data obtained by EB-TECH Co. and IPC the suitable doses in this case are determined as around 1 kGy for the flow rate of 15,000 m^3 wastewater per day (since the 3,000m^3 of wastewater is returned to initial stage with sludge). Therefore, three accelerators with the total power of 300kW and treatment system are designed for,

- Decreasing the operation cost of wastewater treatment facility
- Improving the removal efficiency of organic impurities below 25 in COD
- Increasing the re-circulation rate up to 80%

Expected construction period includes 11 months in civil and installation works and 3 months for trial operation. After the successful installation of electron beam treatment facilities, up to 80% of wastewater could be re-used in paper producing process (Figure 3). Overall construction cost are estimated around $US 5 and operating cost for above plant are estimated as $US 0.5M per year.

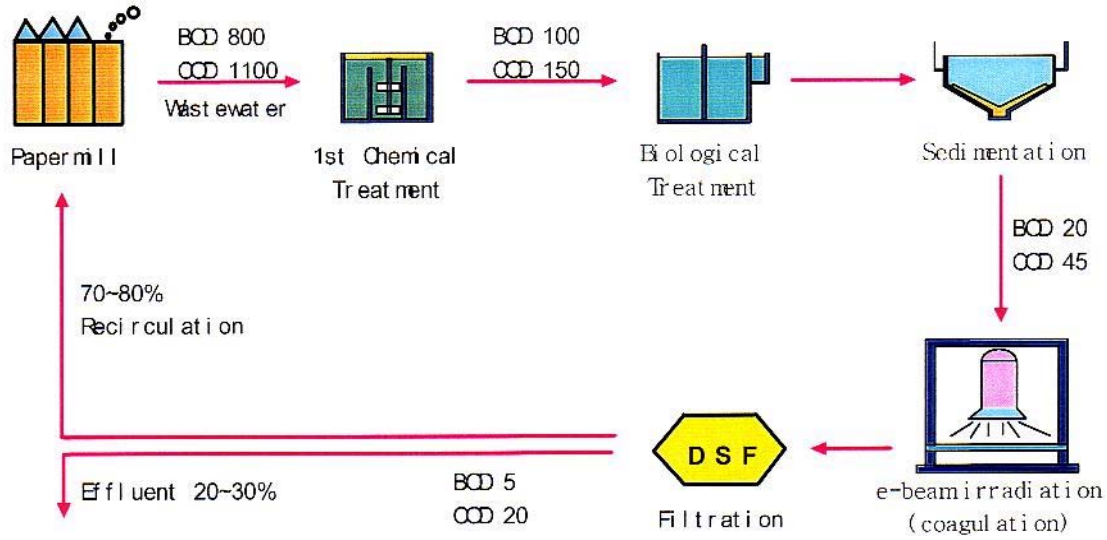

FIG. 3. Process flow of e-beam facility for wastewater from papermill.

3. WASTEWATER FROM TEXTILE DYEING COMPAINES

The complex wastewaters from textile dyeing companies in Daegu Dyeing Industrial Complex (DDIC) were investigated in this study. DDIC includes about hundred factories occupying the area of 600,000m^2 with 13,000 employees in total. A majority of the factories has equipment used for dip dyeing, printing, and yarn dyeing. The production requires high consumption of water (90,000m^3/day), steam, and electric power, being characterized by large amount of highly colored industrial wastewater. Purification of the wastewater is performed by union wastewater treatment facilities (chemical treatment and 2 steps of biological treatment). Current facility treats up to 80,000m^3 of wastewater per day, extracting thereby up to 500m^3 of sludge. Rather high cost of purification results from high contamination of water with various dyes and ultra-dispersed solids.

Characteristics of DDIC wastewater undergo both short-term and long-term variations, the former being equal to 10-13 % while the latter amounting up to 20 % of mean value. Overall characteristics of influent wastewater, 5 day's biological oxygen demand (BOD_5), chemical oxygen demand measured by permanganate method (COD_{Mn}), suspended solid (S/S) are presented in Table I.

TABLE I. TYPICAL CHARACTERISTICS OF TEXTILE DYEING WASTEWATER OF DDIC [4]

Parameter	pH	BOD_5(mg/l)	COD_{Mn}(mg/l)	S/S (mg/l)	Color units
Raw wastewater	12	2,000	900	100	1,000
Chemical treated	6.8~7.5	1,700	450	50	500
After 1st Bio-treat	7.0~8.0	1,300	250	50	400
After 2nd Bio-treat	7.0~8.0	50	80	50	250

Chemical composition of the wastewater also is not constant. Data of chemical analysis showed the composition of dissolved organic impurities in influent wastewater consist of organic compounds, organic dyes, surfactants and other organic compounds. In the organic compounds, terephtalic acid (TPA) and ethylene glycol (EG) are the major components of the pollutants. Organic dyes and surfactants, even at comparatively low concentration, determine such objectionable properties of the wastewater as color and foaming, so concentration of these compounds should be substantially reduced. Among other organic compounds there are: hexane, carboxy-methyl and hydroxyl-methyl cellulose, phenols, starch, waxes, etc. Inorganic compounds are presented mainly by sulfate anion and sodium cation (as result of pH adjustment) and small amounts of chlorides and carbonates. Besides, some amount of hydrogen peroxide may be present. The latter, unlike other inorganic compounds, can take part in radiation induced transformations of organic compounds.

Because of increase in productivity of factories and increased assortment of dyes and other chemicals, substantial necessity appears in re-equipment of purification facilities by application of efficient methods of wastewater treatment. The existing purification system is close to its limit ability in treatment of incoming wastewater.

3.1. Laboratory-scale feasibility study.

The laboratory scale studies had been carried out regarding the possibility of electron beam application for purification of wastewater. With the co-works of EB-TECH Co., Korea Dyeing Technology Center (DYETEC) and Institute of Physical Chemistry of Russian Academy of Sciences in Moscow, Russia (IPC), the experiments on irradiation of model dye solutions and real wastewater samples (from various stages of current treatment process) had been performed.

In order to develop the most efficient method for re-circulation of wastewater, the experiments were conducted with samples in various stages of treatment. In the experiments, electron accelerator of 1 MeV, 40 kW with the dose rate of 40 kGy/s is used. To carry out the experiments, the laboratory unit was constructed for irradiation under flow conditions. The initial water is placed in storage vessel, which serves as saturator-equalizer. Wastewater from the vessel was moved with controlled consumption by pump to multi-jet nozzle. Diameter of each jet was equal to 4 min; it is equal to the range of 1 MeV electrons in water. The rate of wastewater moving at the exit of the nozzle was controlled within the range of 2-4 m/s (it corresponded to the rate of wastewater in the industrial plant under design). The wastewater injected directed in parallel each other in horizontal plane; their flight length was equal to ~1.5 m (at the initial rate 3 m/s). The wastewater injected along horizontal part of their flight was treated by electron beam. Then irradiated wastewater was collected into the special container.

The results of laboratory investigations of representative sets of samples showed the application of electron beam treatment of wastewater to be perspective for its purification (Figure 4). The most significant improvements result in decolorizing and destructive oxidation of organic impurities in wastewater. Installation of the radiation treatment on the stage of chemical treatment or immediately before biological treatment may results in appreciable reduction of chemical reagent consumption, in reduction of the treatment time, and in increase in flow rate limit of existing facilities by 30-40%.

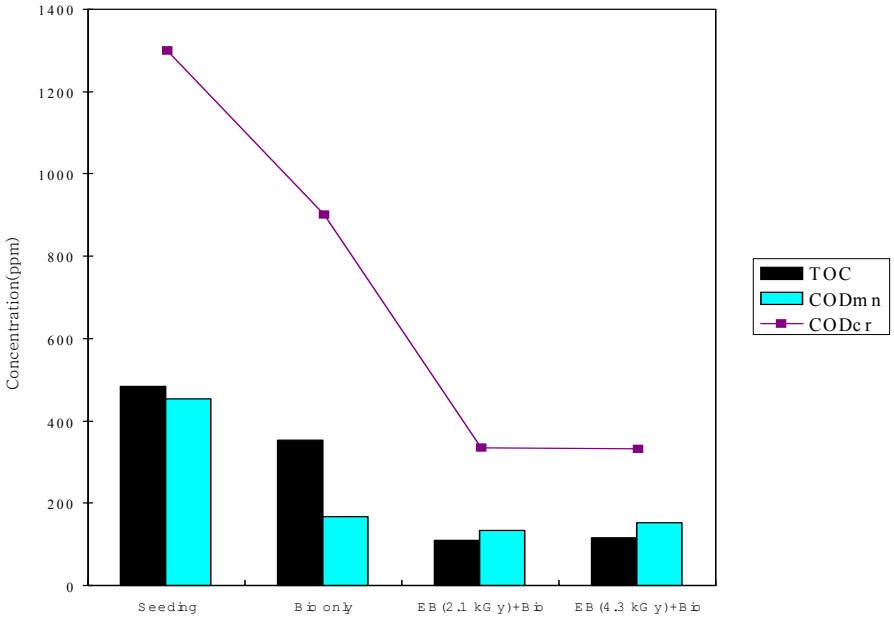

FIG. 4. Combined effect of e-beam and biological treatment.

3.2. Construction of pilot plant

Being convinced with the feasibility of laboratory scale tests, a pilot plant for a large-scale test (flow rate of 1,000m^3 per day) of wastewater has constructed and is now under operation with the electron accelerator of 1MeV, 40kW (Figure 5). The size of extraction window is 1500mm in width and Titanium foil is used for window material. The accelerator was installed in Feb. 1998 and the technical lines are finished in May 1998. For the uniform irradiation of water, nozzle type injector with the width of 1500 mm was introduced. The wastewater is injected under the e-beam irradiation area through the injector to obtain the adequate penetration depth. The speed of injection could be varied upon the dose and dose rate. Once the wastewater has passed under the irradiation area, then directly into the biological treatment system. The Tower Style Biological treatment facility (TSB) that could treat up to 1,000 m^3 per day has also installed in October 1998. TSB is composed of equalizer, neutralizer, and 6 steps of contact aeration media. Each aeration basin is filled with floating or fixed bio-media to increase the contact area.

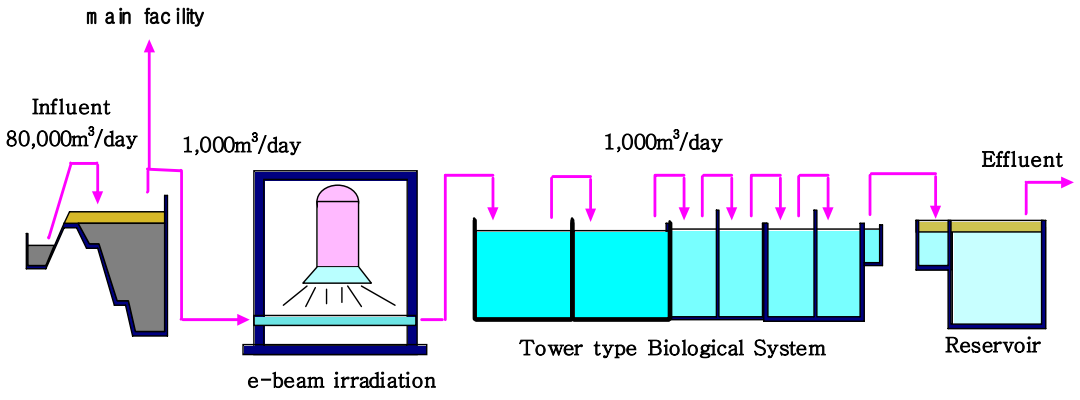

FIG. 5. Schematic diagram of Pilot Plant with e-beam

3.3. Result of pilot plant operation.

Pilot plant inlet flow is a mixture of two flows: raw wastewater from dyeing process and wastewater from polyester fiber production enriched with Terephtalic acid (TPA) and Ethylene glycol (EG); relative flow rate of the latter being 6-8% of total inlet flow rate. TPA concentration of influent is about $2 \cdot 10^{-2}$mol/l that is much higher than total concentration of all other dissolved pollutants. This concentration corresponds to electron fraction of TPA about 0.2% that makes direct action of radiation on TPA (or other pollutant) be negligible when treating the wastewater by electron beam. On the other hand, this concentration is high enough to prevent recombination of radical products of water radiolysis in the bulk of solution, taking into account high rate constants of reactions of both reducing (hydrated electrons, hydrogen atoms) and oxidizing (hydroxyl radicals) particles with terephtalate anion [5]:

- $e^-_{aq} + 1,4\text{-}C_6H_4(COO^-)_2 \longrightarrow R^1$ $\quad k = 7.3\times10^9$ l mol^{-1} s^{-1}
- $H + 1,4\text{-}C_6H_4(COO^-)_2 \longrightarrow R^2$ $\quad k \cong 1\times10^9$ l mol^{-1} s^{-1}
- $OH + 1,4\text{-}C_6H_4(COO^-)_2 \longrightarrow R^3$ $\quad k = 3.3\times10^9$ l mol^{-1} s^{-1}

Besides, because of high relative concentration of TPA comparing to other polluting compounds, competition between listed reactions and reactions of radical products from water with other compounds appears to be much in favor of the former ones. It follows from above mentioned that the main (if not the only) result of electron-beam treatment of pilot plant influent would be radiolytical transformations of TPA that can improve its removal by biological treatment. Radiolytical transformations of other initially present compounds, if those take place at all, can proceed via radical or molecular products from TPA.

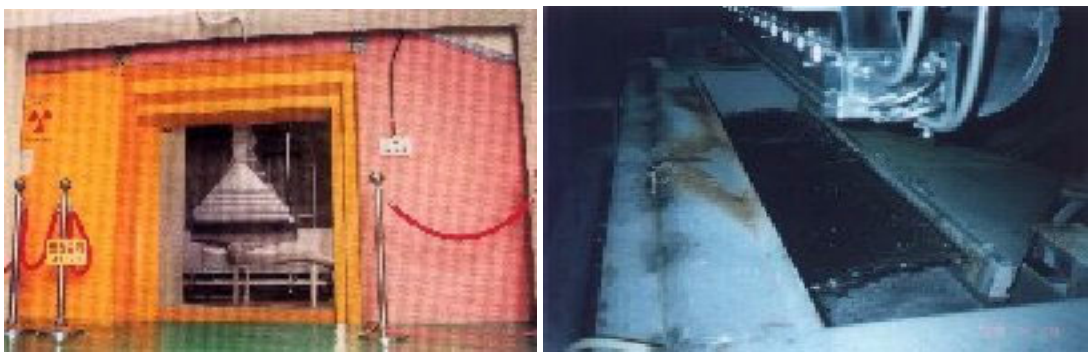

FIG. 6. Electron accelerator and wastewater under injection

Figure 7 shows that TPA enriched wastewater can be efficiently purified by biological treatment. However, preliminary electron-beam treatment improves the process, resulting in more significant decreasing TOC, COD_{Cr}, and BOD_5. As concerns changes in TOC, COD_{Cr}, and BOD_5 during biological treatment, from the data presented in Figure 7 it follows that preliminary electron-beam treatment make it possible to reduce bio-treatment time twice at the same degree of removal. Coincident results were obtained in a separate set of experiments on the same pilot plant but with reduced wastewater flow rate (~130 l/day). In this case inlet flow was divided into two flows: the first one passed only biological treatment while the second one passed electron-beam treatment, then biological treatment with reduced hydraulic retention time (HRT). Averaged for one month's period decrease in TOC values amounted 72%, for the first flow (48h HRT biotreatment), and 78%, for the second flow (1 kGy electron-beam treatment followed by 24 h HRT biotreatment).

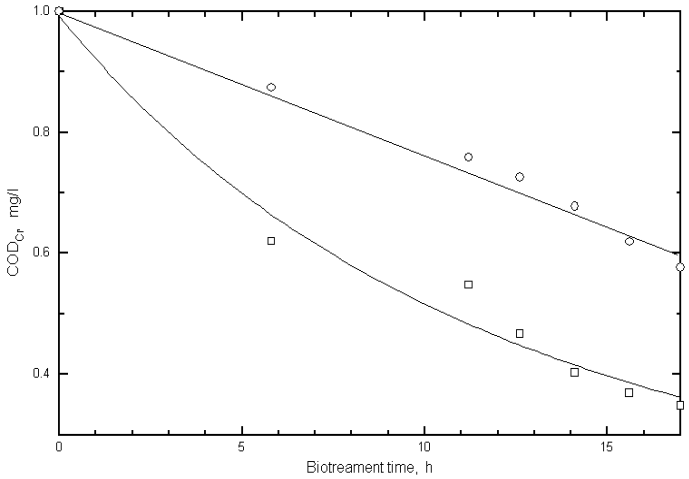

FIG. 7. Effect of electron-beam treatment on biological treatment of wastewater: kinetics of biotreatment of irradiated (1) and unirradiated (2) wastewater

Usually, increase in biodegradability after radiation treatment of aqueous-organic systems is due to radiolytical conversions of non-biodegradable compounds. In present experiments the improvement of biological treatment of wastewater after preliminary electron-beam treatment was found to be caused by radiolytical transformations of biodegradable compound. Electron-beam treatment should not appreciably affect total biodegradability of pollutants if the main pollutant is biodegradable, but can improve biodegradation process at initial stages. In other words, irradiation at comparatively low doses (several Grays) for this case does not change total amount of biodegradable substance characterized by BOD_5 , but convert part of it into easier digestible form. This is confirmed, also, by the data presented in Figure 4 where one can see that decrease in TOC, COD_{Cr}, and BOD_5 during biological treatment is close to linear one for non-irradiated wastewater, while for electron beam treated wastewater the decrease is faster at the beginning of biological treatment and decelerates during the process [6].

3.4. Construction of industrial plant.

On the evaluation of economies and efficiency of pilot scale electron beam treatment facility, industrial scale plant for treating textile dyeing wastewater is under construction from 2003 for :

- decreasing the amount of chemical reagent up to 50%
- improving the removal efficiency of harmful organic impurities by 30%
- decreasing the retention time in Bio-treatment facility

According to the data obtained in laboratory and pilot plant experiments with DDIC wastewater, the optimum absorbed dose for electron-beam treatment was chosen to be near 1 kGy. For that purpose 400 kW electron accelerators with three separate irradiators was proposed as a source of ionizing radiation. The plant is located on the area of existing wastewater treatment facility in DDIC and to have treatment capacity 10,000 cubic meters of wastewater a day using one 1MeV, 400kW accelerator, and combined with existing bio- treatment facility.

The process of wastewater treatment consists of the following steps [7]:

- collecting the inflow wastewater in primary (stock) basin;
- pumping the wastewater from primary basin to reactor;
- irradiating the wastewater inside reactor, cooling air being also irradiated;
- collecting irradiated wastewater in secondary basin;

- bubbling irradiated air (containing ozone) through the wastewater in a basin;
- pumping the wastewater from secondary basin to outlet line

Total technological scheme of the installation on of electron-beam treatment is presented in Figure 8. It includes three principal technological chains: wastewater flow, cooling/ozonizing air flow, and ventilating air flow. Coordinated functioning of those chains is assured by monitoring and control systems. Wastewater flow passes the following elements (in series): Inlet System – Primary Basin – Water Pump 1 (P1) – Nozzles – Reactor – Secondary Basin – Water Pump 2 (P2) – Outlet Line. All the steps of wastewater flow chain are correspondent to flow rate 420,000 kg/h (about 10,000 m^3/day).

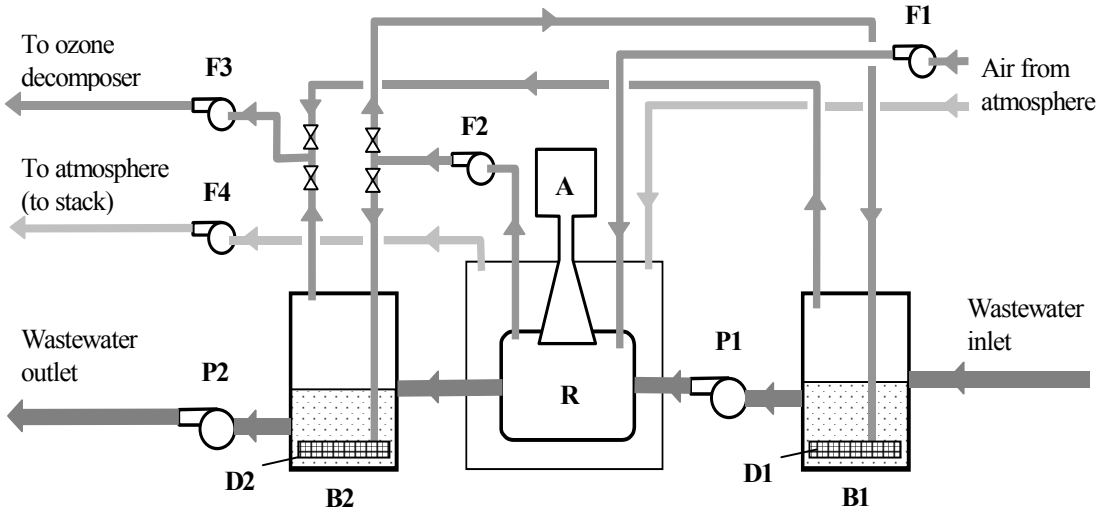

*FIG. 8. Simplified technological scheme of the plant. **F1-F4** – Air fans, **P1-P2** – Water pumps, **D1** and **D2** – Diffusers, **A** – Accelerator, **R** – Reactor, **B1** and **B2** – Primary and secondary basins.*

The building consists of two floors: the ground and the first ones. Profile views of the building are presented in Figure 9. Following notations are used: 1 - Accelerator/Generator Room, 2 - Reactor Room, 3 - Reactor, 4 - Collector, 5 - Reactor Output Channels, 6 - Ceiling Window, 7 - Instruments Room, 8 - Montage Area 9 - Control Room, 10 - Safe Door (First Floor), 11 - Safe Door (Ground Floor), 12 - Primary Basin, 13 - Secondary Basin. Entrance to building is by staircase to montage area on the first floor. Ceiling window may be used for primary installation of accelerator units into the building as well as for repair needs.

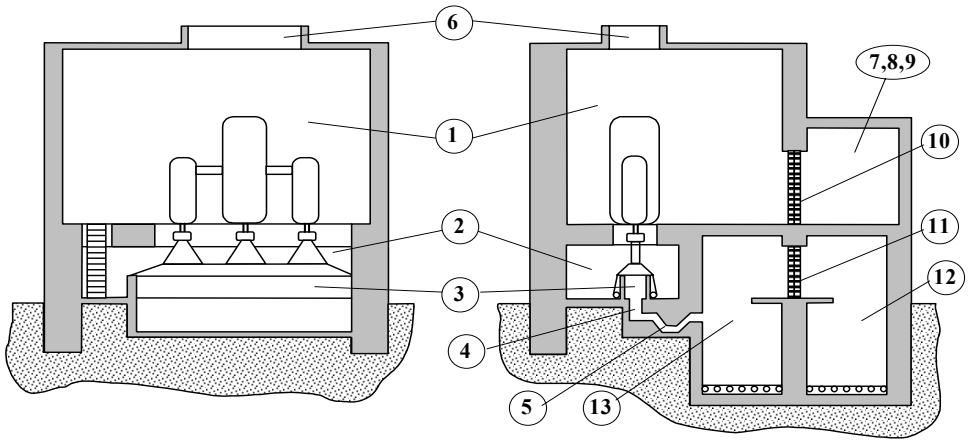

FIG. 9. Profile views of the building.

Where: 1 - Accelerator/Generator Room, 2 - Reactor Room, 3 - Reactor, 4 - Collector, 5 - Reactor Output Channels, 6 - Ceiling Window, 7 - Instruments Room, 8 - Montage Area 9 - Control Room, 10 - Safe Door (First Floor), 11 - Safe Door (Ground Floor), 12 - Primary Basin, 13 - Secondary Basin.

4. CONCLUSIONS

Electron beam treatment combined with conventional purification methods such as coagulation, biological treatment etc. is suitable for reduction of non-biodegradable impurities in wastewater and will extend the application area of electron beam. A pilot plant for treating 1,000m^3 of textile dyeing wastewater per day with electron beam has constructed and operated continuously since October 1998. This plant is combined with biological treatment system and it shows the reduction of chemical reagent consumption, and also the reduction in retention time with the increase in removal efficiencies of COD_{Cr} and BOD_5 up to 30~40%. On the basis of data obtained from pilot plant operation, construction of actual industrial scale plant has started in 2003, and will be finished by 2005. This plant is located on the area of existing wastewater treatment facility in DDIC and to have treatment capacity 10,000m^3 of wastewater per day using one 1MeV, 400kW accelerator, and combined with existing bio- treatment facility.

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TECHNICAL AND ECONOMIC ASPECTS OF RADIATION HYGIENIZATION OF MUNICIPAL SEWAGE SLUDGE USING GAMMA IRRADIATOR

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Abstract

Disposal of sewage sludge, especially in large metropolitan cities throughout the world, is emerging as a serious problem for urban authorities as sludge contains a high load of microbes which are a serious threat to public health.. There exists a need for development of cost effective and environment friendly technologies to overcome the problem. High-energy ionizing radiation can create extremely reactive species like free radicals or ions at room temperature or even at low temperature in any phase and in a variety of substrates without addition of external additives. The highly reactive intermediates produced during radiolysis of water namely, hydroxyl radical OH, hydrated electron $e^-_{aq.}$, and hydrogen atom H have the capability to react with the pathogenic microbes present in the sludge resulting in its disinfection. In India, Department of Atomic Energy (DAE) has designed, built and has been operating, since 1992, a Sludge Hygienization Research Irradiator (SHRI), adjacent to the municipal sewage sludge treatment plant in Vadodara for reduction of pathogens in the sludge. The plant has provided valuable experience regarding the design, operational parameters of the irradiator and irradiated sludge has been shown to be useful as a fertilizer in agricultural practice. The operational experience of SHRI facility, current status, some recent developments and the economic aspects of this technology are presented.

1. INTRODUCTION

The pollution load on the environment is increasing due to increase in population, industrial activities and extensive use of pesticides and other chemicals for cultivation. Of particular concern are wastes that contain potentially infectious microorganisms. The most critical and voluminous source being the city sewage sludge. Sewage is wastewater produced by domestic premises and contains a number of pollutants, which have a major impact on the environment. Sewage typically contains 99.9% water and 0.1% solid. The solid waste, which is organic in nature, is broken down to simple organic compounds in sewage treatment plants and results in sewage sludge as the by-product. The end product of the conventional sewage treatment plant process typically contains about 3 - 5% solid sludge.

Organic materials constitute about 40-80% of the dry weight of solid sludge. The major organic loading originates from human excreta, and is a complex mixture of fats, proteins, carbohydrates, lignin, amino acids, sugars, celluloses, humic materials and fatty acids. A large proportion of these organic materials are in the form of both live and dead microorganisms. The presence of various micronutrients and ability to enhance the organic matter of the soil make sewage sludge a potentially useful material. Although the sludge can be a valuable fertilizer, due to the contamination sludge must undergo a stabilization process. There are various types of treatment systems that are used to treat sewage. The choice system depends on many factors such as:

- Size of the population for which systems is required
- The characteristics of sewage, whether only domestic or containing industrial effluents
- The location of the sewage system – proximity to sea or river
- Effluent standards required for discharge of treated liquors.

A modern conventional sewage treatment processes consists of four levels of treatment, primary, secondary and tertiary. The primary treatment process consists of physical and physico-chemical process wherein the plastics, cloth rags, debris, glass pieces and bigger solid particles are removed by screening, grit chambers and primary settling tanks.

The secondary treatment is essentially a biological processes wherein microbes are encouraged to grow either in aerobic or in anaerobic condition, to consume and degrade the dissolved organic matter. At this stage, the maximum BOD/COD loads are reduced. Some examples of such units are activated sludge process, trickling filter, up flow anaerobic sludge blanket, aerated lagoon etc. The tertiary treatment includes additional removal of solids and organic material and constituents such as nitrogen and phosphorous. The sludge, drawn from the bottom of primary and secondary settling tanks is thickened by various means, digested either aerobically or anaerobically and then, dewatered mainly by sludge drying beds. After dewatering, it is disposed off as landfill material or agriculture manure. Disinfection is rarely done prior to its disposal. The final disposal of the sludge is generally carried out in the following ways:

- Incineration – high energy demand, expensive
- Sanitary Landfill – not desirable as resources are wasted
- Disposal in the sea – not possible everywhere, polluting
- Disposal on agricultural Land – desirable, needs hygienisation.

Due to intensive cultivation practices, agricultural lands worldwide have become vulnerable to degradative processes such as soil erosion from wind and water, nutrient depletion and loss of organic matter, and have suffered a consequent decline in soil productivity. The restoration and rehabilitation of these degraded soils to an acceptable level of productivity can be enhanced by using various off-farm sources of organic wastes, including sewage sludge, municipal solid wastes, and agricultural wastes.

The nutrient content of most sludge makes them useful as fertilizers or as soil conditioners if properly mixed with the surface soil. Sludge has the potential to be an excellent soil conditioner because the humus material in the sludge provides a good matrix for root growth, while the nutrient elements are released in approximately the right combination for optimal plant growth. Soil microbes will assist in further stabilization of any biodegradable organics remaining. Nutrient release with sludge is slower than with chemical fertilizers, allowing the nutrients to become available, as the crop needs it. Land spreading of sludge will become more popular as energy and nutrients become scarcer.

Thus, recycling of the sewage sludge to agricultural land can be an important outlet provided it is carried out in a manner that protects human and animal health as well as environment at large. For the sewage treatment plant operators, it offers a way of generating a value added by-product from the waste whose disposal otherwise is a matter of environmental concern. The sludge after conventional treatment processes still contains a heavy pathogenic microbial load and therefore needs to be hygienized before application in the agricultural processes. In many advanced countries directives requires that the sludge, before applying to agricultural land must undergo an appropriate process to kill off disease causing organisms, which may be present in the sludge, to an acceptable level.

US EPA guidelines have recommended that the number of E.Coli, indicator bacteria for pathogens, should not exceed 1000 per gram of the dry sludge if it is to be applied for agricultural practices [1]. Since land application of sewage sludge on a larger scale is relatively recent, it may not have been considered in the design of sewage treatment plants. The utilization of sewage sludge on a large scale and in a safe manner will necessitate development of technologies that can treat the sludge in a reliable, efficient and cost effective manner.

2. HYGIENISATION OF SEWAGE SLUDGE USING RADIATION

As discussed above, the sludge generated by a sewage plant still contains a high level of pathogens that limit the reuse of this waste that is otherwise a rich source of nutrients. Its disposal in the present form is an economic loss to the country.

Further processing of the sludge to reduce pathogens is therefore necessary, before the beneficial utilization or recycling of solid sludge can be recommended. Thus, there exists a need to further extend the treatment process to include a step that ensures removal of the pathogenic bacteria with a high degree of reliability.

The high-energy gamma radiation from radioactive sources such as Cobalt-60 has the ability to inactivate the pathogens with a very high degree of reliability, and in a clean and efficient manner. The ionizing radiation interacts with matter in two ways: directly and indirectly. In direct interaction, the ionizing radiation interacts with the critical molecules like DNA and proteins present in the microorganism causing cell death. During indirect interaction, radiolysis products of water results in the formation of highly reactive intermediates which then react with the target biomolecules culminating in the cell death.

Ionizing radiation : $H2O \longrightarrow \bullet OH, H\bullet, e\text{-}aq, H2, H2O2$

$\bullet OH, H\bullet, e\text{-}aq, +$ DNA (in microorganism) $\rightarrow$ Damage to DNA, (Inactivation of bacteria)

Presence of oxygen is important in the process as the oxygen is a known radio-sensitizer which helps in fixing the radiation damage done to cells thereby inhibiting their self repair mechanism and resulting in inactivation of the microorganism. The radiation dose required to inactivate the pathogenic bacteria is generally defined in terms of the D10 value, which is the radiation dose required to inactivate or kill microbial concentration by a factor of 10 or by 1 log cycle.

In fact, this has formed the basis of producing radiation sterilized single use medical products which is now a well established industry worldwid [2]. On the same principle, the pathogens present in the sewage sludge can also be effectively removed from the sewage sludge by exposing it to high-energy radiation. The radiation treatment of sewage sludge can offer an efficient, simple and reliable method to produce pathogen free sludge, which can be further upgraded to produce a value added bio-fertilizer and allow recycling of the waste products. Therefore, irradiation of sewage sludge as a tertiary treatment process has been investigated in last few years [3-5].

3. MATERIALS AND METHODS

3.1. The sludge hygienization facility

In India, Sludge Hygienisation Research Irradiator (SHRI), a technological scale demonstration plant, was indigenously designed and established by Isotope Group of Bhabha Atomic Research Centre in collaboration with Vadodara Municipal Corporation and Govternment of Gujarat. The technological demonstration facility at Baroda was established with the goals to:

- Establish the basis for radiation hygienisation of sewage sludge
- Indigenous Design, installation and operation of liquid sludge irradiator
- Demonstrate compatibility with conventional sewage treatment plants
- Study feasibility of utilizing municipal sewage sludge as soil conditioners in agriculture
- Evaluate economic feasibility of the whole process

The plant at its designed capacity of 18.5 PBq (18.5×10^{15}Bq or 500kCi) can treat up to 110 cubic meter per day of sewage sludge emanating from 6 MGD (27 MLD) conventional sewage treatment plant serving domestic population of 0.3 million of Baroda (Fig. 1). The initial source strength of 150 kCi was loaded in 1989. AERB gave its authorization for plant operation in Oct'90. It was formally commissioned in 1992 and has been operational since than. In early 2001 the source strength was augmented to 220 kCi by adding 185 kCi to take care of the depleted Cobalt-60 source.

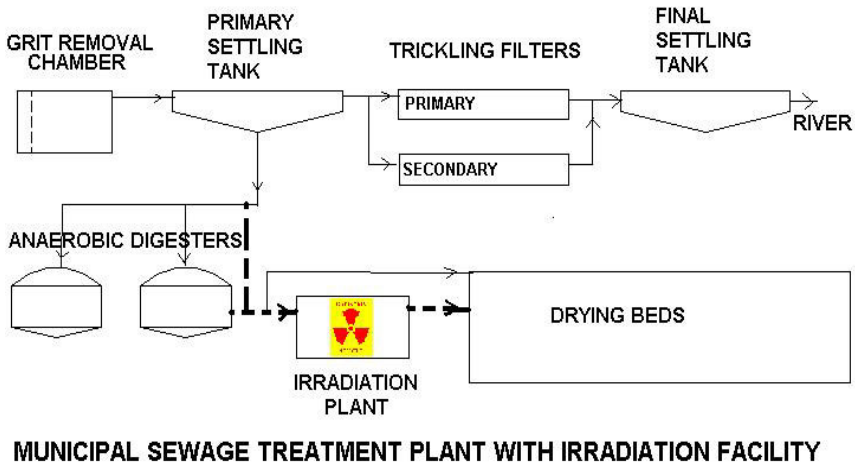

FIG. 1. Location of the radiation plant in the conventional set up at S.H.R.I

3.2. Sludge hygienisation process

Hygienisation of the sludge is carried out in a gamma irradiator with a cobalt-60 source. It consists of typically three main components:

- a cobalt-60 gamma energy source housed in an irradiation vessel
- a concrete-walled room for housing the irradiator and to provide biological shield
- a product handling system that moves sludge into and out of the irradiation vessel.

3.3. Validation of the radiation hygienisation process

One of the important aspects of radiation hygienisation is that the process is mainly dependent on the radiation dose absorbed by the microorganism. Increasing amount of radiation dose decreases the numbers of surviving microorganisms present in the product. The quantity and species of pathogens present in sewage sludge can vary considerably depending upon factors such as time, location, local circumstances and current health of the population.

The published information shows a wide range of concentrations may be present. Since it is not practical to monitor treated sludge for the presence of pathogens, surrogates have to be used for routine evaluation of sludge quality. The surrogate should be an organism found commonly in the sludge that has similar resistance to treatment as pathogens. E.Coli is one such microorganism that has been suggested for monitoring of the quality of irradiated sludge. The details of the microbiological work are presented elsewhere [6].

3.4. Testing of radiation hygienized sludge for use as organic fertlizer

The large scale field trials to study the effect of irradiated sludge on growth and yield of green gram (Vigna radiata (L.) Wilczek), locally known as moong, were carried out at Kasumbia village, near Vadodara during summer of 2003. A randomised block design was used during the trial. The plot size was 12x10 meter. The following treatments were applied for the study:

- T_1: Recommended dose of FYM 10t/ha + chemical fertilizer 20-40-0 NPK kg/ha

- T_2: Sludge based biofertilizer 10t/ha + chemical fertilizer 20-40-0 NPK kg/ha
- T_3: Seed treat. With bio-fertilizer +sludge based bio-fertilizer 5 t/ha
- T_4: Seed treat. With bio-fertilizer +sludge based bio-fertilizer 10 t/ha
- T_5: FYM 5t/ha + sludge based bio-fertilizer 5 t/ha
- T_6: Seed treatment with biofertilizer + T5
- T_7: Sludge10 t/ha
- T_8: Control

Different crop parameters such as plant height, number of branches per plant, pod length, number of pods per plant, number of seeds per plant, seed weight per plant and seed yield were recorded during the study.

4. RESULTS AND DISCUSSION

Figure 2 shows typical survival characteristics of total coliforms present in sewage sludge as function of absorbed radiation dose. The results show that a dose 3 kGy is adequate to reduce the total coliform counts by about 4 log10 cycles.

4.1. Operational experience with SHRI

The operational experience of more than ten years with Sludge Hygienisation Research Irradiator (S.H.R.I.) has established that:

- About 3 kGy of absorbed dose in sewage sludge removes 99.99% of pathogenic bacteria consistently and reliably in a simple manner.
- The continuous operation of SHRI has been very smooth. There have been no operation/maintenance problems since it's commissioning. The plant can be operated even by non-radiation workers.
- The irradiator system can be easily integrated with conventional treatment plant with flexibility of operation. Various dose treatment can be imparted to sludge with addition of sensitizing agents such as oxygen, air, ozone etc.
- The radioactive source loading, unloading or transport into SHRI is very easy and very safe; it can be accomplished in a day.

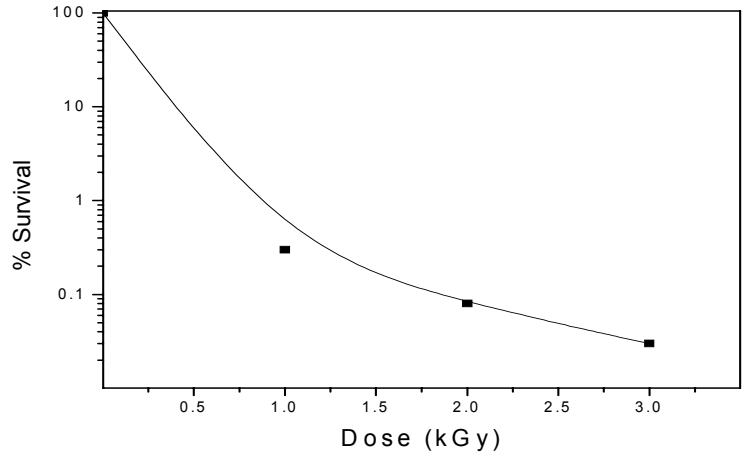

FIG. 2. Survival of Coliform microbial population as a function of radiation dose

4.2. Advantages of radiation hygienisation of sewage sludge

- Radiation treatment plant is easy to integrate with sewage collection and treatment facilities.
- 3 kGy of absorbed dose in sewage sludge removes 99.99% of pathogenic bacteria, which are otherwise responsible for causing diseases such as cholera, typhoid, dysentery etc. Contrary to the conventional processes, the radiation treatment of sewage sludge kills the pathogens in a simple, efficient and reliable way.
- Radiation processed sludge being free from pathogens has potential to be used as manure in agricultural fields. While the continuous use of chemical fertilizers deteriorates the quality of the soil, the sludge enhances the quality and fertility of the soil as it contains useful soil-conditioning materials.
- The radiation processed sludge material is free of any odour.
- The radiation treatment plant can be operated in simple and reliable manner.

4.3. Radiation hygienised sludge as a fertilizer product

The irradiated sludge being pathogen free can be beneficially used as manure in the agricultural fields as it is rich in nutrients required for the soil. Initial field trials, in villages around Vadodara city, of sludge as manure in agriculture fields in winter wheat crops as well as in summer green gram crops has been very encouraging.

Since the irradiated sludge is free from bacteria, this can also be used as a medium for growing soil useful bacteria like rhizobium and azetobactor to produce bio-fertilizers, which can be used to enhance the crop yields. Large scale field trials of utilizing radiation processed municipal sewage sludge in the agricultural fields have been conducted under the supervision of Krishi Vigyan Kendra (KVK, Vadodara). The trials conducted so far have shown that the hygienized sludge based bio fertilizers are very effective in increasing the yields of many crops and can be utilized as organic fertilizer. The results of the study conducted for summer green gram is shown in Table I. These results clearly show the usefulness and effectiveness of hygienized sludge as a manure under actual field conditions.

TABLE I. EFFECT OF NUTRIENT MANAGEMENT TREATMENT ON GROWTH, YIELD AND YIELD ATTRIBUTES OF SUMMER GREEN GRAM

Treatment	Plant Height (cm)	No.of Branches per plant	Pod length (cm)	No. of Pods per plant	No.of seeds per Pod	Seed Yield (kg/ha)
FYM 10t/ha+ chemical fertilizer 20-40-0 NPK kg/ha	33.13	4.33	6.28	35.07	10.73	1086
Sludge based Biofert 10 t/ha +chem..fert. 20-40-0 NPK kg/ha	32.60	4.47	6.35	32.97	11.20	1078
Seed treatment with bio-fert. +sludge based biofert. 5 t/ha	31.60	4.30	6.14	30.37	10.73	835
Seed treatment with bio-fert. +sludge based biofert 10 t/ha	34.44	4.70	6.26	35.47	10.67	1208
FYM 5t.ha+ sludge based biofert. 5 t/ha	31.90	4.10	5.71	34.53	10.07	733
Seed treatment with biofert. + FYM 5t.ha+ sludge based biofert. 5 t/ha	30.35	3.77	6.33	29.93	10.80	766
Sludge 10 t/ha	32.40	4.77	5.98	32.33	10.13	743
Control	28.17	2.93	5.46	26.37	8.93	576
S.Em.	1.01	0.27	0.19	0.98	0.28	54.24
Cd at 5%	3.07	0.81	0.59	2.98	0.84	165
C.V.%	5.51	1.11	5.33	5.30	4.60	10.70

5. ECONOMIC ASPECTS OF RADIATION HYGIENIZATION

Estimated cost analysis of radiation treatment of sewage sludge to treat 376 m^3 of sludge originating from a 48 mld sewage treatment plant:

Total capital investment

- Capital investment : $US 377,500
- Cost of 1800 kCi Co-60 radioactive source: $US 2,000,000
- Total capital cost: $US 2,377,500

Annual fixed cost

- Depreciation (30 years) $US 11250
- Interest on capital 10% per annum $US 236250
- Radioactive source replenishment (12 % per annum) $US 245000

Annual operation cost

- Salaries $US 27,000
- Utilities(Electricity for 20 kW) $US 22500
- Maintenance $US 13500

Total annual cost $US 555,500

Total annual production:

- Wet thickened sludge (376 cu.m. per day) 131600 cu.m.
- Dry sludge (4 % solid content) 5264 tons

Unit cost:

- Wet thickened sludge $US 4.2 per m^3
- Dry sludge $US 105 per ton

The quantification of the economics of cost-benefit relationship of agriculture application of the radiation hygienised sewage sludge needs further evaluation due to the following considerations:

The quantitative analysis of increase in crop productivity will vary from crop to crop and economic benefits derived will vary accordingly. Work is in progress to test the use of radiation hygienised sludge for a variety of crops on large fields to work out this data. It is difficult to quantify the economic gain on improved soil conditions, soil conservation, especially when the magnitude of the improvement may be variable and depends upon the soil and sludge characteristics. Consideration of only the crop nutrient value of sewage sludge neglects these other benefits.

Although the qualitative benefits of agricultural use over other methods of waste disposal such as incineration, land fill or sea disposal are obvious, it is difficult to quantify the economic gain of the radiation process keeping in view the environmental advantages offered by it.

The present practice of disposing sludge increase the load of pathogenic microorganisms in the environment thereby increasing the risk of illness from such microorganisms. The potential economic impact of the pathogen related illnesses include diagonostic cost, treatment cost and lost productivity in the workplace and at home. The treatment of sludge by radiation will eliminate these health risks and hence improve overall quality of life and reduce potential pressure on the country's health care system.

In the field trials for use of hygienised sludge as a soil conditioner, it has been observed that soil treated with the sludge has better water holding capacity as compared to the normal soil. Thus, utilization of sewage sludge in agricultural practices will reduce the demand of water and make this

valuable resource for other applications. The reduced demand of water may also be beneficial in reducing the problem of salinity of the soil associated with excessive use of water in agriculture.

6. CONCLUSIONS

Successful operation of SHRI facility since 1989 has shown very good results on all the above aspects. About 3 kGy of absorbed dose in sewage sludge removes 99.99% of pathogenic bacteria (about 4 log cycle reduction). The continuous operation of SHRI has been very smooth. It produces high value manure containing plant nutrients and soil conditioners for immediate reuse. The technology is easy to integrate with the existing plants and the hygienized sludge has to found to be an effective natural fertilizer in the large field scale trials.

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HIGH POWER ACCELERATORS FOR ENVIRONMENTAL APPLICATION

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Abstract

Cost-effective treatment of municipal and industrial wastewater containing refractory pollutant with electron beam is actively studied in EB TECH Co. EB treatment of wastewater is caused by the decomposition of pollutants as a result of their reactions with highly reactive species formed from water radiolysis (hydrated electron, OH free radical and H atom). However, to have advantages over existing processes, the electron beam process should have cost-effective and reliable in operation. Therefore high power accelerators (400kW~1MW) are developed for environmental application and they show the decrease in the cost of construction and operation of electron beam plant. In other way to reduce the cost for wastewater treatment, radical reactions accompanied by the other processes are introduced, and the synergistic effect upon the use of combined methods such as electron beam treatment with ozonation, biological treatment and physico-chemical adsorption and others also show the improvement of the effect of electron beam treatment for the wastewater purification.

1. INTRODUCTION

In developed and developing countries, where the industries are concentrated in urban areas, resulting in severe water pollution problems in most large cities. Major sources of water pollution include chemical-intensive industries such as textiles, metal plating, electronics, papermill and refineries. Typical contaminants include non-biodegradable substances, grease and oils, acids and caustics, heavy metals such as cadmium and lead, sludge and a long list of synthetic organic compounds. On current trends, industrial water use will more than double by the year 2025 with a four-fold increase in pollutant emissions to watercourses. In some countries, industrial water demand will rise even more sharply. Therefore, the treatment of municipal and industrial wastewater becomes a more important subject in the field of environment engineering.

EB-treatment gives essential change of various properties of pollutants - solubility, volatility, absorptivity, reactivity etc. It stimulates the development of productive combination of EB-method and various conventional methods of wastewater treatment. At present the EB-treatment has not wide application and spreads less than conventional methods. However, first experience of the industrial application shows that EB-treatment can occupy the quite essential place at future. Already now the EB-technology and its combination with conventional ones provide noticeable economy of time, area and industrial power to wastewater treatment. Continuous reinforcement of ecological standards is additional motivation for elaboration and industrial application of EB-treatment.

2. BENEFITS OF ELECTRON BEAM TREATMENT

2.1. Barriers for applications

2.1.1. Public acceptances

When radiation technologies are involved, the same as in other industrial applications, people are uneasy for their safeties even in the case of electron beam, which are machine-generated controllable radiation sources. Also they worry about the production of radioactive material and dangerous new species by radiation. Up to now, most industrial accelerators are operated with not more than 10 MeV and the possibility to produce radioactive material is not worth consideration. Nevertheless, the change of toxicity and transmutation that might occur in wastewater due to the irradiation needs more careful studies.

2.1.2. Technical problems

In comparison with flue gas purification, radiation treatment on wastewater has much more complicated problems. In flue gas purification, the targets to clear would be simply SO_X, NO_X (and in some cases Dioxin and others) and the treatment process are almost universal to each different case, however in wastewater treatment, each wastewater has different contaminants and therefore independent verification and process set-ups are required. Moreover, the by-products are no useful and difficult to analyze. Table I shows the comparison between e-beam flue gas purification and wastewater treatment in several points.

2.1.3. Competition with other process

Conventional biological systems are inexpensive for large amount of wastewater. Normally they cost around $US 1 per cubic meters of wastewater for operation. However some organics are not removed or long time for removal. Ozonation and membrane filters are acceptable for small to medium scale plant, but not economical for larger plant. Electron beam facilities with low doses are competitive power in large plant.

2.1.4. Benefits

Electron beam processing is usually considered as expensive process since the initial investment cost is high due to the cost of accelerator. Additional investment for alternatives or by-pass is required in case of shutdown of electron beam facilities since the most of all the environmental protection facilities should operate all the year round. Non-universality of processing technology in each wastewater treatment facility also costs for additional test and studies before installation.

TABLE I. COMPARISON OF FLUE GAS PURIFICATION AND WASTEWATER TREATMENT WITH E-BEAM

	Flue gas purification	Wastewater Treatment
Contaminants to clear	SO_X, NO_X, (Dioxin)	Complex
Cleaning Process	Simple	Limitation in depth Combined with other process
Competitiveness to others	Proved by commercial operation	Complicate to analyze
Technology	Fully developed	Lab. to pilot scale
Economies	Proved through pilot and commercial plant	Complicate to analyze
By-products	Useful for fertilizer	-

2.2. How to improve benefits

The key to successful application of electron beam in environmental protection is how to estimate the cost benefits ratio. To compete with other processes the electron beam installation has to consider the following parameters:

- Reduce the required doses
- Improve efficiencies
- Reduce the cost for electron beam facilities

For industrial wastewater with low impurity levels such it is desirable to keep the irradiation doses not more than several kGys (less than 1 kGy is preferable). To reduce the required doses application of useful additives or addition of radical enhancer/reducer is effective. The combined electron beam processing with other methods, such as biological treatment or coagulation is another way to decrease the irradiation doses.

Treatment of textile dyeing wastewater is one example of combined electron beam and biological treatment. With the low doses like 1kGy, an accelerator should treat several thousand tons of wastewater per day, and effective wastewater delivery system and efficiency of accelerator machine itself is also important. The water delivery system should give the uniform dose distribution as well as the massive delivery of water under electron beam. In our experiments we choose injection of wastewater through flat nozzle and it showed the possibility of massive delivery of water with dose uniformity.

Most important factor to control the economies is the cost of electron accelerator in use. As is shown in Table II and Fig. 1, the price of accelerators are governed by the power and the accelerators of power as high as possible has relatively lowest cost for unit power generation and most economical to apply in environmental application. Up to now accelerators of several hundred kilowatts are available from accelerator manufacturers in the world.

TABLE II. TYPICAL PRICE OF ACCELERATORS

Power(kW)	Price(M$US)	Cost for 1kw ($10^4$$US)
20	0.5	2.5
40	0.8	2.0
100	1.0	1.0
200	1.5	0.75
400	2.0	0.5

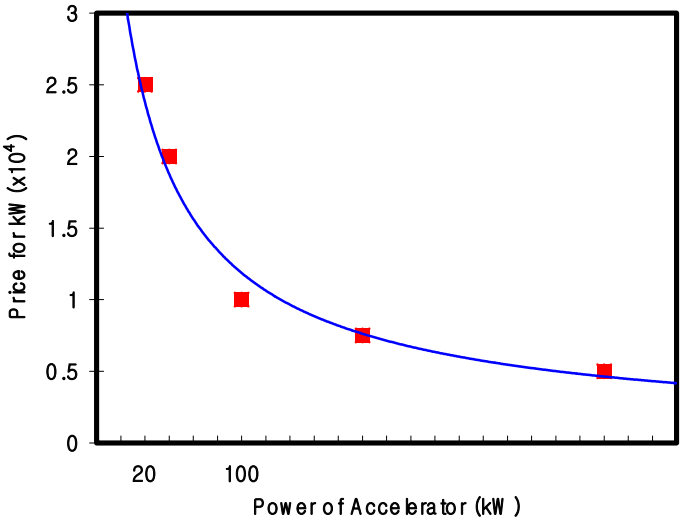

FIG. 1. Typical price of accelerators

3. HIGH POWER ACCELERATORS

The key to successful implementation of electron beam in environmental protection depends on how to control the cost-benefits ratio. To compete with other processes electron beam system should operate with cost-effective accelerator with low doses.

Therefore high power accelerators are introduced to environmental application. The most powerful and reliable accelerator for wastewater treatment is 1MeV, 400 kW accelerator which is produced together by EB-TECH Co. and BINP, Russia. This accelerator can deliver the maximum power of 400 kW. Titanium foils are used for windows, which are cooled by air jet blow and water. High voltages are generated through the inductions of coils in main body and SF_6 gases are used for protecting electrical discharges.

The first accelerator of this model is under installation in Daegu, Korea for treating industrial wastewater form textile dyeing industries (Fig. 2). Figure 3 shows the design of this accelerator.

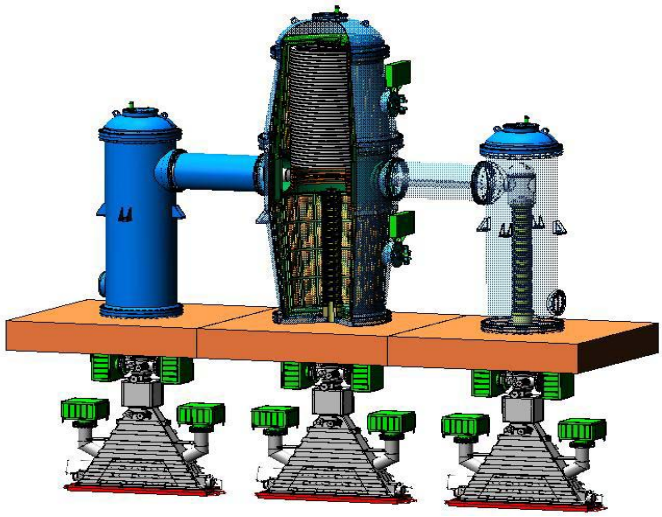

FIG. 2. Typical model of the accelerator of 1MeV, 400kW in assembly

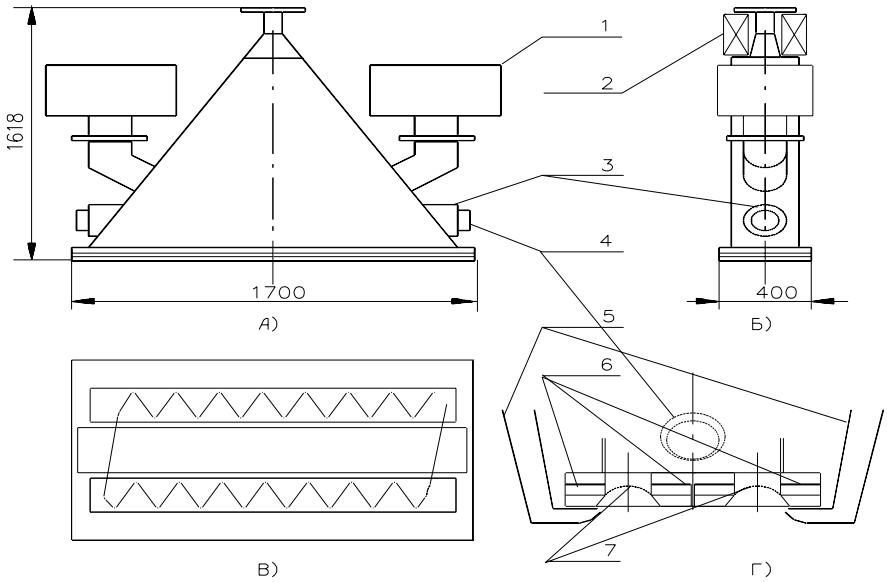

FIG. 3. Double extraction window : 1 ion pumps, 2 scanning system, 3 cylinder flange, 4 protection cylinders, 5 foil blow cooling, 6 foil fixation frame, 7-extraction foils

4. ECONOMIC EVALUATION OF COMMERCIAL PLANT

Based on the data obtained in the economical evaluation and pilot plant experiments, the suitable doses cost-effective electron beam plant are determined as around 1 kGy or less for the flow rate of 20 000m^3 effluent per day. Therefore, the cost assessment of radiation processing plant with e-beam is accomplished based on 1 kGy and 400 kW electron accelerator.

Cost for such as high power accelerator is around US$ 2.0M, while building, piping, other equipment and construction works can be estimated of US$ 1.5M. Even by considering the additional cost for tax, insurance and documentation as of US$ 0.5M, the overall capital cost for plant construction and the operation cost are approximately US$ 4.0M and US$ 1.0M respectively, as stipulated in Table III and Table IV.

TABLE III. CONSTRUCTION COST FOR INDUSTRIAL PLANT (UNIT US$ M)

	Cost	Remarks
Accelerator - 1MeV, 400kW, double window	2.0~2.5	Cost for Land, R&D, Approval from Authorities are not included
Water reactor & other Raw Material	1.0~1.5	
Installation cost – welding/piping/inspection etc.		
Design		
Shield Room & Construction works		
Others - transportation, tax, insurance etc.	0.5	
Total	4.0~4.5	~ 4M$

TABLE IV. OPERATION COST FOR INDUSTRIAL PLANT (UNIT $US K)

Items		Addition of E-beam	Remarks
Operation Cost	Investment (US$ k)	(4,000)	
	Interest	240	6%
	Depreciation	200	20 yrs
	Electricity	320	800 kW
	Labour	100	3 shifts
	Maintenance, etc.	80	2%
Total cost		940	US$~ 1M/yr

Above estimation doesn't include the cost for land, R & D and the cost for the approval form authorities. Expected construction period includes 11 months in civil and installation works and 3 months for trial operation. To estimate the operation cost, the electricity consumption of accelerator and other equipment is calculated as 500kW (80% efficiency) and 300kW to the total of 800kW. Based on the year round operation (8000 hr/yr), it costs 320,000 US$/yr when the cost of electricity (kWh) was assumed to be 0.05 US$. The labor cost of operator is calculated on 3-shift work and is approximately 100,000 US$/yr.

Therefore, the actual operation cost for 20,000 m^3/day plant comes up to around 1.0M US$/yr including the interest and depreciation of investment and is approximately $US 0.12 in construction and US$ 0.03 in operation for each m^3/day of wastewater. When compared to other advanced oxidation techniques such as Ozonation, UV techniques etc., the radiation processing is more cost-effective and convenient treatment for wastewater.

5. SUMMARY

5.1. Requirements of electron accelerator for environmental application

Accelerators for environmental application should be satisfied in followings;

- Accelerator itself has strong and firm configuration for year round operation
- Economical in power consumption
- Easy and safe operation

Introduction of more powerful accelerator with adequate wastewater delivery system could make economical and technical advantages in competition with other methods.

5.2. Generals comments for wastewater treatment with electron beam

For industrial wastewater with low impurity levels such as contaminated ground water, cleaning water and etc., purification only with electron beam is possible, but it should be managed carefully with reducing required irradiation doses as low as possible. Also for industrial wastewater with high impurity levels such as dyeing wastewater, leachate and etc., purification only with electron beam requires high amount of doses and far beyond economies.

Electron beam treatment combined with conventional purification methods such as coagulation, biological treatment, etc. is suitable for reduction of non-biodegradable impurities in wastewater and will extend the application area of electron beam.

A pilot plant with electron beam for treating 1,000m^3/day of wastewater from dyeing industries has constructed and operated continuously since Oct 1998. Electron beam irradiation instead of chemical treatment shows much improvement in removing impurities and increases the efficiency of biological treatment On the basis of data obtained from pilot plant, construction of industrial scale plant has started in 2003, and will be finished by 2005. This plant is located on the area of existing wastewater treatment facility in DDIC and to have treatment capacity 10,000m^3 of wastewater per day using one 1MeV, 400kW accelerator, and combined with existing bio- treatment facility.

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HIGH POWER ACCELERATORS AND PROCESSING SYSTEMS FOR ENVIRONMENTAL APPLICATION

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Abstract

Radiation processing can be successfully applied to treat gas, liquid and solid phase of matter to solve specific problems related to environment protection. The most important reasons why the high power accelerators may be used for the purification purpose are: ability to transfer large amount of energy into irradiated object and the biocide properties of beam of accelerated electrons. Appropriate accelerator and under beam equipment selection should be performed to meet all technical and economical conditions for successful process implementation. The optimization of geometry of irradiation unit, energy of accelerated electrons and other process and equipment parameters should be performed to minimize beam power losses and increase process effectiveness what may create suitable conditions for radiation technology application in the field of environment protection.

1. INTRODUCTION

The requirements in environmental protection due to enforcing of environmental protection regulations results in intensification of research and practical implementation radiation technology in this particular field. Reduction of non-organic and organic pollutants as well as destruction of bacterial flora, particularly including pathogenic bacteria, parasites and viruses became very important practical issue related to the utilization of ionizing radiation. The most important reasons why the accelerator technology may be used for the purification purpose are ability to transfer large amount of energy into irradiated object and the biocide properties of beam of accelerated electrons. Radiation processing can be applied to treat gas, liquid and solid phase of matter (Table I). Flue gas treatment, VOC removal, drinking water purification, wastewater and industrial wastes treatment, reduction of sewage sludge sanitary contamination, solid agriculture wastes transformation are the best examples of practical implementation of radiation technology in environment protection.

TABLE I. RADIATION TECHNOLOGY APPLIED IN ENVIRONMENT PROTECTION

Phase	Object	Additives	Process
Gas	Flue gas	SO_2; NO_x	Removal
	VOC	Organic compounds	Degradation, removal
Liquid	Drinking water	Chemical pollutants	Degradation, removal
	Wastewater	Bacteria; viruses; parasites	Disinfection
	Industrial wastes	Organic and nonorganic compounds	Degradation, removal
Solid	Sewage sludge	Bacteria; viruses; parasites	Disinfection
	Solid materials	Agriculture wastes	Transformation

The utilization of radiation technology for environment protection has been introduced in several developed countries years ago. Research on the application of radiation technology for environmental protection has dealt with sewage sludge to be used in agriculture, and involved study on the effect of radiation on the sanitary condition of sediments and their technological properties. One of the radiation processes which was successfully demonstrated in many laboratories and pilot plant facilities is the reduction of SO_2 and NO_x pollutants from flue gases emitted during fuel combustion used in electrical power and heat production. The industrial implementation of electron beam process for flue gases treatment has been introduced in China and Poland few years ago.

When assessing the effectiveness of radiation processing the product characteristics apart from the investment and facility operation costs should be considered (Table II). Economic viability of radiation method selected to solve particular problem of environment protection should be always compared with more classical approach.

TABLE II. RADIATION PROCESS EFFECTIVENESS

Acceptable price of 1 W electron beam power	Type of radiation process	Product characteristics
100-250 $US/W	Semiconductors modification	Low dose Small scale High unit price
100-50 $US/W	Radiation sterilization	Medium dose Large scale Medium unit price
<2.5 $US/W	Flue gas treatment	Low dose Very large scale No commercial value

The advantage of radiation process in destruction and removal of chemical and biological waste is connected to its high efficiency and possibility to transfer high amount of energy directly into the object under treatment. Disadvantage which is mostly related to high investment cost of accelerator may be effectively overcome in future as the result of use high power accelerators and accelerator technology new developments. It should be noticed that environmental applications of electron beam technology are examples of high risk and not very high payoff but radiation technology transfer to environmental application could be in result a substantial improvement in public health.

2. ELECTRON ACCELERATORS FOR RADIATION PROCESSING

2.1. Criteria for selection of accelerators

Although there are many accelerator manufacturers offering a wide range of accelerators performance ratings, only few would be suitable for particular application. To perform suitable selection the general requirements should be form to evaluate each specific offer. The basic criterions of accelerator selection are formulated in Table III. Radiation facility should be designed with possibly:

- Low electron energy to reduce investment and unit operation costs,
- High beam power to increase productivity and reduce unit operation cost,
- High accelerator electrical efficiency to reduce exploitation and unit operation costs,
- High beam utilization to increase productivity and reduce unit operation cost.

The present achievement of accelerator technology for radiation processing illustrates Table 4. High power accelerators have been developed to meet specific demands of environmental application and high throughput processes to increase the capacity and reduced unit cost of operation. Such accelerator construction must be compromise between size, efficiency and cost in respect to the field of its application.

TABLE III. CRITERIONS OF ACCELERATOR SELECTION

No	Criterion of selection	Remarks
1	Fundamental accelerator parameters: Electron energy Average beam power	The basic requirements which define technological capabilities and facility productivity
2	Terms of accelerator purchase: Price Producer Terms of delivery and installation Warranty conditions Exploitation cost	Economical aspects of accelerator purchase which define investment and exploitation costs; period of time needed for facility completion
3	Auxiliary accelerator parameters: Scan performances Auxiliary parameters Measures and control Main components and systems Auxiliary components and systems Accelerator external supply service	Auxiliary parameters which may characterize accelerator quality and provide necessary data for facility design

TABLE IV. ACCELERATORS FOR RADIATION PROCESSING (RECENT ACHIEVEMENTS)

Accelerator type	Direct DC	UHF 100-200 MHz	Linear 1.3-5.8 GHz
Beam current	< 1,5 A	< 100 mA	< 100 Ma
Energy range	0.1-5 MeV	0.3-10 MeV	2-10 MeV
Beam power	400 kW	700 kW	150 kW
Efficiency	60-80 %	25-50 %	10-20 %

Higher number of accelerators build by certain accelerator producer may significantly reduce the cost of accelerator manufacturing. The same effect can be obtained due to progress in accelerator technology development based on new constructions and components. The substantial effect could be achieved by support of R&D study of accelerator technology by governmental and international institutions. Basic parameters of selected accelerator constructions are included to the Table V.

TABLE V. SELECTED ACCELERATORS FOR RADIATION PROCESSING (BASIC PARAMETERS)

Manufacturer (accelerator type)	Energy [MeV]	Current [mA]	Power [kW]	Price* [M$]	Cost [$/W]
IBA, Belgium (UHF)	10	15	150	6.1	40.7
RDI, U.S.A. (DC)	5	50	250	4.9	19.6
NHV, Japan (DC)	5	30	150	5.0	33.3
Vivirad, France (DC) (under development)	5	200	1000	4.4	4.4
INP, Russia (UHF)	5	10	50	1.2	24.0
INP, Russia (DC)	1	400	400	2.0	5.0

* First quarter of 2004

Electrical energy consumption becomes more important for high electron beam power accelerators with low price per 1 W of beam power (low investment cost). The cost of electrical energy is the significant part of exploitation cost for flue gas facility in spite of high electrical efficiency of the accelerator which was applied in this facility what is illustrated by Table VI.

TABLE VI. ECONOMICAL ASPECTS OF ACCELERATOR ELECTRICAL EFFICIENCY

Parameter	Radiation sterilization facility	Industrial facility for flue gas treatment
Accelerator parameters: electron energy beam power electrical efficiency	 10 MeV 20 kW 10 %	 1 MeV 1,200 kW 80 %
Investment cost	7,500 k$US	18,000 kW
Operational cost: total power line	 1,550 k$US 40 k$US	 2,940 k$US 480 k$US
Share of electrical power cost	2.6 %	16.3 %

Future progress in high power accelerator technology development will be related to:

- High power induction linac,
- CW linear electron accelerators,
- Compact high power HF transformer accelerators,
- Very high power transformer accelerators (1-5 MeV; 0.5-1 MW),
- Modern power components applications in accelerator technology,

2.2. Transformer accelerators

DC voltage is used to accelerate electrons in direct acceleration method. DC voltage power supplies as high voltage sources are usually based on use high power, oil or gas filled transformers with rectifier circuit. They are relatively simple and the most reliable accelerator component. HV cable is frequently applied to connect power supply and accelerating head when voltage level not higher than 1 MV. The voltage level above 1 MV in conventional transformer is impractical because of technical problem with insulation and dimensions of such device. Medium energy (0.5-5 MeV) can be obtained by significant modification of high voltage generator. Different type of inductance or capacitance coupling makes possible to multiply AC primary voltage and obtain up to 5 MV of output voltage. The main parameters of selected transformer accelerators are shown in Table VII.

High voltage iron core transformer and semiconductor rectifier circuit was used in EPS-800-375 accelerator developed by Nissin HV, Japan. Oil insulating system was adopted. Low frequency power line (50/60 Hz) is applied to ensure high efficiency of AC to DC conversion (over 90%). Described above power supply construction was successfully implemented by different accelerators manufacturers. The MW beam power level may be obtained on the base of exiting technology this type. Many different constructions have been built by major accelerator producers like Energy Science Inc. USA, BINP, Russia, NIIEFA Russia, Radiation Dynamics USA and others.

The Cockroft-Walton HV cascade multistage rectifier circuit was used in addition to relatively low voltage transformer in EPS-4 electron accelerator built by Nissin HV, Japan. Multistage rectifier circuit and 3 kHz AC voltage are applied. Accelerators this type are applied in the field of radiation sterilization. High voltage coreless transformer concept was applied in ELV 12 accelerator manufactured by BINP, Russia. The certain number of secondary coils is needed to obtain required output voltage. There is no central magnetic guide what simplifies the high voltage source design. The central pressure tank is used to install HV transformer, accelerating section and scanner. Two more tanks are used with additional accelerating tube and scanning devices. SF_6 gas insulating system is used.

Coreless accelerators are usually operated at AC voltage with frequency 0.4-1 kHz to reduce the accelerator dimensions. Electron energy 0.2-2.5 MeV can be obtained in such accelerators.

TABLE VII. PARAMETERS OF SELECTED TRANSFORMER ACCLERATORS

Accelerator type / Parameter	EPS-800-375	EPS-4	ELV 12
Nominal energy	700 keV (800)	1-5 MeV	0.6-1,0 MeV
Energy stability		± 2%	
Nominal beam current	375 mA	30 mA	450 mA
Beam current stability		± 2%	
Beam power	262.5 kW (300)	150 kW	400 kW
Scan width	225 cm	140 cm	160 cm
Dose uniformity	± 5 %	<± 5%	
Mode of operation:	continuous	continuous	continuous
No of accelerating heads	2 sets x 2 heads	one head	3 heads (*1)
Total beam power	1050 kW (1200)	-	450 kW
Power consumption	1198 kW (1364)	220 kW	600 kW
Electrical efficiency	88 % (88)	68 %	66 %
Producer:	Nissin HV	Nissan HV	BINP

*1 - Maximum beam current per one head 200 mA.

Electrical efficiency of accelerator facility should be carefully investigated. Table 8 shows data related to EPS 800-375 accelerator. As can be easily noticed relatively big part of electrical energy consumption is related to intense air cooling and ozone exhaust blower what increases the total electrical energy consumption of the facility.

TABLE VIII. ELECTTRICAL SUPPLY OF ELECTRON ACCELERATOR EPS 800-375

Beam power	600 kW
Power consumption	
Power supply 6 kV, x 3 Control power 380 V, x 3 Vacuum power 380 V, x 3	667 kW 10 kW 5 kW
Power consumption	682 kW
Power efficiency	88 %
Ventilation and air cooling	
Window cooling blower	150 kW
Ozone exhaust blower	11 kW
Total power consumption	843 kW
Total power efficiency	71 %

2.3. UHF accelerators

Resonant UHF accelerators are based on one large resonant cavity working at the frequency over hundred MHz. The high power vacuum tubes are applied to provide necessary electromagnetic energy which is used to accelerate electrons in accelerator this type. UHF accelerator requires relatively simply and compact DC or pulse modulators to generate UHF oscillations. Medium and high electron energy level with appropriate beam power can be obtained (Table IX) in such accelerator. ILU 10 accelerator construction is based on one coaxial resonator operating in pulse regime. The resonator is made of two separate halves mounted inside of stainless steel vacuum envelope. The central cylindrical part of resonator forms the accelerating gap.

The electron injector consists of a grid, made in upper electrode to control beam current by changing the value of positive bias voltage on the cathode with respect to the grid. The self-excited

generator consist of two industrial vacuum triodes is used to form UHF oscillation inside of coaxial cavity and provide necessary energy for electron acceleration process.

TABLE IX. SELECTED PARAMETERS OF UHF ELECTRON ACCELERATORS

Accelerator type/ Parameter	ILU 10	RHODOTRON TT 300
Nominal energy	5.0 MeV	5 and 10 MeV
Energy stability	±2.5%,	+0 keV-250 keV
Nominal beam current	10 mA	15 mA
Beam current stability	±2.5%	-
Beam power	50 kW	150 kW
Accelerating voltage frequency	115±5 MHz	107.5 ±1 MHz
Operation mode	pulse	continuous
Pulse duration	0.35-0.5 ms	-
Pulse repetition frequency	2-50(60) Hz	100 Hz ±5%
Scan width	98 cm	100 cm
Dose uniformity	<±10%.	<±5%
Power consumption	180 kW	<370 kW
Electrical efficiency	28%	40%
Producer:	BINP, Russia	IBA, Belgium

Rhodotron operates on the basic principle that electrons gain energy when they cross a region where an electric field exists. The unique feature of Rhodotron construction is related to its single cavity construction, which has been crossed by electrons several times to gain energy (accelerating cavity of the TT-300 is a half-wavelength coaxial line shorted at both ends and resonating at 107.5 MHz). In the Rhodotron TT 300, each time the electrons are crossing the cavity, they gain 1 MeV. Ten passes and nine magnets are therefore required to obtain 10 MeV electron beam energy. New Rhodotron electron accelerator arrangement TT-1000 was developed. Using multi pass system across resonant cavity 5 and 7.5 MeV electron energy and up to 700 kW beam power was obtained.

2.4. Linear microwave accelerators

The main feature of accelerator this type is the microwave energy use in electron accelerating process. Power supplies as microwave generators are usually built for S-band or L-band frequencies (3000 - 1300 MHz). A large number of small resonant cavities are used. Microwave energy source parameters are playing the crucial role in linacs performance. The klystrons are more stable in frequency and power but they have efficiency of 40-50% in comparison with 70% efficiency of magnetrons.

Linacs can be built with traveling or standing wave configuration. The last technology allows obtain higher accelerating gradient in cost of more sophisticated microwave power system and acceleration section technology. Accelerators those type are not suitable for environmental application due to low electrical efficiency (10-20%) and limited beam power (50 kW). Continuous wave (cw) operation may significantly improve electrical efficiency (up to 40%) and afford MW beam power level in future.

3. FLUE GAS IRRADIATION

The reduction of SO_2 and NO_x pollutants from flue gases, emitted during fuel combustion in electrical power and heat production, is one of the radiation processes which were successfully demonstrated in many laboratories and pilot plant facilities.

The process of those pollutants removal was invented in Japan. The research on technology development was also continued in USA, Germany and Poland in different pilot plant installations.

The main objectives of the research carried out in pilot plant facilities made electron beam scrubbing a viable and useful process for industrial application. In particular following topics were investigated:

- quantitative characteristics of the process;
- test multistage irradiation;
- optimize collecting process and by product handling systems;
- study and evaluation commercial characteristic of the process;
- evaluate the reliability of the process for long operation;
- energy consumption optimization;
- high power accelerator construction;
- improve necessary areas of the facility.

Full scale industrial plants have been operated in China and Poland. Other industrial facilities for flue gas treatment are under consideration and construction. It was clearly established that the industrial implementation of electron beam process for flue gases treatment requires accelerator modules with beam power 300 kW or more and electron energy in the range 0.8 to 1 MeV. Table X shows basic parameters of selected pilot and industrial facility. Accelerators suitable to fulfill such requirements are based on high power high voltage transformers according to present state of art in accelerator technology.

TABLE X. BASIC PARAMETERS OF PILOT AND INDUSTRIAL FACILITIES FOR FLUE GAS TREATMENT

Parameter	Nagoya Pilot Plant, Japan	Kawęczyn Pilot Plant, Poland	Pomorzany EPS, Poland
Flue gas stream	12,000 Nm^3/h	20,000 Nm^3/h	270,000 Nm^3/h
Removal efficiency:			
de-SOx	94 %	96 %	80 %
de-NOx	80 %	72 %	70 %
Accelerators:			
Energy	800 keV	700 keV	700 keV
Beam Power	3 x 45 mA	2 x 72 mA	4 x 375 mA
Vessel size	2.4 x 1.9 x 14 m	Ø 1.6 m x 10 m	2 x Ø 2.6 m x 14 m
Vessel cross section	4.9 m^2	2 m^2	2 x 5.3 m^2
Beam utilization	66 %	64 %	68 %
Gas velocity (max.)	1 m/s	2,8 m/s	7 m/s
Dose	10.5 kGy	11.5 kGy	8 kGy
Thickness of 1st foil	38 µm (Ti)	50 µm (Ti)	50 µm (Ti)
Thickness of 2nd foil	30 µm (Ti)	50 µm (Ti)	50 µm (Ti)

The construction of the double window system was carefully investigated at Kawęczyn Pilot Plant to increase efficiency of the energy transfer from electron beam to the gas phase inside reaction vessel with axial symmetry [1]. Beam power losses in double window system consisted of two titanium foils 50 µm thick and placed at a distance 70 mm were identified (Table XI).

The optimization of reaction vessel dimensions, energy of accelerated electrons, gas temperature should be performed to minimize beam power losses. The better quality window material with smaller thickness is recommended. The higher initial electron energy should be applied with suitable size process vessel to reduce total losses of the beam power and increase effectiveness of the flue gas treatment process.

TABLE XI. BEAM POWER LOSSES IN DOUBLE WINDOW OUTPUT SYSTEM AT AWECZYN PILOT PLANT (Two titanium foils 50 μm thick, placed at a distance 70 mm, reaction vessel with axial symmetry)

Electron energy	500 keV	600 keV	700 keV
Window (Ti, 2x 50 μm)			
Reflection	24.9 %	17.8 %	12.2 %
Absorption in window	13.5 %	11.0 %	9.3 %
Air (70 mm)			
Absorption	2.5 %	2.0 %	1.7 %
Reaction Vessel			
Wall losses	7.0 %	-	-
Bottom losses	-	-	12.5 %
Total losses	47.9 %	30.8 %	35.7 %

4. WASTEWATER IRRADIATION

Radiation processing was found effective in water purification (decomposition of toxic substances) and wastewater disinfection. The facility throughput can be increased and unit cost of wastewater decreased by improvement the coefficient of electron beam utilization. Higher process efficiency and lower unit cost can be obtained by implementation lower dose level. The minimum dose depends on origin, specific properties and contamination of certain waste water. It can vary from 0.2 up to 2 kGy. Fig. 1 shows depth dose distribution in one side (single layer) wastewater irradiation process. Calculation was performed with application of ModeRTL 2.1 computer program.

The water and wastewater treatment process was investigated in laboratory and pilot plant facilities in U.S.A, Japan, Brazil, Russia, Austria, Korea and several other countries. The Miami Electron Beam Research Facility was equipped with transformer accelerator (electron energy 1.5 MeV, beam power 75 kW) [2]. Initial research was focused on determining the disinfection kinetics of bacteria in different wastewater streams at large scale (flow rate of 2-8% ss digested sludge 0.46 m^3/min; 645 m^3/day). A selected influent stream was presented to the scanned beam in falling sheet approximately 122 cm wide, using weir system. Influent water enters from the bottom and is debubbled as it rises to spill over weir. At the designed flow the water is approximately 3.8 mm thick. The scanner window is protected against water splashes by secondary titanium window. Since the maximum penetration in water is 7.4 mm for 1.5 MeV, some electrons pass through the stream and not all of the beam energy is transferred to the water. Energy transfer efficiency is 65%. Operating and maintenance cost (dose 5 kGy; flow rate 0.61 m^3/min) is 1.1 \$US/m^3 (facility amortization and overhead costs are not included).

Up-flow irradiation devices for electron beam wastewater treatment process was demonstrated in pilot plant establish in Brazil [3, 4]. An up-flow delivery system significantly alleviates energy transfer to the stream of wastewater and allows apply accelerators with relatively low electron energy. Irradiation system efficiency 67-76% was obtained in certain device configuration. Titanium foil 40 μm thick was used in some experiments to protect accelerator window and allows the irradiation device to work as a closed system. The estimated process cost was found to be 1.2 \$US/m^3 for dose rate 2 kGy, flow rate 70 m^3/h, electron energy 1.5 MeV and beam power 60 kW.

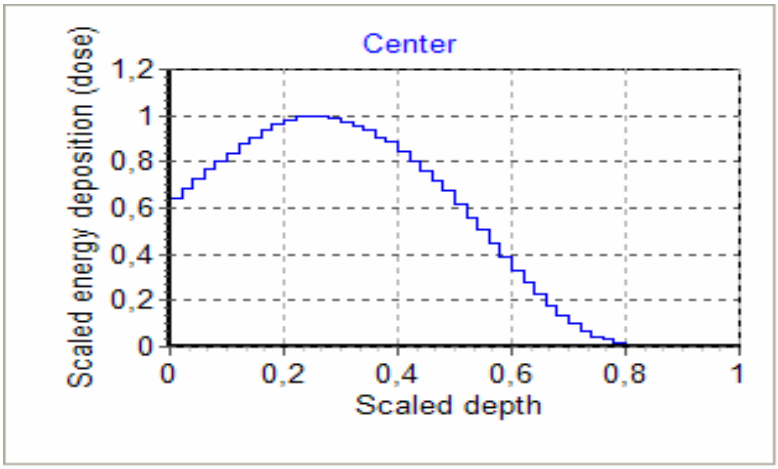

FIG. 4. Depth dose distribution in one side (single layer) wastewater irradiation process:electron energy 1 MeV; beam current 150 mA; surface dose 2.0 kGy; max. dose 3.2 kGy; depth of wastewater layer 5 mm (density 1 g/cm^3); useful depth 2.5 mm; scan length 160 cm; distance from window to waste water surface 30 cm; wastewater flow speed 10 m/s.

Combined ozone-electron beam treatment of water in the aerosol flow was developed in Russia with application low cost, low energy accelerator [5]. The facility output 500 m^3/day was obtained for the dose 1.3 kGy, electron energy 0.3 MeV and beam power 15 kW. Pump and 4 sprayers are used to form the foam in the channel 70 cm wide and 9 cm thick. Speed of wastewater flow 9.6 m/s and speed of air feed up to 290 dm3/s was applied. Upon electron beam treatment the aggregation of disperse particles occurs and participate is formed. It is collected at the bottom part of irradiation chamber and removed together with purified water. The cost of wastewater electron beam treatment was found to be 0.34 \$US/m^3. Accelerator with 300 kW beam power should be applied to purify wastewater stream with flow rate 10,000 m3/day according to estimation performed on the base of pilot plant operation.

Combined ozone-electron beam treatment was applied in Austria for groundwater remediation [6]. Turbulent flow conditions of irradiated water have been demonstrated in bench scale facility. 3 mm thick water layer was treated successfully by 500 keV electrons (penetration range 1.4 mm). It was found that the presence of ozone allowed reduce the dose from 370 Gy to 45 Gy what decrease estimated cost of the treatment from 0.25 to 0.07 \$US/m3.

TABLE XII. RADIATION TREATMENT OF WATER AND WASTEWATER IN DIFFERENT PILOT PLANTS

Parameter	Unit	U.S.A. [2]	Brazil [3, 4]	Russia [5]	Austria [6]
Electron energy	[MeV]	1.5	1.5	0.3	0.5
Beam power	[kW]	75	60	15	12,5
Energy transfer eff.	[%]	65	67-76	-	50
Dose	[kGy]	5	2	1.3	0,37 kGy/s
Flow rate	[m^3/min]	0.61	1.2	0.35	0,05
Cost of treatment	[\$US/m^3]	1.1 (*)	1.2	0.34	0,25

* – amortization and overhead not included

Estimated investment and exploitation costs of industrial facility for wastewater treatment (equipped with one 1 MeV, 400 kW electron accelerator) is displayed in Table XIII. Taking into account accelerator cost including installation, training and the cost related to the building construction, irradiation chamber and other spending the total investment cost can be evaluated.

The exploitation cost consists of variable and fixed costs. If the bank credit is adopted (8% for 20 years) and electricity cost 0.05 $US/kWh is applied the annual exploitation cost will amount bellow $US 1.0 M. Estimated costs for wastewater electron beam treatment process in above conditions operating with dose 2 kGy can be estimated of 0.25 $US/t. The facility throughput can be increased and unit cost of wastewater treatment slightly decreased by application of more powerful accelerators (5 MeV; 1 MW), which are under development.

TABLE XIII. ESTIMATED COST FOR WATER ELECTRON BEAM TREATMENT

Initial capital (US$)		
Accelerator	1 MeV; 400 kW	5 MeV; 1 MW
Total	3,700,000	6,600,000
Accelerator, spare parts, installation and training Accelerator building Waste water treatment chamber, piping and auxiliary equipment	2,200,000 700,000 800,000	4,600,000 800,000 1,200,000
Fixed costs		
Capital amortization, equipment maintenance, administration	428,919	744,071
Variable costs		
Labor, electricity, Equipment maintenance	500,400	1,140,000
Total annual costs	929,319	1,884,071
Annual throughput	3,650,000	9,125,000
Total cost per tone [$US/t]	0,25	0.2

Remarks:

- The operating schedule: 3 shifts per day; 24 h/day; 7 days/week; 365 day/year with availability 96% what corresponds to 8,410 h/year of facility operation.
- The capital cost is assumed to be financed at 8% interest for 20 years period.
- The electricity cost 0.05 US$/kWh.
- Average salary 600 US$/month. That includes: net salary, social security and welfare costs, pension funds, direct and indirect cost of training, payroll taxes and addition costs that occur in connection with employment,
- Dose 2 kGy and beam utilization 60%.

Higher process efficiency can be obtained by implementation lower dose level. Significant improvement of economical factors can be obtained for dose level lower than 1 kGy. Combined processes like ozone-electron beam treatment or specific properties and contamination of irradiated wastewater may allow apply the dose 0.2 kGy according to some literature data.

Industrial demonstration plant for treating wastewater from dyeing process is under construction in Korea. The main objectives are as follow: demonstrate technological and economical advantages of water and wastewater treatment by ionizing radiation and promote the commercialization of e-beam wastewater treatment process. Particularly amount of chemical reagent will be decreased up to 50%, efficiency of biological treatment will be improved by 30% and retention time in biological treatment will be decreased. Radiation unit will be combined with existing biological treatment facility. Maximum flow rate of 10,000 m3/day with one 1MeV, 400 kW accelerator will be achieved. Construction should be finished within 17 months in the middle of 2005.

5. SEWAGE SLUDGE IRRADIATION

Sludge irradiation facility can be built as additional systems to the conventional wastewater treatment plant, fore the sole purpose of disinfection. Liquid sludge irradiators can be designed to

operate in batch or continuous flow mode with the use of pipes or pumping. Irradiation of dewatered or dry sludge is carried out in facilities similar to those used in medical product sterilization equipped with conveyor system. Electron accelerators present capabilities are very well suited for continuous treatment of the wastes, in dewatered, dried or prepackaged form with appropriate thickness under the beam of accelerated electrons at a steady rate. Fig. 2 shows depth dose distribution in one side (single layer) sewage irradiation process. Calculation was performed with application of ModeRTL 2.1 computer program.

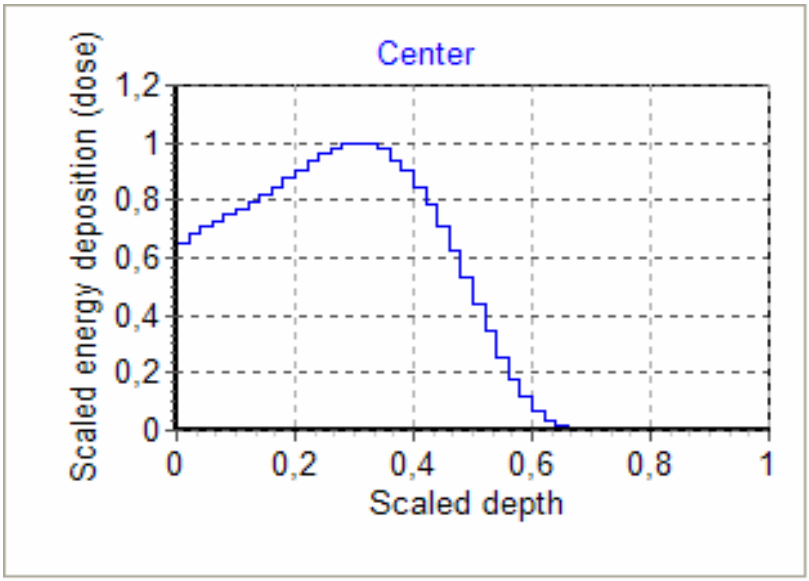

FIG. 5. Depth dose distribution in one side (single layer) sewage sludge irradiation process: electron energy 5 MeV; beam current 10 mA; surface dose 12 kGy; max. dose 18.5 kGy; depth of sewage sludge layer 5 cm (density 0.8 g/cm^3); useful depth 2.3 cm; scan length 160 cm; distance between window and sludge surface 15 cm; speed 9,1 cm/s.

The sewage treatment station for Polish city Otwock has been designed on the basis of the research performed by Institute of Nuclear Chemistry and Technology and Institute of Environmental Protection [7, 8]. Dewatered sewage sludge containing 30% of dry matter has been spread on the transporter and disinfected by electron irradiation with the dose 5 kGy. Capacity of the installation is 70 t/day, what corresponds to wastewater stream 48,000 m3/day. The capital cost of radiation processing unit was estimated on 4.0 M$, what mainly depends on the cost of accelerator and its building.

The City of Edmonton invited entries for a competition regarding Sludge Management Project in 1993 [9, 10]. The work should involved turn-key operation to process anaerobically treated digested sludge and for the off-side disposal or marketing of the processed end product. Heavy metal or toxic chemicals were well bellow prescribed levels for Edmonton sludge. The design team prepared the proposals consisting dewatering, air drying, sterilization, nutrient enhancement, and marketing of a variety of organic-based fertilizer product. The 10 MeV and 50 kW accelerator was foreseen to introduce electron treatment of the sludge at a rate 23,000 dry t/y (63 t/d) what represents a mass throughput 97 t/day. Dried product starts in a hopper, for which a controlled mass flow rate is measured. The product passes in front of the accelerator, in line with long axis of the beam.

The processing channel 5 cm high and 20 cm wide is used and dose rate 15 kGy is apply. The product enters a second hopper in preparation for shipping. An option to provide dewatering enhancement from electron processing was also proposed. The row sludge would be irradiated before it is sent to the dewatering system. The interesting aspect of the details Edmonton facility analysis is that capital cost of dewatering process was estimated as higher (80 $US/dry t) than those which is related to irradiation facility (49 $US/dry t). Finally sewage sludge electron beam treatment process

has not been yet implemented on industrial scale. Table XIV illustrates evaluation economical aspects of irradiation process performed with application of different accelerators. Higher investment cost related to the price of accelerators is compensated by bigger beam power of the accelerators as it can be concluded from presented data. Low cost, effective, high power and high energy accelerators will be adequate technical and economical solution for sewage sludge electron beam treatment process.

TABLE XIV. ESTIMATED COST FOR SEWAGE SLUDGE ELECTRON BEAM TREATMENT (60% facility utilization; exploitation during 7300 h/y)

Irradiated waste material	Sewage sludge 30% ss	Sewage sludge 35% ss	Sewage sludge 35% ss
One accelerator Max. productivity	27 t/h	9 t/h	90 t/h
Capacity in tons:			
Per hour [t/h]	56	12.5	
Per day [t/d]	1,120	250	
Per year[t/y]	408,800	91,250	912,500
Accelerator type	Rhodotron TT 300	ILU 10	Under construction
Manufacturer	IBA, Belgium	INP, Russia	Vivirad, France
Electron energy [MeV]	10	5	5
Beam power [kW]	3x150	2x50	1x1000
Dose [kGy]	12	12	12
Investment cost [$US]	25,500,000	4,364,000	8,800,000
Exploitation cost [$US]	3,662,800	919,500	2,000,000
Unit cost [$/t]	9.0	10.1	2.2

6. FINAL REMARKS

The most important feature of radiation processing is relatively high capital cost of an irradiation facility. This means that capital amortization is a major item in the operating cost. Appropriate accelerator selection should be performed to meet all technical and economical conditions for successful process implementation. The unit costs tend to decrease as the throughputs increase. Optimization of electron beam utilization by proper arrangement of irradiation unit increases productivity and reduces unit operation cost. Significant unit cost reduction can be obtained for implementation lower dose level.

Some general conclusions can be formed on the base of experience gained from research and pilot plant exploitation and large facility projects devoted to electron beam application in the field of environment protection:

- Sufficiently high facility throughput allows obtain relatively low unit cost what increase competitiveness radiation processing against more conventional methods.
- Demonstration full scale facility for radiation treatment is needed to overcome conservative attitude industry and government institution in new technology adoption.
- The government policy should promote radiation technology applied for environmental application when risks and benefits are equally important.

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