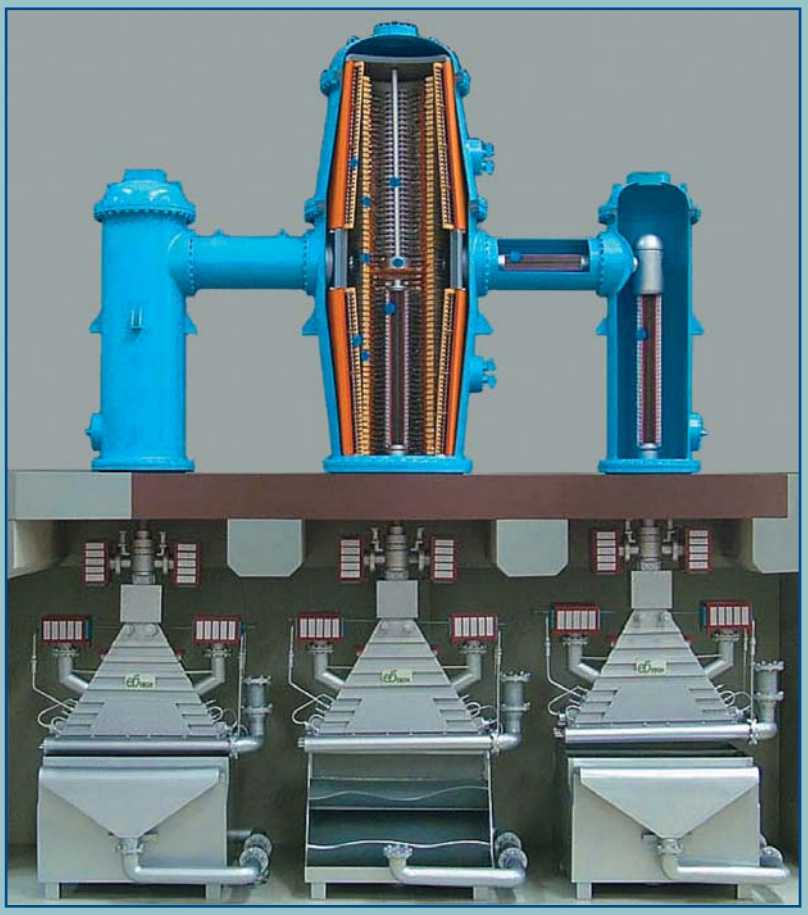
RADIATION PROCESSING: ENVIRONMENTAL APPLICATIONS



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RADIATION PROCESSING: ENVIRONMENTAL APPLICATIONS

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FOREWORD

A number of radiation processing technologies aimed at ensuring the safety of gaseous and liquid effluents discharged to the environment have been developed in recent years. Pilot and industrial scale installations have demonstrated that radiation processing based technologies for flue gas treatment (SO_x and NO_x removal), wastewater purification and sludge treatment can help to mitigate environmental degradation. Countries around the world, including many developing countries, have expressed great interest in the use of radiation technology to process effluents, particularly to remediate polluted waters and wastewater. This publication provides a summary of information relevant to radiation processing for environmental applications. It is expected to serve as a basis for the preparation of guidelines for and feasibility studies of the further implementation of radiation processing technologies, and to play an important role in promoting these technologies worldwide.

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Chapter 1

INTRODUCTION

In recent years, the problems of environmental damage and degradation of natural resources have received increasing attention throughout the world. Population growth, higher standards of living, increased urbanization and enhanced industrial activities all contribute to environmental degradation. For example, fossil fuels — including coal, natural gas, petroleum, shale oil and bitumen — are the main primary sources of heat and electrical energy production, and are responsible for a large number and amount of pollutants emitted to the atmosphere via exhaust gases from industry, power stations, residential heating systems and vehicles. All of these fuels are composed of major constituents such as carbon, hydrogen and oxygen, and other components including sulphur and nitrogen compounds and metals. During the combustion process, different pollutants are emitted, such as fly ash (containing diverse trace elements (heavy metals)), SO_x (including SO_2 and SO_3), NO_x (including NO_2 and NO) and volatile organic compounds (VOCs). Air pollution caused by particulate matter and other pollutants not only directly impacts the atmospheric environment but also contaminates water and soil, leading to their degradation. Wet and dry deposition of inorganic pollutants leads to acidification of the environment. These phenomena have a negative impact on human health and on vegetation.

Widespread forest damage due to atmospheric pollution has been reported in Europe and North America. Many cultivated plants are also affected by pollutants, especially in the early stages of growth. Mechanisms of pollutant transformation in the atmosphere are described by environmental chemistry. Photochemistry plays an important role in these transformations. In the presence of water vapour, SO_x and NO_x are oxidized, forming sulphuric and nitric acids. Fog and droplets result in so-called acid rain (i.e. acid precipitation). In recent years, investigations have shown that VOC emissions to the atmosphere can cause stratospheric ozone layer depletion and ground level photochemical ozone formation, and can have toxic or carcinogenic effects on human health. These emissions contribute to the global greenhouse effect, adding a new dimension to the environmental degradation resulting from the burning of fossil fuels.

Ironically, coal, the dirtiest fuel among the natural hydrocarbons, is expected to remain the principal fossil fuel for the next two centuries (Table 1). Its increased use will be necessary to meet the rising energy demands of both developed and developing countries. Thus, there is an urgent need to develop

TABLE 1. ESTIMATED NET INCREASE IN FUEL CONSUMPTION, 1995–2020

Fuel	Power (GW)		
	Installed in 1995	Estimated 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

technologies that reduce or minimize the pollution associated with the use of coal.

The dramatic increase in the world population (Table 2), combined with industrialization, urbanization, agricultural intensification and water intensive lifestyles, is expected to result in a global water supply crisis. Currently, about 20% of the world population lacks access to safe drinking water. While water is a renewable resource, it is also a finite resource. Only 3% of the world's water is fresh, of which one third is inaccessible.

TABLE 2. WORLD POPULATION (10^9) BY REGION, 1980–2020

Region	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[a]	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
South Asia	909.5	1146.0	1937.9
Total	4347.9	5293.4	8091.7

[a] Commonwealth of Independent States.

Global freshwater consumption rose sixfold between 1900 and 1995. At the same time as consumption rates are increasing, water resources in surface and underground water bodies are being polluted with industrial and municipal wastes. Until a few decades ago, most of the wastes discharged to water bodies were animal and human excreta, and other organic components from industry. 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 bodies. However, increasing urbanization over the past two centuries has been accompanied by an expansion of sewage collection systems with inadequate or no treatment. Liquid waste loads have become so large that the self-purification capacity of receiving bodies of water downstream of large populations can no longer prevent adverse effects on water quality. These wastes now constitute significant sources of water pollution. Industrial effluents carry chemical contaminants such as heavy metals, organic pollutants, petrochemicals, pesticides and dyes, while the discharge of sewage and sludge gives rise to microbiological contamination of water bodies. Some pollutants are synthesized in situ, as, for example, chloro-organic compounds arising from the application of chlorine for water and wastewater disinfection. The discharge of industrial effluents, sewage and sludge into water bodies is responsible for infection risks, health effects caused by contaminated drinking water and offensive odours.

Approximately one third of the world's population (Fig. 1) currently lives in countries with moderate to high water supply stress — areas where water consumption is more than 10% of the available renewable freshwater supply. If the present consumption patterns continue, by 2025 two thirds of the human population will live under water stress conditions. In terms of quantity and quality, the declining state of the world's freshwater resources may prove to be the dominant issue on the global environmental and development agendas of this century. In the decades to come, water security, like food security, will become a major national and regional priority in many areas of the world. There is thus a need to develop improved technologies that can control the pollution of this precious resource.

As these examples illustrate, humankind's environmental problems are no longer merely local or regional, but have become continental in scope. Economically and technically feasible technologies for controlling pollution from gaseous emissions and liquid effluent streams are being sought by experts working in a variety of areas, including radiation technology.

Since the discovery of high energy radiation more than one hundred years ago, radiation's ability to modify physicochemical properties of materials has found many applications. The use of radiation technology applying gamma sources and electron accelerators for the treatment of materials is well

established. Worldwide, there are over 200 industrial gamma irradiators and 1300 industrial electron accelerators in use for applications such as the sterilization of medical instruments, food irradiation and polymer processing. Radiation processing is a well established, multibillion dollar global industry that provides unique high value products for humankind in an environmentally friendly manner.

Over the past few decades, extensive research has been carried out concerning the use of radiation technology for environmental remediation.

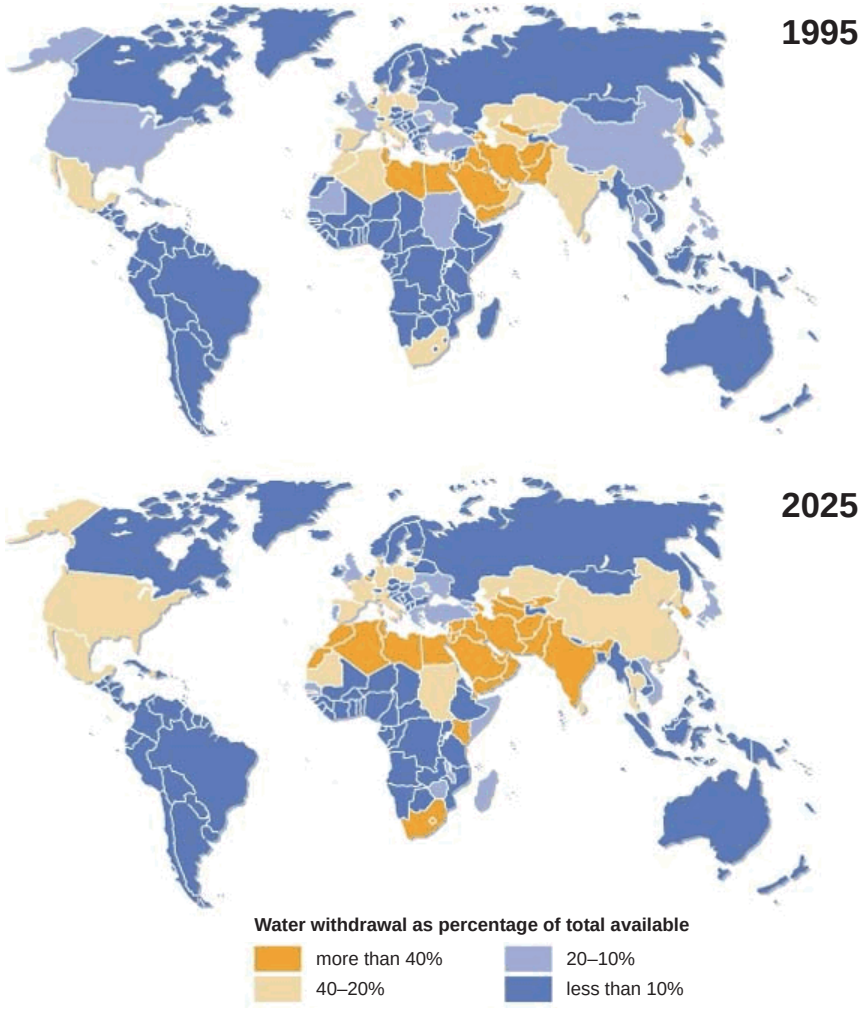

FIG. 1. Global water stress according to the Global Environment Outlook 2000 [1.1].

This work includes the application of radiation technology for simultaneous removal of SO_x and NO_x from flue gases, purification of drinking water, wastewater treatment and disinfection of sewage sludge for use in agriculture.

1.1. FLUE GAS TREATMENT

Electron beam technology for flue gas treatment was developed in Japan in the early 1970s. The process was later used at pilot scale plants in Germany, Japan, the Republic of Korea and the United States of America. In Poland, the process has been demonstrated at the Kawęczyn power station in a pilot installation with a throughput of 20 000 Nm^3/h using two electron accelerators (50 kW and 700 keV each).

Recently, a pilot installation with a throughput of 10 000 Nm^3/h and three accelerators (30 kW and 800 keV each) was constructed at the Maritsa East 2 thermal power plant in Bulgaria to treat SO_x gases from the combustion of low grade, high sulphur lignite coal. The plant has demonstrated very good process parameters, and the efficiency of pollutant removal ranges from 87 to 97% for SO_x and from 85 to 90% for NO_x. The by-product yield is of good quality, with a moisture content of less than 1%. The nitrogen content is approximately 21% or higher, which is the value recommended for use in commercial fertilizer. Ammonium sulphate makes up 96–97% of the by-product, with ammonium nitrate accounting for another 2%.

Industrial scale electron beam flue gas treatment installations are currently in operation in coal fired plants in Poland and China. The accelerators installed at the Pomorzany electric power station in Poland have a capacity of more than 1 MW, making it the largest irradiation facility ever built. The Pomorzany plant treats approximately 270 000 Nm^3 of flue gas per hour. The removal of SO_x and NO_x is highly efficient (up to 95% for SO_x and up to 70% for NO_x), and the by-product is a high quality fertilizer stock. The total investment costs of the Pomorzany installation were approximately $20 million. The capital costs of the installation at the plant in Chengdu, China, which was designed to treat SO_2 (using accelerators with low power capacity), were approximately $11 million.

Another possible application of the technology is for the removal of VOCs and polycyclic aromatic hydrocarbons (PAHs), for example, in flue gas treatment facilities of municipal waste incinerator plants.

1.2. WASTEWATER TREATMENT

Radiation processing of wastewater is non-chemical and makes use of the rapid formation of short lived reactive particles that can interact with a wide range of pollutants. Such reactive radicals are strong oxidizing or reducing agents that can transform the pollutants in liquid wastes.

In Daegu, the Republic of Korea, a pilot scale plant equipped with an electron beam accelerator was constructed to treat 1000 m^3 of textile dyeing wastewater per day. Its successful operation led to the construction of an industrial scale plant for treating 10 000 m^3/d on the same site. On the basis of data obtained in laboratory and pilot experiments, suitable doses were determined to be approximately 1–2 kGy for a throughput of 10 000 m^3/d. Therefore, an accelerator with 400 kW of power was installed, increasing the cost effectiveness and compactness of the plant. The cost of this high power accelerator was approximately $2.0–2.5 million; the cost of building materials, piping, other equipment and construction was estimated at $1.0–1.5 million. Considering the additional costs of taxes, insurance and documentation (approximately $0.5 million), the overall cost of the plant was approximately $4.0–4.5 million.

1.3. SEWAGE TREATMENT

Sewage is water borne waste from domestic premises and industry. Since it carries human waste, it is a source of various human pathogens that need to be controlled for safe disposal. The cleanup of sewage is a multistage process that includes primary and secondary treatment. Primary treatment removes the heaviest of the solid materials; the secondary stage includes the activated sludge and trickling filter processes. Sewage sludge, also known as biosolids, is the solid waste yield after completion of the secondary stage of wastewater treatment. The annual production of sewage sludge has been increasing around the world as stricter clean water laws have begun to take effect. The United States of America and the countries of Europe together produce approximately 13 800 Mt of dry sewage sludge per year. This sludge is a rich source of many micronutrients and a valuable source of fixed nitrogen, making it a valuable fertilizer.

Currently, about 41% of the sludge produced in the United States of America is utilized for land application; in European countries, utilization for land application ranges from 13% in Austria to approximately 80% in Luxembourg and Portugal. The United Kingdom utilizes about 55% and France about 50% of the generated sludge for land application. The presence

of pathogenic microorganisms in the sewage sludge has been a source of concern regarding its use in agricultural applications. Currently, heat and lime treatment are the methods most commonly used for processing sewage sludge; however, other processes are being explored. Sewage sludge irradiation is a very promising technology for sludge treatment and has been approved by the US Environmental Protection Agency as a method for producing 'class A' sludge that is safe for agricultural use.

A plant for liquid sludge treatment using gamma radiation from a ^{60}Co gamma source has been in operation in Vadodara, India, since 1992. The plant is designed to treat 110 m^3 of sludge from a conventional treatment plant per day. The plant's operational experience has shown that the process is simple, effective and easy to integrate into an existing sewage treatment plant, and that the radiation treated sludge can be used as a fertilizer in agriculture. Similarly, an electron beam accelerator can also be used to treat dewatered sludge.

REFERENCE TO CHAPTER 1

[1.1] UNITED NATIONS ENVIRONMENT PROGRAMME, Global Environment Outlook 2000, UNEP, Nairobi (1999).

Chapter 2

RADIATION PROCESSING OF GASEOUS SYSTEMS

Among the conventional technologies for flue gas treatment aimed at SO_2 and NO_x emission control are wet, dry and semi-dry flue gas desulphurization (FGD) and selective catalytic reduction (SCR). Volatile organic compounds can be adsorbed on particulate matter, but this process has rarely been useful for low concentrations of hydrocarbons. These conventional gas cleaning technologies are complex chemical processes that result in the generation of wastewater, gypsum and depleted catalyst [2.1–2.5]. Electron beam technology is among the most promising advanced technologies available for the treatment of flue gases, particularly in the light of the drawbacks of conventional technologies.

Electron beam technology is a dry scrubbing process for simultaneous SO_2 and NO_x removal with no generation of waste. Irradiation of flue gases using an electron beam can bring about chemical changes for the ready removal of SO_2 and NO_x. The main components of flue gas are N_2, O_2, H_2O and CO_2, with much lower concentrations of SO_x and NO_x. Ammonia may be present as an additive. The gas components absorb radiation energy in proportion to their mass fraction in the mixture. Fast moving electrons slow down, and secondary electrons are formed that play an important role in overall energy transfer.

In electron beam irradiation, fast electrons interact with the gas, creating divergent ions and radicals. The 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, oxidizing radicals $^{\bullet}OH$ and $HO_2^{\bullet}$ and excited species such as $O(^3P)$ are the most important products formed. These excited species react in a variety of ways such as ion–molecule reactions, neutralization reactions and dimerization [2.6]. The SO_2, NO, NO_2 and NH_3 present cannot compete with these reactions because of their very low concentrations, but they react with N, O, OH and HO_2 radicals.

After humidification and cooling, flue gases flow to a reaction chamber, where they are irradiated by the electron beam. Ammonia is injected upstream of the irradiation chamber. There are several pathways for oxidation of NO. In the case of electron beam treatment, the most common reactions are as follows [2.7]:

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

$$O(^3P) + O_2 + M \rightarrow O_3 + M$$

$$NO + O_3 + M \rightarrow NO_2 + O_2 + M$$

$$NO + HO_2^{\bullet} + M \rightarrow NO_2 + {}^{\bullet}OH + M$$

After oxidation, the NO_2 is converted to nitric acid in the reaction with the $^{\bullet}OH$ radical according to the following equation:

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

The HNO_3 aerosol reacts with the NH_3, forming ammonium nitrate through a neutralization reaction written as:

$$HNO_3 + NH_3 \rightarrow NH_4NO_3$$

The NO is partly reduced to nitrogen gas.

There also can be several pathways for SO_2 oxidation, depending on the conditions. In electron beam treatment, the most important are radiothermal and thermal processes [2.8].

The radiothermal reaction proceeds through radical oxidation of SO_2 via the following equation:

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

The HSO_3 is then converted to ammonium sulphate through 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:

$$SO_2 + 2NH_3 \rightarrow (NH_3)_2SO_2$$

$$(NH_3)_2SO_2 \xrightarrow{O_2, H_2O} (NH_4)_2SO_4$$

The total amount of SO_2 removed is the combined yield of the thermal and radiothermal reactions, written as follows [2.9, 2.10]:

$$\eta_{SO_2} = \eta_1(\phi,T) + \eta_2(D, \alpha_{NH_3}, T)$$

The yield of the thermal reaction depends on the temperature and humidity, and decreases with an increase in temperature. The yield of the radiothermal reaction depends on the dose, temperature and ammonia stoichiometry. The main parameter in NO_x removal is the irradiation dose, with other parameters playing minor roles in the process. In industrial applications, dose distribution and gas flow distribution are important as well [2.11].

2.1. RADIATION TREATMENT OF SULPHUR DIOXIDE AND NITROGEN OXIDES

The removal of SO_2 using electron beams from a linear accelerator (linac) (2–12 MeV, 1.2 kW) was first demonstrated by Japanese scientists in the early 1970s. Irradiation of flue gas at a dose of 50 kGy and a temperature of 100°C resulted in the conversion of SO_2 to an aerosol of sulphuric acid droplets that were easily collected [2.12]. In these experiments, the Ebara Corporation 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_4NO_3 that was usable as a fertilizer.

Using the Ebara process, two larger scale plants were constructed in Indianapolis, United States of America [2.13], and Karlsruhe, Germany [2.14]. The Indianapolis plant was equipped with two electron beam accelerators (0.8 MeV, 160 kW each) and had a gas flow capacity of $1.6–3.2 \times 10^4$ m^3/h, with the gas containing 1000 ppm SO_2 and 400 ppm NO_x. In Karlsruhe, two electron accelerators (0.3 MeV, 180 kW total power) were used to treat $1–2 \times 10^4$ m^3 of flue gas per hour, with the gas containing 50–500 ppm SO_2 and 300–500 ppm NO_x.

The engineering design technology for electric utility applications was further developed at pilot plants in Nagoya, Japan [2.15], and Kawęczyn, Poland [2.16]. At the plant in Kawęczyn, many new engineering solutions were applied, including longitudinal double gas irradiation, the use of an air curtain to separate the secondary window from corrosive flue gases and modifications of the humidification/ammonia system (high enthalpy water or steam injection, aqueous ammonia injection). The results obtained (Fig. 2) confirm the previously discussed physicochemistry of the process.

A high dose is required for NO_x removal, while SO_x can be removed at optimized design conditions, with low energy consumption. These new

developments have led to economic and technical feasibility improvements and to the installation of industrial scale plants.

An industrial scale plant in Chengdu, China, constructed by the Ebara Corporation, was designed primarily for SO_2 removal; accelerators with 320 kW of power were installed to treat 300 000 m^3 of flue gas per hour (Fig. 3). The reported removal efficiency is 80% for SO_x and 20% for NO_x [2.17].

Another industrial scale installation for the treatment of flue gases is located at the Pomorzany electric power station in Szczecin, Poland [2.18] (Figs 4–6). The installation purifies flue gases from two Benson boilers (65 MW(e) and 100 MW(th) each). The maximum flow rate of the gases is 270 000 Nm^3/h, and the total electron beam power exceeds 1 MW. There are two reaction chambers, each with a nominal gas flow rate of 135 000 Nm^3/h. Each chamber is irradiated by two accelerators (260 kW, 700 keV each) installed in series.

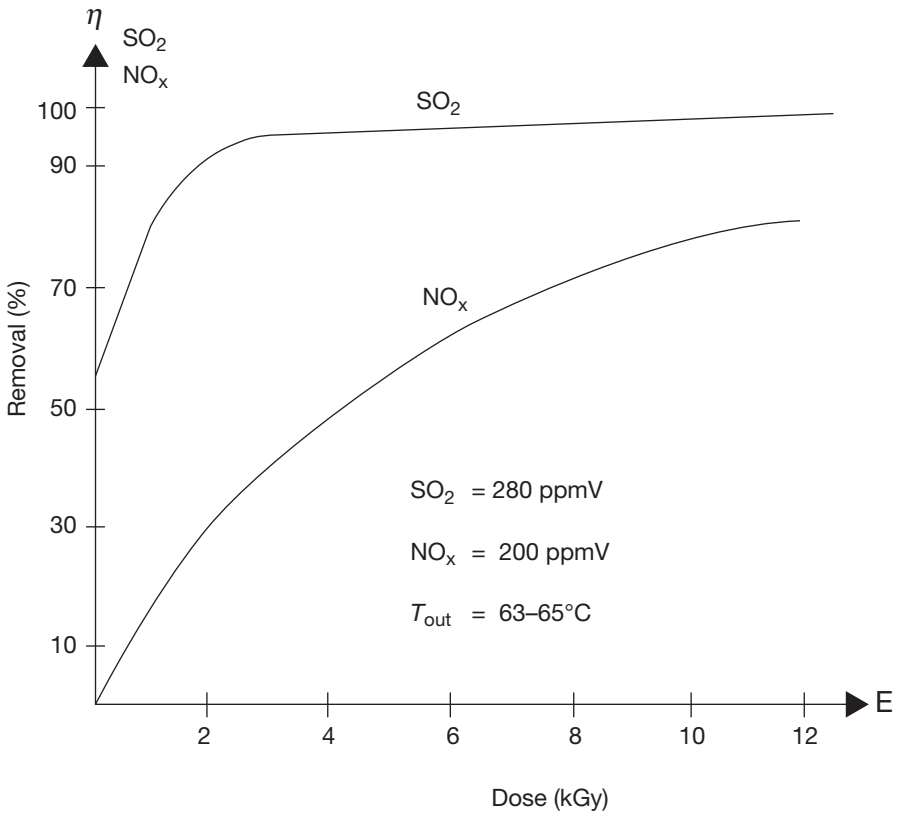

FIG. 2. Efficiency of SO_2 and NO_x removal versus dose: results from pilot plant experiments and theoretical calculations [2.8].

FIG. 3. Flue gas treatment plant in Chengdu, China.

FIG. 4. The Pomorzany electric power station in Szczecin, Poland.

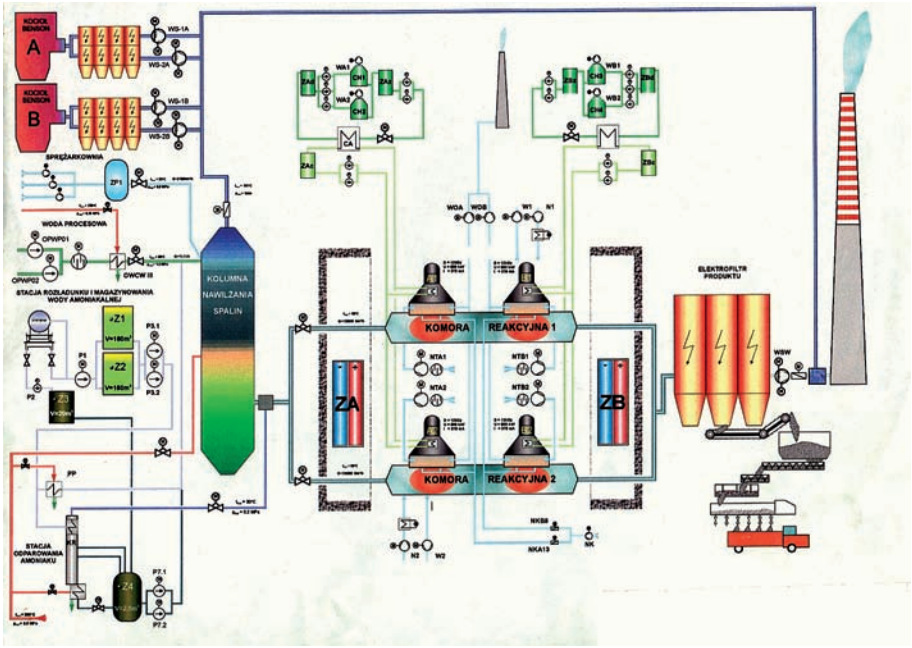

FIG. 5. Flow diagram of the industrial plant at the Pomorzany electric power station [2.19].

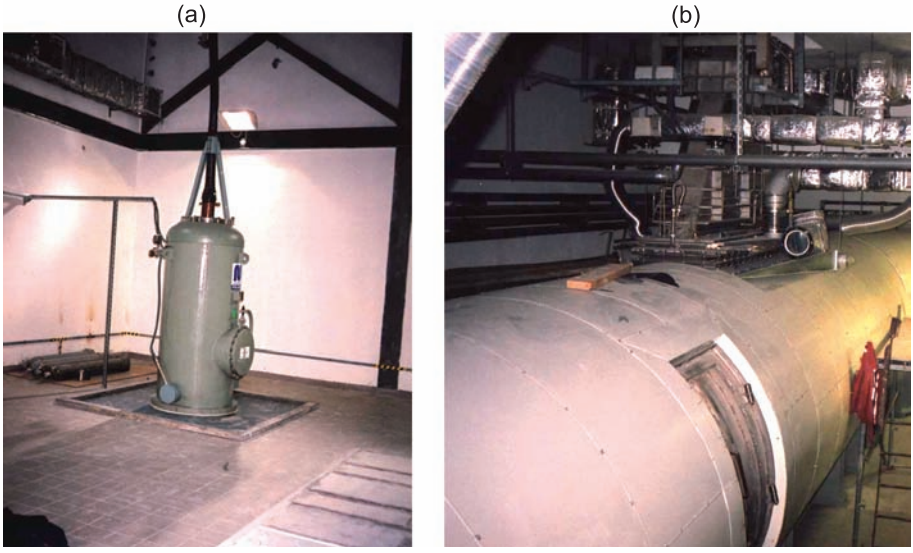

FIG. 6. One of four accelerators (a) and the gas irradiation process vessel (b) at the Pomorzany installation [2.19].

The applied dose is in the range of 7–12 kGy. At these doses, the removal efficiency approaches 80–90% for SO_2 and 50–60% for NO_x. The by-product is collected by an electrostatic precipitator and shipped to a fertilizer plant.

2.1.1. Industrial plant construction

A feasibility study of an industrial scale electron beam process for a 350 MW power plant treating a flue gas flow of 1 500 000 m^3/h (normal temperature and pressure, NTP) has been carried out, with comprehensive engineering and cost evaluations [2.19]. This study shows that large scale electron beam plants for flue gas treatment have cost advantages over plants using conventional technologies.

A typical industrial installation consists of four main parts:

(1) A flue gas conditioning unit;
(2) An ammonia storage and feed system;
(3) A process vessel with an accelerator system;
(4) A by-product collection and storage unit.

2.1.1.1. Flue gas conditioning unit

The function of the flue gas conditioning unit is to cool and humidify flue gas. The temperature of the flue gas from boilers or other combustion systems is too high and the humidity level is too low for optimal process performance. These levels are adjusted using water spray and/or steam injection. The optimal temperature after cooling is in the range of 60–75ºC, and optimal humidity is 12–14%. These optimal levels can be achieved by evaporation of water only if the inlet gas temperature (T_0) is high (170–190ºC). Reaching the optimal levels also depends on the humidity of the inlet flue gas; the higher the humidity of the gas, the lower T_0 can be.

Different types of water spray system can be used to adjust these levels, including:

(a) A dry bottom spray column;
(b) A cooling column with circulating water;
(c) Spray in the inlet duct.

The use of a dry bottom spray column is a simple process, but it has some disadvantages. In such a column, the water needs to be sprayed in very fine droplets (<100 μm in diameter); thus, two medium air–water nozzles must be applied and the consumption of compressed air is high. When the temperature

of the inlet flue gas is below 180ºC and the humidity of the flue gas after cooling is too low, the water content is increased by injecting steam into the column, which increases the operating costs of the process. An advantage of using the dry bottom column is that the walls remain dry, and thus corrosion is not significant.

Cooling columns with circulating water were tested in the Kawęczyn, Shin Nagoya and Maritsa East 2 electron beam pilot plants, and have been used in the Chengdu electric plant. This type of water spray system is preferable when the plant is designed mainly for SO_2 removal and low gas temperatures after humidification are required. In cases where the temperature of the inlet flue gas is low, the enthalpy is too low to ensure adequate humidification of the gas and the circulated water stream has to be heated. In this case, a significant amount of additional heat (steam) is needed for evaporation of water. The temperature of the outlet gas, T_1, is normally in the range of 55–65ºC. If the temperature of the gas is too low, part of the flue gas stream can be bypassed to maintain the proper temperature.

The advantages of the cooling column with circulating water are that:

- Water droplets can be much larger than with a dry bottom spray column;
- Only one medium water nozzle needs to be installed;
- The amount of compressed air needed can be reduced;
- The fly ash content of the flue gas is significantly reduced.

The disadvantages of this solution are:

- The serious corrosion of column elements;
- The generation of wastewater containing collected particulates and absorbed HCl, SO_2 and SO_3.

A third possibility is to spray water so that it evaporates into the flue gas within the inlet duct. This solution can be applied if the duct is long and its cross-section is large. In such a case, it is possible to apply finely atomized water, and systems with full evaporation of water or with water circulation are available. This process has not yet been tested for use in electron beam technology. It may reduce investment costs by 5–7%.

2.1.1.2. Ammonia storage and feed system

Ammonia is preferably stored in pressurized vessels in the form of liquid (anhydrous) ammonia. Before injection into the flue gas, the liquid ammonia is

vaporized in a small evaporator. Ammonia that comes into contact with SO_2 forms inorganic solids that can deposit on the nozzles of the ammonia feed system. Special devices need to be installed close to the spray nozzles to prevent these deposits from plugging nozzle orifices. Some ammonium salts may also settle on the walls of ducts after the ammonia is introduced. In another ammonia injection system that has been tested by the Ebara Corporation, ammonia is mixed with hot air and sprayed using gas–water nozzles at the inlet to the process vessel. The water dissolves the ammonium salts and prevents the formation of deposits. This system is preferable for low temperature operations aimed primarily at SO_2 removal [2.20]. Another option is the use of aqueous ammonia as an ammonia source. This option has not been tested at the pilot scale, but it has been applied at the industrial scale at the Pomorzany electric power station.

Ammonia can be introduced into the system in two ways. In the first, gaseous ammonia is desorbed from aqueous ammonia in a distillation column and introduced into the flue gas duct. In the second, ammonia is evaporated in a cooling column through a nozzle system. Preliminary tests have indicated that the best choice is a combined method in which part of the aqueous ammonia is sprayed into a cooling column and the remainder is added downstream in a gaseous form. As the use of aqueous ammonia increases the operating and investment costs, the use of anhydrous ammonia is preferred. Mixing ammonia in the flue gas stream usually does not create any problems, provided the ammonia is injected at several points, since the gas stream flow is turbulent and good mixing conditions prevail in the duct.

2.1.1.3. Process vessel with accelerator system

This system consists of:

- Accelerators equipped with:
  - Power supplies;
  - A water cooling system;
  - Window cooling systems;
  - Windows;
- A reaction chamber;
- X ray shielding with a ventilation system.

Accelerators are the most notable and advanced equipment applied in electron beam technology. Only a few companies in the world manufacture high power electron accelerators suitable for this technology. The price of such a unit depends on its power. The required accelerator power is proportional to

the mass flow of flue gas and the radiation dose needed for efficient SO_2 and NO_x removal. Generally, a dose of 4–5 kGy is required for efficient SO_2 reduction, whereas a dose of 8–12 kGy is usually necessary for efficient NO_x reduction. Therefore, if high NO_x removal efficiency is required, the application of combined (i.e. simultaneous removal) processes should be taken into consideration. For example, the application of low NO_x burners can reduce the concentration of NO_x upstream of the electron beam installation. Several high power accelerators have been constructed to meet this requirement, and technical solutions are still being improved. The main parameters of the accelerators are the electron energy and the beam current. Typically, accelerators with an electron energy of up to 800 keV and a beam current of up to 500 mA are applied in the electron beam flue gas treatment process.

The electrons are introduced into the process vessel through a titanium foil window (e.g. 50 μm thick), passing through with the gas flow. The process vessel is an empty cylinder with a circular or rectangular cross-section. The dimensions of the process vessel should match the penetration range of electrons in the irradiated gas. The dose deposition in the cross-section of the reactor is not uniform. Theoretical studies carried out at the Institute of Nuclear Chemistry and Technology in Poland have shown that the use of non-uniform flow, which increases the gas velocity in the space where higher energy is deposited, can improve removal efficiencies by 10–14% [2.21].

Furthermore, the air used for cooling the windows is strongly irradiated, resulting in the generation of some ozone. Up to now, this ozonized air has been discharged to the atmosphere via the ventilation system. It appears that this air could be advantageously used in the process, or in the combustion chamber of the boiler.

2.1.1.4. By-product collection and storage unit

After the injection of ammonia and the excitation of gas molecules by electrons, the radical and chemical reactions lead to the formation of ammonium salts that condense in the form of very fine submicron aerosols. These aerosols, which are of a chemical composition valuable in fertilizer production, are typically filtered from the off-gases in an electrostatic precipitator (ESP).

Among the filters that have been tested at various pilot plants are:

— ESPs [2.22];
— Bag filters [2.23];
— Gravel bed filters [2.24];
— Venturi scrubbers.

The first two of these filters use dry filtration, and the last two use wet filtration. Because wet filtration is very complicated and generates a quantity of wastewater that must be treated before being discharged, it is not recommended in the case of dry technology application.

Bag filters have very good aerosol particle removal efficiency and provide additional removal of SO_2 within the filter cake layer. However, they are not applied at the industrial scale because they cannot be adequately cleaned: the by-product is sticky, and the pulse jet method used in cleaning does not guarantee full regeneration of the bag surface. Alternative methods of filter bag regeneration exist and can be applied, and membrane filters might be used in the future.

Electrostatic precipitators have proved their usability for electron beam technology in industrial scale plants. The product precipitated in ESPs is hygroscopic; if it becomes wet, corrosion may occur. Therefore, to prevent condensation of water, the bottom of the ESP is electrically heated. The walls are made of stainless steel and are insulated. The same material is recommended for electrodes.

As previously mentioned, the SO_2 and NO_x removal efficiencies depend strongly on the process conditions. The highest efficiency obtained for SO_2 is 95%, while that for NO_x is about 70% (Fig. 7).

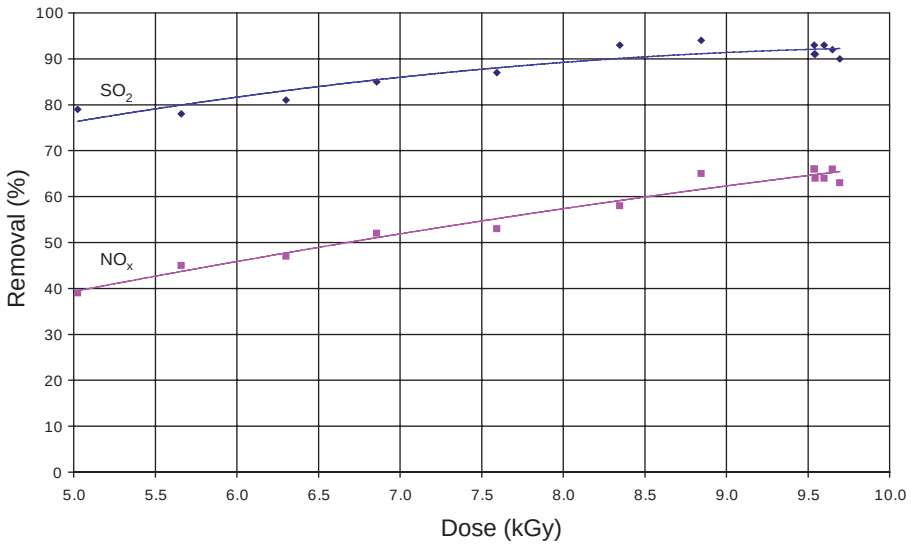

FIG. 7. Efficiency of SO_2 and NO_x removal versus dose; results obtained at an industrial installation (SO_2 inlet concentration of 1500–1630 mg/Nm^3; NO_x inlet concentration of 470–540 mg/Nm^3).

Industrial results are in agreement with results previously reported based on pilot plant experiments and theoretical calculations. The data obtained during industrial operation confirm hypotheses on the impact of process parameters on NO_x and SO_2 removal efficiency. In the case of NO_x removal, the most important parameters are the radiation dose and the inlet concentration of NO_x. The total removal (in mg/Nm^3) increases with the inlet concentration of NO_x, while the relative removal (in %) decreases with an increase in this parameter. Ammonia stoichiometry has very little impact on NO_x removal (Fig. 8).

For SO_2 removal, a greater number of parameters affect pollutant removal efficiency. The most important among these is the temperature of the gas after humidification, which is due to the thermal reaction contribution. Another important factor is the radiation dose. Although humidity appears to have a major impact on efficiency, it is difficult to establish this beyond a doubt because of the strong correlation between humidity and the temperature of the process (wet bulb temperature is a factor of both these parameters). During water evaporation, the temperature decreases and the humidity increases. The ammonia stoichiometry ratio has an unexpectedly strong influence on SO_2 removal efficiency (Fig. 8); other factors such as flue gas flow rate and inlet concentration have much less of an impact in this regard. During experiments,

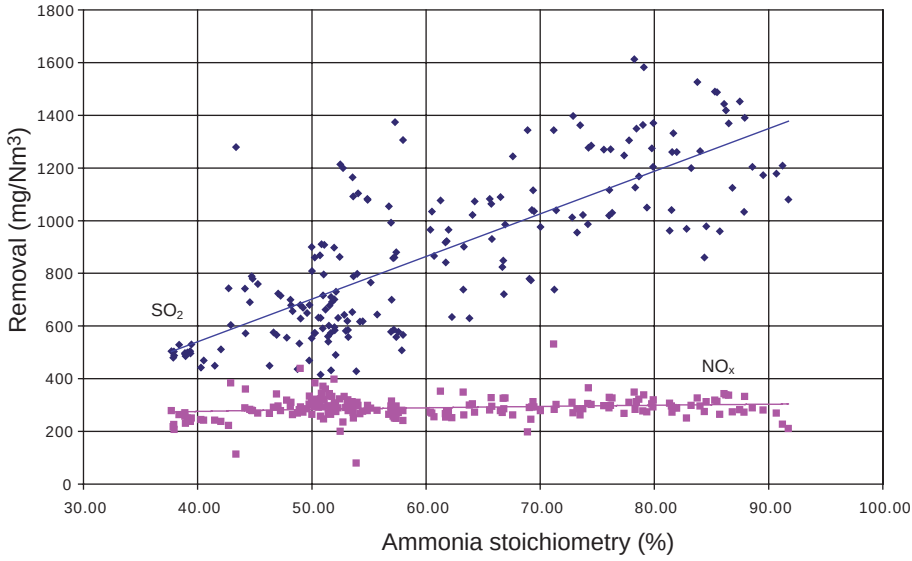

FIG. 8. Efficiency of SO_2 and NO_x removal versus ammonia stoichiometry; results obtained at an industrial installation.

it has been observed that the mode of ammonia injection also influences the removal efficiency, and thus has an impact on the entire process. It has also been observed that injecting part of the aqueous ammonia feed directly into the humidification tower increases SO_2 removal efficiency. This phenomenon is now under investigation [2.25].

2.1.2. Economics of the process

The conventional technology most often applied for flue gas treatment is a combination of wet FGD and SCR. The investment costs of retrofitting wet FGD installations are usually $80–250/kW(e), depending on the unit capacity and site conditions. In comparison, the investment costs of retrofitting SCR installations are $59–112/kW(e), depending on the plant size and the difficulty of retrofitting the facility [2.26]. For new facilities, the costs are $45–60/kW(e). According to Ref. [2.27], the combined costs of wet FGD and SCR for 50 and 300 MW(e) units are estimated to be $474/kW(e) and $270/kW(e), respectively.

The annual operating costs for wet FGD methods are about $2500–3000/MW(e), while those for SCR methods are $3800–4600/MW(e) [2.26]. Thus, the costs of removing both types of pollutant using conventional methods are $6300–7600/MW(e) annually. A comparison of the costs of different emission control methods for a 120 MW(e) unit is presented in Table 3.

2.2. RADIATION INDUCED REMOVAL OF VOCs FROM EXHAUST GASES

The primary reactions in the decomposition of VOCs by irradiation are similar to those in SO_2 and NO_x removal, namely, free radicals attacking organic compound chains or rings, causing VOC decomposition [2.28–2.30].

For the decomposition of chlorinated aliphatic hydrocarbons (e.g. chloroethylene), Cl dissociated secondary electron attachment and the reaction of Cl and OH radicals with VOCs play very important roles.

For aromatic hydrocarbons, VOC decomposition primarily involves the following steps:

(1) *Positive ion charge transfer reactions*

$$M^+ + RH = M + RH^+$$

where RH is a VOC, for example, benzene or PAHs.

TABLE 3. COSTS OF SELECTED EMISSION CONTROL METHODS FOR A RETROFITTED 120 MW(e) UNIT

Emission control method	Investment costs ($/kW(e))	Annual operating costs ($/MW(e))
Wet FGD	120	3000
SCR	110	4600
Wet FGD + SCR	230	7600
Electron beam flue gas treatment	160	7350

Because RH has a lower ionization energy (9.24 eV for benzene and <10 eV for PAHs) than most of the primary positive ions (>11 eV) formed above, part of the VOCs will be decomposed by rapid charge transfer reactions.

(2) *Radical–neutral particle reactions*

Hydroxyl radicals play a very important role in VOC decomposition, especially when the water concentration is approximately 10%. These radicals react with VOCs in two ways:

(i) Hydroxyl radical addition to the aromatic ring (e.g. toluene):

$^{\bullet}OH + C_6H_5CH_3 = {}^{\bullet}R1$

(ii) Hydrogen atom abstraction (for the alkyl substituted aromatic compounds) or hydrogen atom elimination (for benzene, naphthalene and the higher PAHs):

$C_6H_5CH_3 + {}^{\bullet}OH = {}^{\bullet}R2 + H_2O$ (hydrogen atom abstraction)

$C_6H_6 + {}^{\bullet}OH = C_6H_5OH + H$ (hydrogen atom elimination)

The radicals formed above ($^{\bullet}R1$, $^{\bullet}R2$) undergo very complex reactions such as O_2 addition, oxygen atom release, or the formation of aromatic -CHO (-aldehydes) or -OH compounds, or ring cleavage products:

$^{\bullet}R$ ($^{\bullet}R1$, $^{\bullet}R2$) + $O_2 = {}^{\bullet}RO_2$

$2^{\bullet}RO_2 = 2^{\bullet}RO + O_2$

$^{\bullet}RO_2 + NO = {}^{\bullet}RO + NO_2$

$^{\bullet}RO + O_2 = HO^{\bullet}_2$ + products (aromatic-CHO, -OH)

$^{\bullet}RO \rightarrow$ aliphatic products

The possibility of applying this process for dioxin removal from flue gases has been studied [2.31, 2.32], and recent pilot studies have demonstrated that the process is technically and economically feasible [2.33, 2.34].

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Chapter 3

RADIATION PROCESSING OF WASTEWATER

Rapid population growth and increased agricultural and industrial development have led to the generation of large quantities of polluted industrial and municipal wastewaters. The recognition that these polluted waters may pose a serious threat to humans has led technologists to seek cost effective technologies for their treatment. A variety of methods based on biological, chemical, photochemical and electrochemical processes are being explored for decomposing the chemical and biological contaminants present in these wastewaters [3.1]. Radiation technologists have been investigating the use of high energy radiation for their treatment. The primary advantage of radiation processing over alternatives is that the reactive species are generated in situ during the radiolysis process without the addition of any chemicals. Several pilot scale and a number of industrial scale wastewater treatment plants based on radiation technology are in operation or under construction [3.2, 3.3]. The results of practical applications have confirmed that radiation technology can be easily and effectively utilized for treating large quantities of wastewater [3.2–3.6].

3.1. WATER RADIOLYSIS

Radiation processing in wastewater treatment is an additive free process that uses the short lived reactive species formed during the radiolysis of water for efficient decomposition of the pollutants therein. For practical treatment of wastewater, radiation processing offers the following advantages:

— Strong reducing and oxidizing agents;
— Universality and interchangeability of redox agents;
— A variety of paths for pollutant conversion;
— Process controllability;
— A wide choice of equipment and technological regimes;
— Compatibility with conventional methods.

High energy irradiation produces instantaneous radiolytic transformations through energy transfer from high energy photons or accelerated electrons to the orbital electrons of water molecules. Absorbed energy disturbs the electron system of the molecule and results in the breakage of interatomic

bonds [3.6]. Hydrated electrons (e^-_{aq}), H atoms, $^\bullet OH$ and $HO^\bullet_2$ radicals, H_2O_2 and H_2 are the most important products of the fragmentation and primary interactions (radiolysis products):

$$H_2O \xrightarrow{EB} e^-_{aq},\ ^\bullet H,\ ^\bullet OH, HO^\bullet_2, H_2O_2, H_2$$

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

High reactivity is characteristic of water radiolysis products [3.7]. As a rule, these products' reactions with impurities in water typically require less than 1 μs. At the same time, the reactivity of different radiolytic products is quite different. Hydrogen peroxide and $^\bullet OH$ and $HO^\bullet_2$ radicals are oxidizing species, while H atoms and e^-_{aq} are chemical reducing in nature [3.2]. The simultaneous existence of strong oxidants and strong reductants within wastewater under treatment is remarkable and is one of the important characteristics of the radiation processing technique. The main properties of oxidizing species produced during the radiolysis of water are given in Table 4 [3.2, 3.6–3.8]. The $^\bullet OH$ radical, by virtue of the high radiation chemical yield of the formation as well as its high oxidation potential, is the most predominant species. In fact, the oxidation power of $^\bullet OH$ is much higher than that of conventional industrial oxidants such as Cl_2, O_2, HOCl, $KMnO_4$, $K_2Cr_2O_7$ and O_3.

TABLE 4. PROPERTIES OF OXIDIZING PRODUCTS OF WATER RADIOLYSIS [3.6]

Particle	Hydroxyl radical $\dot{O}H$ ($\dot{O}^-$)	Hydrogen peroxide H_2O_2	Per-hydroxyl radical $H\dot{O}_2$
Formation	$H_2O^+ + H_2O \rightarrow H_3O^+ + \dot{O}H$ $H_2O \xrightarrow{EB} \dot{H} + \dot{O}H$	$2\dot{O}H \rightarrow H_2O_2$ $2H\dot{O}_2 \rightarrow H_2O_2 + O_2$	$\dot{O}H + H_2O_2 \rightarrow H_2O + H\dot{O}_2$ $\dot{H} + O_2 \rightarrow H\dot{O}_2$
Radiation chemical yield after completion of spur reactions G (molecules/100 eV)	3.0 at pH0.4 2.8 at pH7 3.0 at pH13 ($\dot{O}^-$) (≈ 6.0)	0.8 at pH0.4 0.75 at pH7	0.02 at pH0.4
Diffusion coefficient D (10^{-5} cm^2/s)	2.3	1.3	1.7
Standard redox potential E° (V)	2.1 ($OH^-_{aq} \leftrightarrow {}^\bullet OH + e^-$) 2.8 ($H_2O \leftrightarrow {}^\bullet OH + H^+ + e^-$)	0.7 ($H_2O_2 \leftrightarrow O_2 + 2H^+ + e^-$) 1.5 ($H_2O_2 \leftrightarrow HO_2 + H^+ + e^-$)	0.4 ($HO_2 \leftrightarrow O_2 + H^+ + e^-$)

The typical rate constants for the reactions of $^{\bullet}OH$ radicals with nitriles, amides, carboxylic acids, esters and carbonyls are from 1×10^7 to 1×10^9 $dm^3 \cdot mol^{-1} \cdot s^{-1}$; however, with saturated hydrocarbons, alcohols, ethers, amines, alkenes, aromatics, pyrimidines, thiols and disulphides, the rate constants can be greater than 1×10^9 $dm^3 \cdot mol^{-1} \cdot s^{-1}$ [3.7]. During its reactions with ions, the $^{\bullet}OH$ radical captures an electron to form a hydroxyl ion:

$$^{\bullet}OH + Fe^{2+} \rightarrow OH^- + Fe^{3+}$$
$$^{\bullet}OH + NO_2^- \rightarrow OH^- + NO_2^{\bullet}$$

whereas in reactions with unsaturated hydrocarbons, $^{\bullet}OH$ adds to the double bond:

$$^{\bullet}OH + CH_2 = CH_2 \rightarrow HOCH_2 - {}^{\bullet}CH_2$$

and, upon reaction with alkyl compounds, the $^{\bullet}OH$ captures a hydrogen atom to produce a water molecule:

$$^{\bullet}OH + CH_3COCH_3 \rightarrow H_2O + CH_3CO\dot{C}H_2$$

The main properties of reducing particles are given in Table 5 [3.2–3.8]. A hydrated electron is a stronger reductant than a H atom. In reactions involving cations of metals, e_{aq}^- is able to produce neutral atoms and ions having anomalous valency. Typical reactions of e_{aq}^- consist in e^- addition to the reagent, but can be subdivided into two types [3.2]. The first type includes simple addition such as:

$$e_{aq}^- + Tl^+ \rightarrow Tl^0$$
$$e_{aq}^- + NO_3^- \rightarrow NO_3^{2-}$$
$$e_{aq}^- + C_{10}H_8 \rightarrow C_{10}H_8^-$$

The second type includes the dissociative reactions of e^- addition:

$$e_{aq}^- + RCl \rightarrow R^{\bullet} + Cl^-$$
$$e_{aq}^- + C(NO_2)_4 \rightarrow C(NO_2)_3^- + NO_2^{\bullet}$$

TABLE 5. PROPERTIES OF REDUCING PRODUCTS OF WATER RADIOLYSIS [3.6]

Particle	Hydrated electron e^-_{aq}	$^{\bullet}H$ atom
Formation	$H_2O \xrightarrow{EB} H_2O^+ + e^-$ $e^- + nH_2O \rightarrow e^-_{aq}$	$e^-_{aq} + H_2O \rightarrow \overset{\bullet}{H} + OH^-$ $e^-_{aq} + H_3O^+ \rightarrow \overset{\bullet}{H} + H_2O$ $H_2O \xrightarrow{EB} \overset{\bullet}{H} + \overset{\bullet}{O}H$
Radiation chemical yield after completion of spur reactions G (molecules/100 eV)	2.8 at pH7 3.8 at pH13 (≈ 4.5)	0.6 at pH7 3.8 at pH2 0.15 at pH13 (≈ 1.5)
Diffusion coefficient D (10^{-5} cm^2/s)	4.96	7.0
Standard redox potential E° (V)	–2.87 ($e^-_{aq} \leftrightarrow H_2O + e^-$)	–2.3 ($H^{\bullet} \leftrightarrow H^+_{aq} + e^-$)
Rate constant ($dm^3 \cdot mol^{-1} \cdot s^{-1}$) of reactions with:		
Saturated hydrocarbons, alcohols, ethers	$<10^5$	10^7–10^9
Alkenes	$<10^5$	$>10^9$
Aromatics	10^5–10^7	$>10^9$
Pyrimidines, nitro-, bromo-, thiols, disulfides	$>10^9$	10^7–10^9
Nitriles, amides, carboxylic acids, esters, chlorohydrocarbons	10^7–10^9	10^5–10^7
Iodohydrocarbons	$>10^9$	$>10^9$
Cd(II), Pb(II), Cr(III)	$>10^{10}$	$<10^6$
CrO_4^{2-}, $Cr_2O_7^{2-}$, Hg(II)	$>10^{10}$	$>10^9$

Hydrogen atoms have a low radiation chemical yield in neutral solutions. Usually, the reaction of a H atom with an inorganic compound occurs via an electron transfer:

$$^{\bullet}H + Cu^{2+} \rightarrow Cu^{+} + H^{+}$$

In its reactions with organic compounds, the H atom demonstrates higher reactivity than does e^-_{aq}. Reactions with unsaturated hydrocarbons typically consist of the addition of $^{\bullet}H$ to a double bond:

$$^{\bullet}H + CH_2 = CH_2 \rightarrow CH_3 - {}^{\bullet}CH_2$$

Upon reacting with alkyl compounds, $^{\bullet}H$ produces a H molecule and an alkyl radical:

$$^{\bullet}H + CH_3-CH_3 \rightarrow H_2 + CH_3-\dot{C}H_2$$

3.2. MAIN PATHWAYS OF POLLUTANT DEGRADATION

Radiation processing aims at the degradation of pollutants at a faster rate than with conventional processes. Radiation processing of wastewater generally has maximum efficiency at pollutant concentrations of 10^{-3} mol/L (~100 ppm) or less. The treatment of such wastewater is simple, requires a low dose (~1 kGy or less) and provides almost complete elimination of odour, colour, taste and turbidity.

The radiation processing of polluted water containing specific contaminants may require the creation of special conditions to achieve the predominant type of transformation — reduction, oxidation, addition or removal of functional groups, aggregation, disintegration, etc. However, in general, pollutant transformation involves the following pathways: chain oxidation, formation of insoluble compounds, coagulation of colloids and enhancement of pollutant biodegradability.

3.2.1. Chain oxidation

Chain oxidation constitutes one of the most efficient processes realized in radiation processing of wastewater. As a rule, saturation of wastewater with air is needed for chain oxidation. Radiolytic oxidation at a moderate dose leads to the formation of carbonyl, carboxyl, hydroxyl and/or peroxide groups in organic molecules [3.2, 3.3]. The conditions of irradiation can be specifically chosen to achieve chain oxidation of various pollutants. The general mechanism of chain oxidation consists of the following stages:

(a) Initiation :

$$RH + {}^{\bullet}OH \rightarrow R^{\bullet} + H_2O$$
$$R^{\bullet} + O_2 \rightarrow RO_2^{\bullet}$$

(b) Propagation :

$$RO_2^{\bullet} + RH \rightarrow ROOH + R^{\bullet}$$
$$ROOH \rightarrow RO^{\bullet} + {}^{\bullet}OH$$
$$RO_2^{\bullet} + R^{\bullet} \rightarrow ROOR$$

(c) Termination : $2R^{\bullet} \rightarrow R_2$

$2RO_2^{\bullet} \rightarrow ROOR + O_2$

The products of radiolytic oxidation differ from the initial pollutants and are essential in terms of their physicochemical properties and increased ability to undergo biodegradation.

3.2.2. Formation of insoluble compounds

Many organic pollutants can be transformed into insoluble or sparingly soluble compounds upon irradiation, owing to the formation of high molecular weight products [3.9, 3.10]. Usually, organic compounds having a molecular weight of more than 200 amu have low solubility. The formation of large molecules during radiolysis is realized by the recombination of intermediate radicals formed upon radiolytic transformation of pollutants:

$$2R_2HC-\dot{C}R_2 \begin{cases} \rightarrow R_2HC-CR_2-CR_2-CHR_2 \\ \rightarrow R_2HC-CHR_2+R_2C=CR_2 \end{cases}$$

The pollutant radicals formed may undergo dimerization or disproportionation [3.2]. In water, the dimerization dominates because of the cage effect and the rapid delocalization of excess energy by inner redistribution along C–C bonds or its transfer to molecules in the medium. Recombination of radicals competes with the reaction of radical addition to unsaturated hydrocarbons:

$$R_2HC-\dot{C}R_2+R_2C=CR_2 \rightarrow R_2HC-CR_2-R_2C-\dot{C}R_2$$

A high concentration of unsaturated molecules in wastewater can result in the formation of a polymer by chain repetition of the above reaction [3.2].

3.2.3. Coagulation of colloids

Exposure to high energy radiation can result in both stabilization and coagulation of colloidal solutions [3.2, 3.9, 3.10]. The coagulation effect is realizable in colloidal solutions of metals, hydroxides, sulphides, carbon, proteins, polymers, etc., owing to charge interaction and the chemical actions of water radiolysis products on micelles:

$$M^{n+} + me^{-}_{aq} \rightarrow M^{(n-m)+}$$

$$M^{n+} + {}^{\bullet}OH \rightarrow OH^{-} + M^{(n+1)+}$$

$$M^{n+} + mH^{\bullet} \rightarrow mH^{+} + M^{(n-m)+}$$

Therefore, $^{\bullet}OH$ radicals and e^{-}_{aq} reduce the stability of positively charged colloids. The effect of the $^{\bullet}OH$ radicals depends on the primary charge of the micelles and their absorbability by the OH^{-} ions. The presence of H atoms in irradiated colloidal wastewater provides stabilization for positively charged colloids but leads to coagulation for negatively charged ones.

3.2.4. Enhancement of pollutant biodegradability

A large number of substances such as hard surfactants, lignin and pesticides cannot be degraded by conventional biochemical methods; thus they escape decomposition in biological treatment. Biodegradation of wastewater depends on the oxidation level and structure of pollutants, and preliminary oxidation and fragmentation of biologically resistant molecules improve their biodegradability. The mechanism of radiolytic oxidation mentioned above makes possible the transformation of various pollutants. Research and industrial treatments testify to significant improvement of pollutant biodegradability after radiation oxidation of aerated wastewater [3.2, 3.4, 3.5, 3.11]. Usually, a dose of about 1–2 kGy is necessary for complete transformation of pollutants from a biologically resistant to a biodegradable state.

3.3. RADIATION PROCESSING OF INDUSTRIAL WASTEWATER

As described in Section 3.1, high energy irradiation of aqueous solutions generates highly reactive radicals that can interact with a wide range of pollutants. The first studies of the radiation treatment of wastes, principally for disinfection, were carried out in the 1950s. In the 1960s, these studies were extended to the purification of water and wastewater. Laboratory research on industrial wastewaters and polluted groundwater was conducted in the 1970s and 1980s, and in the 1990s several pilot plants, including mobile electron beam facilities, were built for extended research. Aqueous effluents that can be treated by irradiation fall into two groups: industrial wastewater, and natural and contaminated water (including effluents from municipal treatment plants). The main differences between the two groups are the concentration of pollutants and the level of microbial infection, both of which are higher in the

first group of effluents; however, disinfection is the main focus for the second group.

Owing to the great variety of wastewaters generated by different industries, there currently is no universal treatment process for industrial wastewater. The main focus of radiation processing is to convert non-biodegradable pollutants into biodegradable species. Extensive studies have been carried out of the purification of industrial wastewater by radiation processing, although generally at the laboratory and, to a lesser extent, the pilot plant scale. The first such full scale application was reported for the purification of wastewater generated at the Voronezh synthetic rubber plant in the Russian Federation.

The Voronezh plant has two purification lines, each equipped with an accelerator of 50 kW capacity to convert the non-biodegradable emulsifier, or Nekal, present in the plant wastes into a biodegradable form. Nekal is a mixture of isomeric isobutyl-naphthalene sulphonates. The dose required for complete decomposition of 1×10^{-3} mol·dm^{-3} of Nekal in aqueous solution is 0.3 MGy. However, it is only necessary to remove the alkyl or sulphonates group to render the molecule biodegradable, since the products (naphthalene sulphonates, alkyl naphthalene and naphthalene) are biodegradable; the initial G value is about 2 molecules/100 eV [3.2]. On the basis of observation following the radiation treatment, it was discovered that the products of Nekal can be readily degraded further by tertiary biological treatment. The installation treats up to 2000 m^3 of effluent per day.

Another full scale application is combined radiation and biological treatment of textile dyeing wastewater. Initial laboratory investigation indicated that electron beam treatment of textile dyeing wastewater was a prospective means for its purification [3.4]. These improvements resulted in decolorizing and destructive oxidation of organic impurities with low doses of radiation (~1–2 kGy). Radiation treatment combined with biological treatment reduced the chemical reagent consumption and treatment time, and increased the flow capacity. On the basis of laboratory tests indicating the method's feasibility, a pilot scale plant for treating 1000 m^3 of textile dyeing wastewater per day using an electron beam was constructed in Daegu, the Republic of Korea, and has been in continuous operation since 1998 (Fig. 9).

The pilot plant demonstrated reductions of chemical reagent consumption and retention time with an increase in the efficiency of COD(Cr) and BOD5 removal of up to 30–40%. Figure 10 shows the additional effect of electron beam irradiation on the biological treatment of wastewater; Fig. 10(a) presents the kinetics of biotreatment of irradiated and non-irradiated wastewater, while Fig. 10(b) gives the dose effect on combined electron

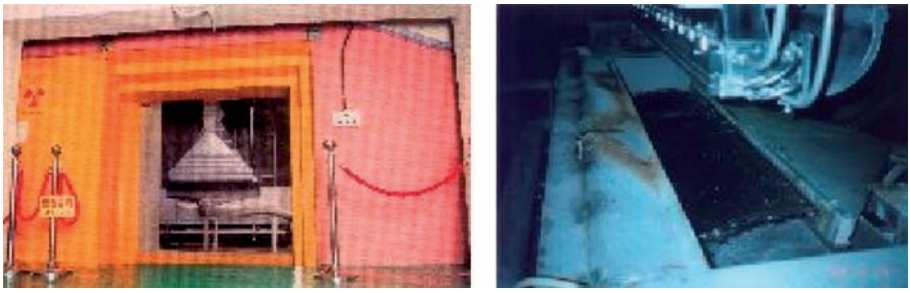

FIG. 9. Pilot scale wastewater plant and wastewater injection system in Daegu, the Republic of Korea.

beam–biological treatment. The increase in removal efficiency after radiation treatment is due to the radiolytic transformation of biodegradable compounds into more readily degradable forms.

On the basis of data obtained from pilot plant operation, it was decided to build an industrial scale plant. Construction of the plant started in 2003, with completion scheduled for 2005. This plant is located in a textile dyeing industrial complex and has the capacity to treat 10 000 m^3 of wastewater per day using a 1 MeV, 400 kW accelerator, combined with an existing biotreatment facility (Figs 11, 12).

Radiation treatment of textile dyeing wastewater and several dyes has also been actively studied in Brazil, Hungary and Turkey [3.12–3.15]. A pilot

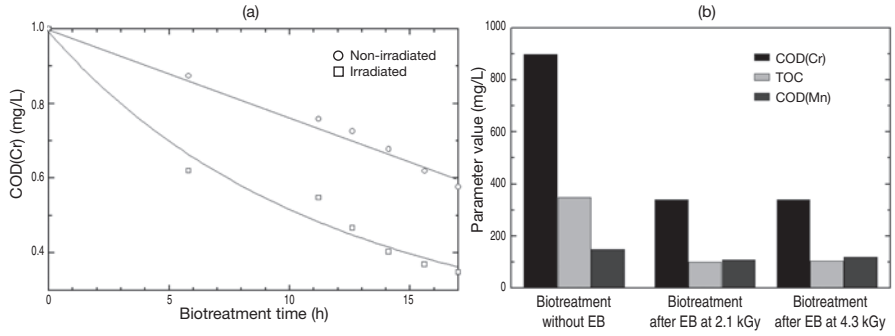

FIG. 10. Effect of electron beam irradiation on the biological treatment of wastewater: (a) kinetics of biotreatment of irradiated and non-irradiated wastewater; (b) dose effect on combined electron beam (EB) and biological treatment.

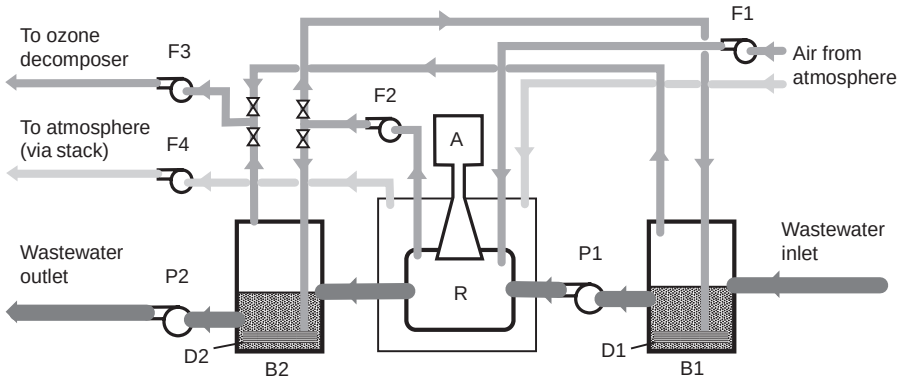

FIG. 11. Industrial scale textile dyeing wastewater treatment plant: F1–F4 are air fans, P1 and P2 are water pumps, D1 and D2 are diffusers, A is the accelerator, R is the reactor, and B1 and B2 are the primary and secondary basins, respectively.

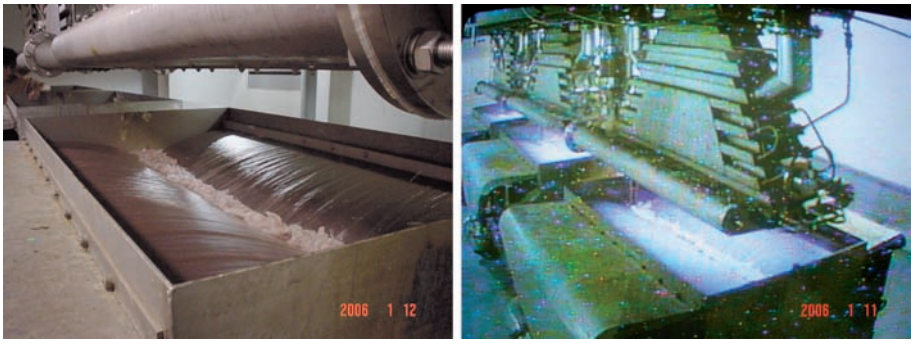

FIG. 12. Industrial electron beam plant for treatment of textile dyeing wastewater in the Republic of Korea.

scale wastewater treatment plant was set up at the Institute of Energy and Nuclear Research (IPEN) in Brazil to study the efficiency of removal and degradation of toxic and refractory pollutants present in industrial wastewater (Fig. 13). Combined biological and radiation treatment of domestic sewage and sludge was carried out to investigate disinfection. Radiation processing of many other industrial wastewater samples collected at the public wastewater treatment plant in São Paulo State was also tested. For industrial wastewater from chemical industries, a dose of 20 kGy was necessary to degrade about 99% of the organic compounds [3.12].

FIG. 13. Pilot scale wastewater treatment plant at IPEN, Brazil.

3.4. RECLAMATION OF EFFLUENT FROM MUNICIPAL WASTEWATER TREATMENT PLANTS

Population growth coupled with declining freshwater supplies has necessitated the development of technologies to reclaim potable water from wastewater. Providing sufficient potable water will be one of the critical challenges of the twenty-first century. The increasing levels of pollution and complexity of effluents from municipalities and industry demand effective technologies to reduce pollutants to the desired levels. The use of current wastewater treatment technologies for such reclamation often is not successful. Some progress has been made, however, in technical approaches to reliably producing potable water using wastewater reclamation technologies. Advanced wastewater treatment technologies are essential for the treatment of municipal wastewater to protect public health and to meet water quality criteria for the aquatic environment and for water recycling and reuse. Moreover, when reclaimed wastewater is to be used for human consumption, disinfection is absolutely crucial, and removal or inactivation of pathogenic organisms is a critical step in final treatment. Among the possible water treatment alternatives, radiation processing is a very effective option, as it can simultaneously degrade both the toxic organic compounds and the biological contaminants that are present.

Radiation disinfection of effluent from municipal wastewater treatment plants has been successfully demonstrated by a number of researchers. Studies have demonstrated that inactivation of faecal coliforms in secondary effluents from municipal wastewater plants can be achieved with radiation doses of less than 1 kGy (Fig. 14). While the water matrix has an adverse effect on the efficiency of conventional disinfectants, it generally has no effect on radiation processing for bacteria inactivation. Moreover, as radiation processing is technically much easier than conventional processes, it has a clear advantage over existing methods for municipal wastewater disinfection.

Reclamation of effluent from municipal wastewater treatment plants using radiation processing has been studied extensively in Austria, Brazil, Ecuador, Jordan and the Republic of Korea. In Austria, research has been carried out on the effect of oxygen on the radiation induced inactivation of selected microorganisms in water. A cost assessment based on a radiation dose of 1 kGy indicated that, for a plant capacity of about 1150 m^3/h, the cost of treating secondary effluent is about \$0.1/m^3, which is acceptable, considering the advantages that the radiation induced disinfection provides compared with conventional technologies [3.16]. In Jordan, a 99% reduction of the microbiological content of irradiated water was achieved at a dose of 2 kGy; to carry out the effective deactivation of nematode eggs, up to 3 kGy of irradiation dose was found to be required [3.17]. In Ecuador, irradiation with an electron accelerator resulted in the decontamination and disinfection of municipal

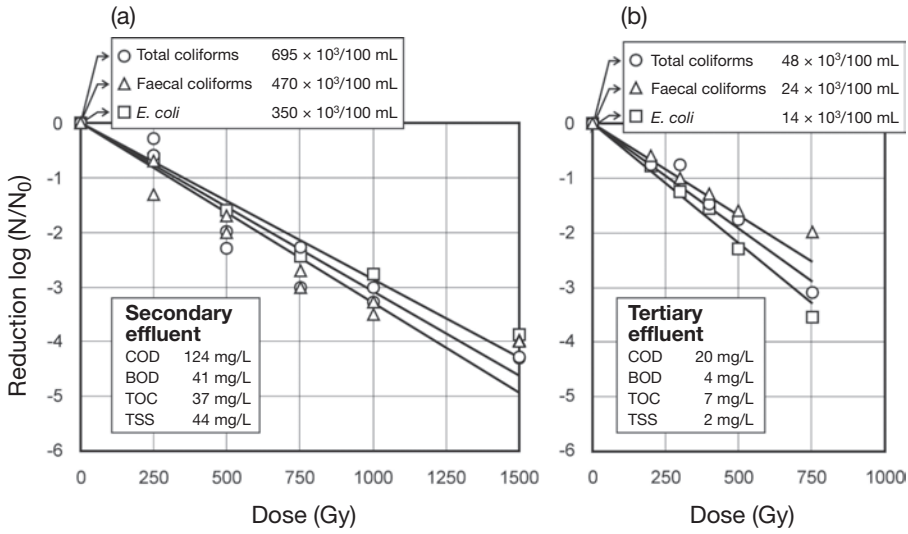

FIG. 14. Inactivation of selected coliforms in (a) secondary and (b) tertiary effluents by means of electron beam irradiation.

wastewater with reductions of 72% of COD, 80% of BOD, 72% of surfactants, and 100% of total microorganisms and faecal coliforms.

Regulations currently exist in some countries concerning the concentration of *E. coli* in secondary effluents, and similar regulations will be adopted by a significant number of countries in the near future. Given that wastewater processed by radiation meets such regulations, and that the process itself has advantages over conventional technologies, radiation processing is a highly attractive technology. However, at present there is no full scale radiation treatment plant in operation.

Radiation processing for effluent treatment is a beneficial alternative to chemical and ultraviolet treatment. Compared with chemical disinfection, it has proved to be a cleaner technology without the formation of hazardous by-products; compared with ultraviolet irradiation, electron beam irradiation is technically much simpler and is almost insensitive to colour, suspended solids or gas bubbles in the effluent stream, as well as to effluent composition and fouling characteristics. Moreover, it requires considerably less maintenance and is very easy to control. Studies of the remediation of contaminated groundwater using radiation have been conducted in the United States of America, and kinetic models and destruction mechanisms have been proposed [3.18]. Remediation of groundwater contaminated by pesticides has been studied in Poland and Turkey [3.19, 3.20].

3.5. RADIATION INDUCED REMOVAL OF HEAVY METAL IONS FROM WASTEWATER

The toxic metals present in industrial effluent streams include heavy metals such as lead, mercury, cadmium, nickel, silver, zinc and chromium. These heavy metals accumulate in soil and are eventually transferred to the human food chain. Exposure to ionizing radiation of aqueous effluents containing these heavy metals leads to the formation of free radicals, radical ions and stable products that subsequently react in the following manner [3.21–3.25]:

The hydrated electron (e^-_{aq}) acts as a strong reducing agent:

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

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

$Cr(VI) + e^-_{aq} \rightarrow Cr(V)$

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

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

$2Pb^+ \rightarrow Pb + Pb^{2+}$

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

$H^\bullet + Pb^+ \rightarrow PbH^+$

PbH^+ decays to produce Pb:

$2PbH^+ \rightarrow H_2 + Pb^{2+} + Pb$

(c) $HgCl_2 + e^-_{aq} \rightarrow HgCl + Cl^-$

$HgCl_2 + H^\bullet \rightarrow HgCl + Cl^- + H^+$

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

$2HgCl \rightarrow Hg_2Cl_2$

The $^\bullet OH$ radical is one of the powerful oxidizing species that lead to the transformation of metal ions to higher valence states [3.1]. However, because concentrations of heavy metals in wastewater are normally very low (in terms of parts per million), the process seems not to be technically feasible, since trace quantities of reduced metals have to be separated mechanically from the wastewater. For higher concentrations, chemical methods (precipitation, ion exchange) or physical methods (membranes, electrolysis) are more feasible from the economic and technical points of view.

3.6. ECONOMICS OF WASTEWATER TREATMENT

The key to the successful application of electron beam technology in environmental protection is economics. To compete with other processes in economic terms, potential uses of the electron beam system must aim to:

- Reduce the required doses;
- Improve efficiencies;
- Reduce the cost of electron beam facilities.

For wastewater such as contaminated groundwater and effluents from municipal plants with low pollutant levels, the treatment is simple. It requires a low dose and completely eliminates odour, colour, taste and turbidity with no more than a few kilograys of radiation. Highly polluted water — for example, industrial wastewater — may, however, require special conditions for irradiation. To reduce the required dose, it is essential to choose and optimize the right type of transformation, for example, reduction, oxidation, addition/removal of functional groups, aggregation, disintegration and application of useful additives that may act as sensitizers.

3.6.1. Combinations with conventional methods

Radiation processing may change various pollutant properties such as solubility, volatility, absorptivity and reactivity. The products formed via degradation of pollutants due to irradiation may be easier to oxidize, reduce or biodegrade when subsequently treated by conventional methods. Therefore, a synergistic effect can be expected when radiation processing is combined with other, conventional processes such as biological treatment [3.2–3.5, 3.11]. As mentioned in Sections 3.2 and 3.3, radiation processing converts many biologically resistant pollutants to biodegradable states. The treatment of textile dyeing wastewater is one example of combined electron beam and biological treatment (see Section 3.3).

The efficiency of separation processes — coagulation, flocculation and foam flotation of suspended solids — can also be improved by radiation processing [3.9–3.11, 3.26, 3.27]. A combination of radiation processing and separation methods is essential in expediting the treatment of wastewater from factories producing monomers and polymers (e.g. polyesters, polyvinylchloride). Radiation processing of such wastewater completes the formation of dispersed polymers and decomposes colloidal particles.

With the special preliminary addition of a monomer, an analogous combination of radiation processing and a separation stage are necessary for the treatment of wastewater [3.2–3.5]. Preliminary radiolytic oxidation improves the efficiency of electrodialysis. The combined action of accelerated electrons and electrodialysis is three to ten times greater than their separate actions. A similar effect has been shown using a combination of radiation processing and absorption employed as a post-treatment process. The reason for such influence is the change in the chemical nature and polarity of the functional groups and the size of pollutant molecules [3.2–3.6].

One way to increase the efficiency of radiation processing is to warm wastewater before treatment [3.2–3.5]. Many factories have warm wastewater as an effluent. In the electron beam process, a higher temperature results in

enhanced yield and faster removal of volatile and gaseous products of pollutant decomposition. Radiation processing of wastewater at a high temperature often has the synergistic effect of destroying spores, vegetative cells, viruses and enzymes. Increased surfactant decomposition is also observed. Combined ozone–electron beam treatment has also been applied in Austria for groundwater remediation [3.28]. Turbulent flow conditions of irradiated water have been demonstrated in a bench scale facility. A 3 mm thick water layer was treated successfully with 500 keV electrons (with a penetration range of 1.4 mm). It was found that the presence of ozone allowed a reduction of the dose from 370 to 45 Gy, with a decrease in the estimated cost of treatment from \$0.25 to \$0.07/m^3.

3.6.2. Technological regimes

With low doses of the order of a few kilograys, an accelerator can treat several thousand tons of wastewater per day. Therefore, the effectiveness of the wastewater delivery system and the efficiency of the accelerator itself are important process parameters. Ideally, the water delivery system will allow uniform dose distribution as well as the delivery of large amounts of wastewater for electron beam treatment. Although the accelerator is the source of the penetrating electrons, the interaction between the beam and the wastewater takes place inside a special reaction vessel — a reactor. Three main types of reactor have been used for practical electron beam treatment of wastewater (Fig. 15) [3.9–3.11, 3.26, 3.27, 3.29].

The first type (reactor (a) in Fig. 15) represents a reactor with perfect displacement. A nozzle injector forms the wastewater flow into a wide continuous water jet. This jet is irradiated by a transverse electron beam and is collected below in a tray. The design thickness of the water jet is tied to the beam energy, that is, to the penetrating power of the accelerated electrons. The width of the water jet corresponds to the width of the beam window. The bottom of the tray is always immersed in a layer of irradiated wastewater; this configuration is necessary, as it aids the penetration of the electrons by thinning and splitting the water jet. The water jet type of reactor makes possible electron beam treatment by powerful accelerators at high rates of wastewater flow.

The second type (reactor (b) in Fig. 15) is based on spraying. Wastewater is pumped through a sprayer to create a pattern of water impingement on the beam window. The spraying method provides suitable conditions for carrying out chain oxidation of pollutants in an air atmosphere. Wastewater spraying provides an extensive surface for contact between the wastewater being irradiated and air. Such contact supports the penetration of oxygen into the

wastewater and promotes the efficient combination of radiolytic ozone and electron beam treatment.

The upflow reactor type (reactor (c) in Fig. 15) represents perfect mixing. Wastewater moves by gravity inside a tray and mixes intensively with a gas or air upon irradiation. The tray can be made of metal, glass, ceramic or concrete. Gas is injected from below by special bubblers or through a porous reactor bottom. Tray space is usually divided into several sections by vertical partitions. Upflow irradiation devices for the electron beam wastewater treatment process have been demonstrated in a pilot plant in Brazil [3.30, 3.31].

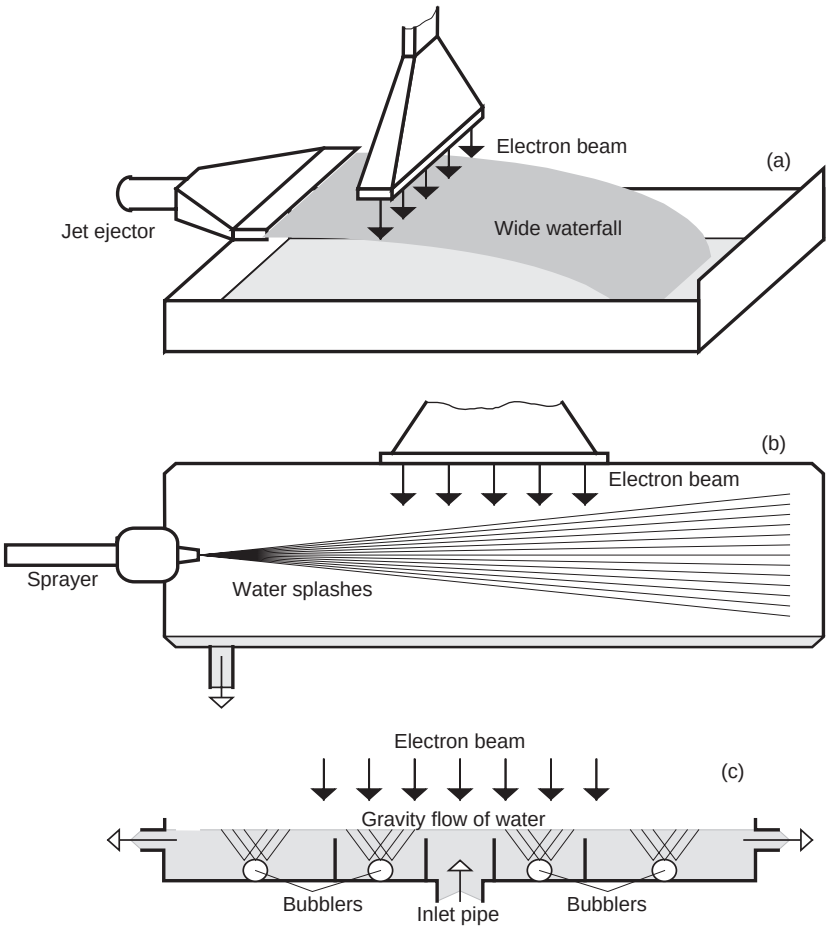

FIG. 15. Various types of wastewater reactor: (a) reactor with perfect displacement; (b) reactor based on spraying; (c) upflow reactor type.

An upflow delivery system significantly improves energy transfer to the wastewater stream and allows the application of accelerators with relatively low electron energies. An irradiation system efficiency of 67–76% was obtained in an optimized design configuration. In some experiments, a 40 μm thick layer of titanium foil was used to protect the accelerator window, which allowed the irradiation device to work as a closed system. The estimated process cost was found to be $\$1.2/m^3$ for a dose rate of 2 kGy, a flow rate of 70 m^3/h, an electron energy of 1.5 MeV and beam power of 60 kW.

3.7. FUTURE PROSPECTS

Rapid population growth combined with industrialization, urbanization and water intensive lifestyles has resulted in severe problems in wastewater management, especially in large cities. In many countries where industry is concentrated in urban areas, severe water pollution problems have arisen in most of the large cities. Hence, the treatment of such wastewater has become an important subject in the field of environmental engineering. At present, wastewater treatment with radiation processing has not found wide application, and it is used much less often than conventional methods. However, in recent years pilot plants and industrial scale studies have shown that radiation processing could occupy an important place in the future.

Already, radiation processing in combination with conventional methods has been shown to provide noticeable reductions in the time, area and power needed for wastewater treatment. Continuous emphasis on ecological standards will be an additional motivation for the elaboration and industrial application of radiation processing. Propagation of radiation processing can improve environmental protection and provide essential support in industrial development.

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Chapter 4

RADIATION PROCESSING OF SEWAGE SLUDGE

The sludge resulting from municipal wastewater treatment usually is in the form of a liquid or semisolid liquid that typically contains 0.25–12% solids by weight, depending on the operations and processes used. Of the components removed in wastewater treatment, sludge is by far the greatest in volume, and the problems associated with its processing and disposal are complex because:

- It is composed of the substances responsible for the offensive character of untreated wastewater;
- The portion of sludge produced from biological treatment and requiring disposal is composed of the organic matter contained in the wastewater, but in a form that can decompose and become offensive;
- Only a small portion of the sludge is solid matter.

The sludge can be used as a soil conditioner and as an additive to animal fodder. However, it contains bacteria, viruses and parasites (and possibly toxic compounds), and should be disinfected prior to any such use. Guidelines by the US Environmental Protection Agency (EPA) recommend that, in sewage sludge to be applied in agricultural practice, the number of *E. coli* bacteria (used as an indicator of the presence of pathogens) not exceed 1000 per gram of dry sludge [4.1].

Since large scale land application of sewage sludge is a relatively recent practice, this use 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 the development of technologies that can treat the sludge in a reliable, efficient and cost effective manner. Many researchers have shown that sewage sludge can be successfully disinfected by exposure to ionizing radiation, and that irradiation often facilitates dewatering by accelerating sedimentation and filtration [4.2–4.13].

4.1. TREATMENT OF SEWAGE SLUDGE USING IONIZING RADIATION

The sludge generated by sewage plants contains a high concentration of pathogens, which limits the reuse of this waste. Because sewage sludge is otherwise a rich source of plant nutrients, its disposal in its original form is an economic loss to society. However, further processing of the sludge to reduce the number of pathogens is necessary before sludge solids can be recycled or put to beneficial use. Thus, it is necessary to enhance the treatment process to ensure the removal of the pathogenic bacteria with a high degree of reliability.

The high energy ionizing radiation from radioactive sources such as ^{60}Co or an electron beam accelerator has the ability to inactivate pathogens with a very high degree of reliability and in a clean and efficient manner. The ionizing radiation interacts with matter both directly and indirectly. Direct interaction takes place with critical molecules like DNA and the proteins present in the microorganisms, thus causing cell death. During indirect interaction, radiolysis products of water result in the formation of highly reactive intermediates that then react with the target biomolecules, culminating in cell death:

$$H_2O \xrightarrow{\text{Ionizing radiation}} e^-_{aq}, {}^{\bullet}OH, {}^{\bullet}H, HO^{\bullet}_2, H_2, H_2O_2$$

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

The presence of oxygen is important in the process, as it is a known radiosensitizer that helps fix the radiation damage done to cells, thereby inhibiting their self-repair mechanism and resulting in the inactivation of microorganisms.

The radiation dose required to inactivate the pathogenic bacteria is generally defined in terms of the D_{10} value, which is the radiation dose required to reduce (through inactivation or cell death) the microbial concentration by a factor of ten or by one log cycle. In fact, this principle was the basis for producing radiation sterilized, single use medical products and is now well established in industry worldwide [4.14]. Based on the same principle, the pathogens present in sewage sludge can also be effectively removed by exposure to high energy radiation. The radiation treatment of sewage sludge offers an efficient, simple and reliable method for producing pathogen free sludge that can be further upgraded to produce a value added biofertilizer and allow waste recycling. Therefore, in recent years, irradiation of sewage sludge as a tertiary treatment process has been investigated [4.15–4.20].

It has been shown [4.21] that a dose of 2–3 kGy destroys more than 99.9% of the bacteria present in sewage sludge and leads to the almost complete removal of helminth eggs and to the inactivation of the agents that cause disease in animals (Fig. 16). Doses of this magnitude are employed for the radiation treatment of sewage sludges at an industrial plant in Geiselbullach, Germany [4.9], and slightly higher doses (4 kGy) are used at a pilot plant near Boston, United States of America [4.22]. Higher doses (up to 10 kGy) are required to inactivate more radiation resistant organisms. Doses of 10 kGy were used at a sewage treatment plant in Albuquerque, United States of America [4.23, 4.24], and at an installation in Ukraine [4.25].

In addition to disinfection, irradiation has a beneficial effect on physico-chemical properties of sewage sludges such as the specific resistance to filtration, water separation and sedimentation. An increase of the sedimentation rate is observed when sludges are irradiated [4.26]. The changes are due to a decrease in the stability of colloidal particles in the irradiated sludges accompanying radiation induced changes in the charge on the particles. The changes in the physical properties of sludges do not affect their quality as fertilizers or fodder additives [4.25].

4.2. PILOT AND INDUSTRIAL SCALE OPERATION

Pilot and industrial scale plants for radiation disinfection of sewage sludge are in operation in several countries around the world. In Geiselbullach,

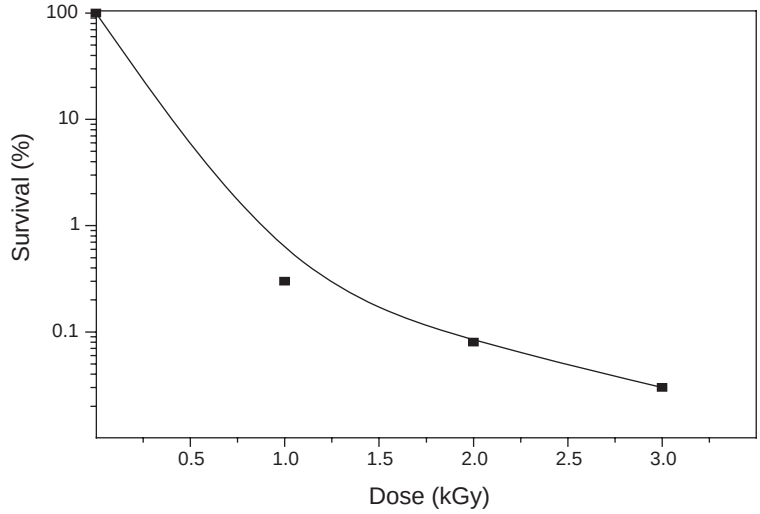

FIG. 16. Survival of coliform microbial population as a function of radiation dose [4.21].

a research and demonstration pilot plant with a ^{60}Co source (with ^{137}Cs added later) went into operation in 1973, producing fertilizer from sewage sludge [4.9, 4.26]. The favourable results obtained at this facility led to its conversion to an industrial operation, with the disinfected sludge being used in agriculture [4.29]. The plant consists of two underground components: the irradiation shaft with a built-in central tube, and the pump shaft containing recirculation and evacuation pumps, valves and pipework. The irradiator operates in a batch mode, treating approximately 5.6 m^3 of sludge at a dose of 3 kGy. This batch type operation continues automatically for 24 h/d; in 1978, it yielded a daily output of 145 m^3 of irradiated sludge. It was subsequently found that the dose could be reduced from 3 to 2 kGy if oxygen was bubbled through the sludge during the irradiation.

Another pilot plant, built primarily for research purposes, was installed in 1978 in Albuquerque and remained in operation until 1985 [4.23, 4.24]. The source of ionizing radiation was ^{137}Cs in the form of caesium chloride encapsulated in stainless steel. The dried sludge (8 t/d) for irradiation was moved slowly past the source on a conveyor belt along an S shaped path, receiving a dose of 10 kGy [4.27].

In Vadodara, India, a ^{60}Co based sludge treatment plant for processing 110 m^3 of liquid sludge per day has been in operation since 1992 [4.12, 4.21]. The advantages of such an irradiator system are that it can be easily incorporated into a conventional treatment plant, with operational flexibility, and that various dosages can be imparted to the sludge with the addition of sensitizing agents such as oxygen, air and ozone. Moreover, the loading, unloading and transport of the radioactive source can be carried out safely, quickly and easily (Fig. 17). The operating experience of this plant has demonstrated that radiation treatment of sewage sludge offers an efficient, simple and reliable method for producing pathogen free sludge that can be further upgraded to produce a value added biofertilizer and allow recycling of the waste products [4.28].

A pilot plant using an electron accelerator as a radiation source was built in Boston in 1976 [4.22, 4.29]. The electron beam energy used at the plant is 1.5 MeV. During the treatment process, the sludge rises in a gradually widening header and then flows in a vertical free fall as a thin sheet, 1.2 m wide and about 4 mm thick, that crosses the horizontal scanning electron beam. The output of the modified facility is up to 655 m^3 of disinfected sludge per day at a disinfection dose of 4 kGy. In the 1980s, similar facilities, also based on electron accelerators, were brought into service near Miami, United States of America [4.22, 4.29], and in Takasaki, Japan [4.30, 4.31].

At the electron beam sewage sludge treatment plant in operation in Takasaki, dewatered sludge is spread through a flat, 20 cm wide nozzle onto a

stainless steel conveyor belt and fed past the electron beam in a 1–10 mm thick layer at a rate that provides an absorbed dose of 5 kGy; the maximum feed rate is 300 kg/h. After irradiation, the sludge is mixed with a bulking agent such as perlite in order to make the product aerobic and is then moved to a conveyor belt where it is composted under conditions of controlled aeration and frequent mixing.

About 3 kGy of absorbed dose removes 99.99% of the pathogenic bacteria from sewage sludge consistently, reliably and simply. A schematic diagram of the main plant in Vadodara, India, is shown in Fig. 18; Fig. 19 shows the irradiation vessel and how the plant is incorporated into a municipal wastewater treatment plant. The process of sewage sludge treatment using radiation is very simple. The incoming sludge is transported to an underground reservoir. It is then fed into a 3 m^3 irradiation vessel and continuously circulated in a loop for a predetermined period of time.

After radiation exposure, the treated sludge is withdrawn from the irradiation vessel and pumped out to drying sand beds; there, the water

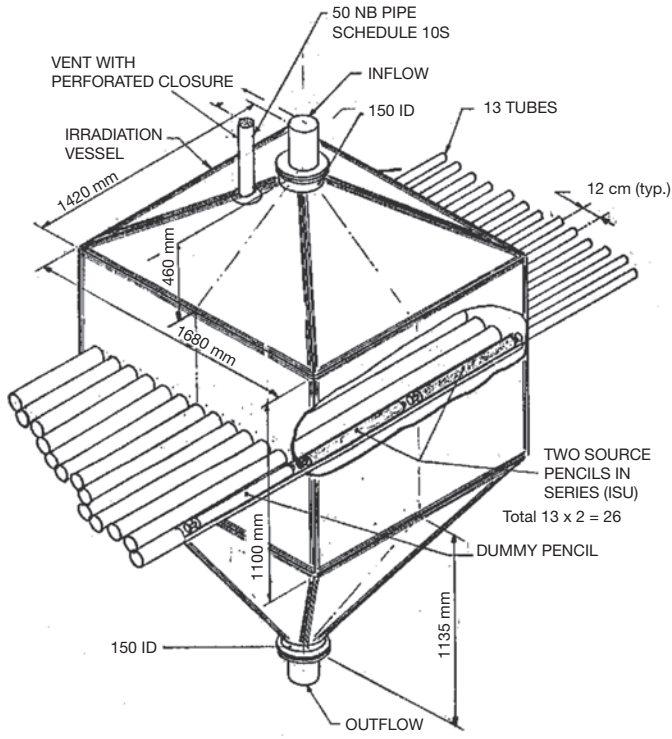

FIG. 17. Structure of the gamma ray irradiator at the plant in Vadodara, India.

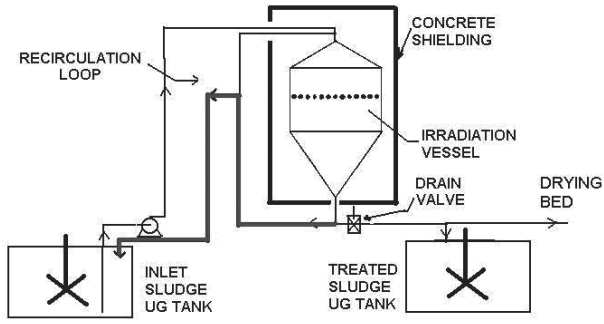

FIG. 18. Sludge treatment process at the Vadadora plant.

evaporates, yielding pathogen free dried sludge. The irradiated sludge, being pathogen free, can be beneficially used as manure in agricultural fields, as it is rich in required soil nutrients. Initial field trials of this use of the sludge for winter wheat and summer green gram crops have been held in villages around the city of Vadadora. The results have been very encouraging, and demand for the sludge has increased on the part of farmers (Figs 20, 21). Since the irradiated sludge is free of bacteria, it can also be used as a medium for growing bacteria that are useful for soil, such as rhizobium and azetobactor, to produce biofertilizer that can be used to enhance crop yields.

Applying the electron beam approach for sludge treatment has also been studied in Israel [4.32]. Digested sludges from municipal wastewater treatment plants have long been used directly in agriculture in Israel (Fig. 22). However, owing to infection by pathogenic microorganisms, the sludge must be processed to reduce the number of pathogens. An industrial scale plant with the capacity to treat 600 m^3 of dewatered sludge per day (18% solids content) with 10 kGy has been planned. This plant will be equipped with two electron accelerators (50 kW each) and handling facilities, and is expected to be more economical

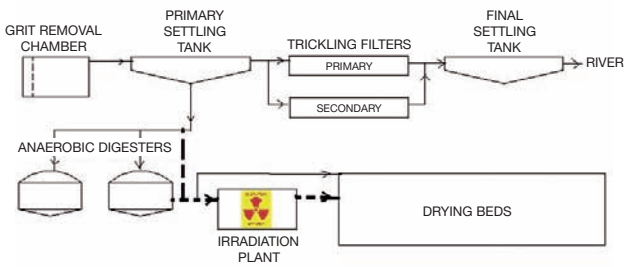

FIG. 19. Location of the irradiation plant in the conventional setup at the Vadodara plant.

FIG. 20. Field trials to study the effect of irradiated sludge on growth and yield of green gram: (a) control; (b) sludge based biofertilizer.

than plants using other sludge disposal processes such as incineration and lime stabilization.

Irradiation of sewage sludge as a tertiary treatment process has the potential to provide a viable solution to the environmental problems related to sludge disposal. Furthermore, it can play an important part in providing organic matter and micronutrients for agriculture, which can be helpful in improving soil characteristics and increasing crop yields.

4.3. ECONOMICS OF SLUDGE TREATMENT

For economic and practical reasons, reducing the dose required for disinfection of sludge is extremely important. Dose reduction can be achieved in

FIG. 21. Field trials of irradiated sludge at a grape and pomegranate plantation in India.

several ways, including thermoradiation of the sludge [4.24], bubbling oxygen through the sludge during irradiation [4.9] and combining radiation treatment with conventional disinfecting agents.

On the basis of the operating experience of the sludge irradiation facility at Vadodara, a sludge treatment plant based on a gamma ray facility has been conceived to serve a population of one million [4.21]. With 1.8 MCi of ^{60}Co, such a facility can treat up to 376 m^3 of sludge per day originating from a sewage treatment plant with a capacity of 48×10^6 L/d. The calculated capital costs, including isotopes, construction and other equipment, were approximately $2.5 million, and the annual operating costs are approximately $0.5 million. Referenced to the annual production (131 600 m^3/a), the unit cost works out to be $4.2/$m^3$ for wet thickened sludge and $105/t for dry sludge (at 4% solids content). Economic feasibility studies indicate that radiation treatment of sewage sludge is generally less costly than conventional treatment methods, particularly when the radiation facilities have a high output capacity. It seems likely, therefore, that industrial use of the radiation process will grow.

Electron accelerators are another promising source of ionizing radiation for the treatment of sewage sludge and offer advantages in the areas of radiation safety and high energy output. However, the limited penetration of electrons is a disadvantage and requires that the sludge be irradiated in a thin layer. A disinfection plant for municipal sewage based on the use of an electron accelerator has a capital cost of approximately $4 million for a capacity of 70 t of dry sludge per day [4.33]. An actual comparison of electron accelerator use with other processes shows the economic advantages of the electron accelerator with respect to both capital costs and operating costs [4.34].

FIG. 22. Land application of digested sludge in Israel.

4.4. ENHANCED COMPOSTING OF RADIATION DISINFECTED SEWAGE SLUDGE

Problems concerning the land application of sewage sludge include the need to improve handling through reduced water content and the need to remove odours and pathogens. Regulations put into place by the EPA require that sludge applied to land surfaces or incorporated into the soil be treated using a process that significantly reduces pathogens (e.g. anaerobic digestion, aerobic digestion, air drying, composting, lime stabilization). In addition, public access to the site must be controlled for at least 12 months, and grazing by animals used as foodstuff for humans must be prevented for at least one month [4.35]. To address these problems, treatment to stabilize the sludge — for example, composting — is recommended.

Composting of irradiated sludge may have two advantages:

(a) The composting period may be shortened by seeding controlled bacterial flora in the sludge;
(b) Contamination by pathogens or their growth can be prevented by inoculating the sludge with innocuous composting bacteria.

Studies on the composting of radiation disinfected sewage sludge have been carried out by a number of researchers [4.36–4.38]. For isothermal composting, the optimal temperature and pH are approximately 50ºC and pH7–8, respectively. The repeated use of the product as seed increased the rate of CO_2 evolution. The rate reached a maximum within 10 h and then decreased rapidly, and the conversion of organic carbon to CO_2 was approximately 40% [4.36]. By composting irradiated sludge, the process can be carried out under optimal conditions; moreover, the composting period is expected to be shorter, because it is not necessary to maintain the fermentation temperature at a high level long enough to reduce the number of pathogens in the sludge. The growth of inoculated microorganisms is greatly affected by the bacterial flora in the medium in which it is growing. Salmonella grew rapidly in irradiated compost, but it was possible to inhibit this growth by saturation of coliform bacteria after irradiation [4.37].

4.5. SOIL REMEDIATION

The EPA has determined that polychlorinated dibenzodioxins (PCDDs) and polychlorinated dibenzofurans (PCDFs) present a serious public health risk, and has established restrictions on the 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 ^{60}Co. Destruction of more than 98% of the contaminants was achieved with a dose of 800 kGy in soil containing 100 ppb of TCDD. The addition of contaminants such as dichlorobenzene and hexachlorobenzene did not affect the result. The addition of 25% water and 2.5% non-ionic surfactant was beneficial to the soil studied [4.39]. Selected aspects of the subjects discussed in this chapter are dealt with in Ref. [4.40].

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Chapter 5

RADIATION SOURCES FOR ENVIRONMENTAL APPLICATION

High energy radiation sources can be divided into two groups: those that employ natural or artificial radioactive isotopes, and those that employ particle accelerators. The first group consists of the classic radiation sources and artificial radioisotopes such as ^{60}Co and ^{137}Cs. The second group includes X ray generators and electron accelerators of various types.

Since the various types of radiation give different depth–dose profiles, the radiation used in an environmental application is determined largely by the nature and size of the objects to be irradiated and the penetration required. For example, stack gases can be irradiated uniformly and efficiently by electrons, while uniform irradiation of bulk wastewater or solid wastes generally requires more penetrating types of radiation.

5.1. RADIOISOTOPE SOURCES

The two artificial radioisotopes ^{60}Co and ^{137}Cs are widely used as gamma radiation sources. Cobalt-60 is produced by irradiating the stable isotope of cobalt (^{59}Co), while ^{137}Cs is separated from spent reactor fuel. Cobalt-60 gives high energy, more penetrating gamma radiation, while ^{137}Cs has the advantage of a longer half-life, necessitating less frequent source replacement (Table 6).

TABLE 6. RADIOISOTOPES COMMONLY USED AS SOURCES OF RADIATION

Isotope	Half-life (a)	Type and energy (MeV) of principal radiation emitted
Cobalt-60	5.272	β_{max} 0.313; β_{ave} 0.094 γ 1.332 γ 1.173
Caesium-137	30.17	β_{max} 1.18 (5%); β_{max} 0.51 (95%) β_{ave} 0.24 γ 0.662 (83%)

Beta rays are emitted with a spectrum of energies; Table 6 gives both the maximum and the average energies of the beta radiation. Gamma rays are emitted with discrete energies (in MeV) that are characteristic of the emitting radioisotope. Cobalt-60, for example, emits equal numbers of gamma photons with energies of 1.332 and 1.173 MeV.

5.1.1. Cobalt-60 sources

Cobalt-60 is the most widely used source of gamma radiation. It decays predominantly by emission of a 0.313 MeV beta particle to give an excited state of ^{60}Ni that loses energy by emitting two gamma photons in cascade with energies of 1.173 and 1.332 MeV. It is produced by exposing natural ^{59}Co to neutrons in a nuclear reactor, where the ^{59}Co(n,γ)^{60}Co reaction gives radioactive Co with activities as high as 50–100 Ci/g (1.9–3.7 TBq/g). Cobalt-59 is generally irradiated in the form of pellets, small slugs or thin disks of metal to provide a uniformly active material. The walls of the metal container serve to filter out beta radiation emitted by the ^{60}Co.

Gamma emitting sources must be surrounded by relatively thick shields of dense material to protect the operating personnel. The ^{60}Co, in the form of a hollow cylinder, encircles a cavity containing the sample to be irradiated, and the whole is surrounded by a compact mass of shielding material, generally lead. Cavity type sources are compact and can be designed to give uniform high intensity that cannot be varied.

Gamma sources designed for industrial use must handle large samples. They are generally in the form of a small shielded chamber that allows the exposure of a high activity ^{60}Co source near the centre of the chamber. Shielding is provided by massive concrete walls with a labyrinth entrance, and additional protection is afforded by the distance between the source and the operating personnel. When not in use, the source is stored below floor level in a shielded container or a water filled pit.

5.1.2. Caesium-137 sources

Caesium-137 is separated from the mixed fission fragments present in spent nuclear fuel elements. The radioisotope is available as caesium chloride, with activities of the order of 25 Ci/g CsCl (0.93 TBq/g). Caesium-137 sources are similar in design to ^{60}Co sources, although the amount of shielding can be reduced because of the lower energy of ^{137}Cs gamma radiation.

The decay of ^{137}Cs is more complex than that of ^{60}Co, with 5.4% of the decays giving the ground state of ^{137}Ba by emission of a 1.176 MeV beta particle and 94.6% of the events giving an energy rich metastable form of ^{137}Ba through

the loss of a 0.514 MeV beta particle. The metastable isotope, ^{137m}Ba, decays to the ground state of the nucleus with a half-life of 2.55 min by emission of a 0.617 MeV gamma photon or of an electron whose energy is 0.617 MeV less than the binding energy of the electron in the atom. With ^{137m}Ba, about 9% of the decay events give conversion electrons with energies of 0.625 MeV and about 2% give conversion electrons with energies of 0.656 MeV.

Caesium-137 has the advantage over ^{60}Co of a longer half-life (30.17 a), so that the frequency of source replacement is reduced. However, caesium chloride is water soluble, and if a ^{137}Cs source becomes damaged, it could contaminate water used as radiation shielding. Careful encapsulation of the caesium chloride in stainless steel reduces this risk to a low level.

The energy of gamma radiation from ^{137}Cs (0.66 MeV) is lower than that from ^{60}Co (mean energy 1.25 MeV). While this results in less expensive biological shielding with a ^{137}Cs source, it also leads to a less uniform dose distribution in the material being irradiated, a less powerful radiation source and greater radiation loss due to self-absorption in the source.

5.2. ELECTRON ACCELERATORS

Over 1300 electron accelerators, with a total power of about 50 MW, are currently in use worldwide for radiation processing and related research purposes. The widespread use of electron accelerators is due in large part to the relatively high power available with electron beams, the extremely low probability of inducing radioactivity in the irradiated products and the fact that, unlike gamma irradiation facilities, the beams can be turned on or off at will. Up to now, they have been used mainly for cable production and in thermo-shrinkable materials, foam sheets, coating and curing, and other applications. However, their use in the area of environmental protection is becoming increasingly important in industrialized countries, and wide ranging investigations have identified several areas of waste control to which radiation processing can contribute.

5.2.1. Electron accelerators for industrial uses

Electrons emitted by a cathode are accelerated in a vacuum chamber by an electromagnetic or electrostatic field passing through a metal foil exit window into the air, where product irradiation takes place. The energy range of electron beams used in environmental applications is from 0.5 to 10 MeV; lower energy electrons are unsuitable because of their lower penetration, while higher energy beams may induce radioactivity in some materials. Beam power

(the product of the electron energy and current) ranges from 5 to about 600 kW, although more powerful accelerators (1 MW and higher) are expected in the future.

Medium energy accelerators produce electrons with energies of 0.5–5 MeV at powers of up to 400–600 kW. They are used for wastewater treatment and for cleaning exhaust gases containing NO_x and SO_x. High energy accelerators, which generate electron beams with energies of 5–10 MeV, are used for radiation disinfection of sludge and, to a lesser extent, for the radiation treatment of hazardous wastes.

Electron accelerators for radiation processing are also classified on the basis of the mode of operation into direct current (DC) or radiofrequency powered machines. With DC accelerators, the electrons are accelerated using a DC voltage, either applied directly between the electron source and an electrode, as with electrostatic machines, or transferred to the electrons inductively, as with transformer type machines. The final energy of the electrons in DC accelerators is numerically equal to the potential difference across the accelerating tube. In radiofrequency accelerators, electrons are accelerated by passage across a pattern of electromagnetic fields that can be set up at very high frequencies.

Linacs, in which electrons are injected in pulses into a straight, segmented waveguide and accelerated by the electric field of an electromagnetic wave that travels down the tube, are also used in radiation processing [5.1].

5.2.2. Transformer accelerators

Direct current voltage is used to accelerate electrons in the direct acceleration method. Direct current voltage power supplies that are used as high voltage sources are usually based on the use of oil or gas filled transformers with a rectifier circuit. They are relatively simple and are the most reliable accelerator component. High voltage cable is frequently used to connect the power supply and accelerating head when the voltage level is less than or equal to 0.7 MV. A voltage level above 0.7 MV in a conventional transformer is impractical because of technical problems with the insulation and dimensions of such a device. Medium energy (0.5–5 MeV) can be obtained by a high voltage generator. A different type of inductance or capacitance coupling makes it possible to multiply alternating current (AC) primary voltage and obtain up to 5 MV of output voltage. The main parameters of selected transformer accelerators are shown in Table 7. Many different configurations have been built by major accelerator producers such as NHV Corporation, Japan; Energy Science Inc., United States of America; the Budker Institute of Nuclear Physics (BINP), Russian Federation; the D.V. Efremov Scientific

TABLE 7. PARAMETERS OF SELECTED TRANSFORMER ACCELERATORS

Parameter	Accelerator type			
	EPS-800-375	EPS-4	Dynamitron	ELV 12
Nominal energy	800 keV	1–5 MeV	1–5 MeV	0.6–1.0 MeV
Energy stability (%)	—	±2	±2	±1
Nominal beam current (mA)	375	30	50	500
Beam current stability (%)	—	±2	±2	±2
Beam power (kW)	300	150	250	400
Scan width (cm)	225	140	200	200
Dose uniformity (%)	±5	<±5	<±5	<±5
Mode of operation	Continuous	Continuous	Continuous	Continuous
No. of accelerating heads	2 sets of 2	1	1	3[a]
Total beam power (kW)	1200	220	250	400
Power consumption (kW)	1364	n.a.[b]	350	500
Electrical efficiency (%)	88	68	71	80
Producer	NHV, Japan	NHV, Japan	RDI, USA	BINP, Russian Federation

[a] Maximum beam current per head is 200 mA.
[b] n.a. = not available.

Research Institute of Electrophysical Apparatus (NIIEFA), Russian Federation; Radiation Dynamics, Inc., United States of America; and others [5.2].

In the EPS-4 electron accelerator, built by NHV, Japan, a Cockroft–Walton high voltage cascade multistage rectifier circuit is used in addition to a relatively low voltage transformer. A multistage rectifier circuit and 3 kHz AC voltage are applied. Accelerators of this type are used in the field of radiation sterilization. The high voltage coreless transformer concept was applied in the ELV 12 accelerator manufactured by BINP, Russian Federation. A certain number of secondary coils are needed to obtain the required output voltage. There is no central magnetic guide that simplifies the high voltage source design. The central pressure tank is used to install the high voltage transformer, accelerating section and scanner. Two more tanks are used with an additional accelerating tube and scanning devices. An SF_6 gas insulating system is used. Coreless accelerators are usually operated on AC voltage with a frequency of 0.4–1 kHz to reduce the accelerator dimensions. Electron energies of 0.2–2.5 MeV can be obtained in such accelerators.

5.2.3. Ultra-high frequency accelerators

Resonant ultra-high frequency (UHF) accelerators are based on a single large resonant cavity working at a frequency higher than 100 MHz. High power vacuum tubes are applied to provide the electromagnetic energy used to accelerate electrons in accelerators of this type. A UHF accelerator requires relatively simple and compact DC or pulse modulators to generate UHF oscillations. Medium and high electron energy levels with appropriate beam power can be obtained in such an accelerator (Table 8). ILU 10 accelerators (BINP, Russian Federation) are constructed based on a single coaxial resonator operating in a pulse regime. The resonator is made of two separate halves mounted inside a stainless steel vacuum envelope. The central cylindrical part of the resonator forms the accelerating gap. The electron injector consists of a grid in the upper electrode to control the beam current by changing the value of positive bias voltage on the cathode with respect to the grid. The self-excited generator, consisting of two industrial vacuum triodes, is used to form UHF oscillation inside a coaxial cavity and to provide the energy necessary for the electron acceleration process [5.2].

The Rhodotron accelerator operates on the basic principle that electrons gain energy when they cross the region of an existing electric field. The unique feature of the Rhodotron accelerator is its single cavity construction, where the cavity is crossed by electrons several times to gain energy. (The accelerating cavity of the Rhodotron 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 cross the cavity, they gain 1 MeV. Ten passes and nine magnets are therefore required to obtain 10 MeV of electron beam energy. A new Rhodotron electron accelerator arrangement was developed for the Rhodotron TT 1000, in which, using a multipass system across a resonant cavity with electron energies of 5 and 7.5 MeV, beam power of up to 700 kW is obtained.

5.2.4. Linear accelerators

The main feature of linacs is the use of microwave energy in the electron accelerating process. Microwave generators as power supplies are usually built for S or L band frequencies (1300–3000 MHz). A large number of small resonant cavities are used. Microwave energy source parameters play a crucial role in the performance of linacs. The klystrons are more stable in frequency and power, but they have an efficiency of 40–50%, compared with the 70% efficiency of magnetrons. Linacs can be built with a travelling or standing wave configuration. The latter technology affords a higher accelerating gradient at

TABLE 8. PARAMETERS OF SELECTED UHF AND RHODOTRON ELECTRON ACCELERATORS

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

the cost of a more sophisticated microwave power system and acceleration section technology. Accelerators of this type are not suitable for environmental applications owing to the low electrical efficiency (10–20%) and limited beam power (50 kW). Continuous wave operation may significantly improve electrical efficiency (up to 40%) and allow megawatt level beam power in the future.

5.2.5. Accelerators required for environmental applications

The most important factor determining the economic feasibility of the use of electron beam technology in environmental applications is the cost of the electron accelerator. Accelerator manufacturers produce many kinds of electron accelerator, with energies ranging from 0.5 to 10 MeV and beam powers ranging from 50 to 400 kW. For flue gas treatment, electron energies of approximately 0.7–1.0 MeV are adequate, but energies above 1.0 MeV are useful for wastewater treatment. Such energy levels provide the necessary penetration of accelerated electrons into wastewater when applied to suitable hydrodynamic wastewater flow regimes. Some accelerators with energies above 5 MeV are manufactured with low beam power (less than 50 kW). Low beam power is adequate for use in experimental and pilot plants, but not for

large scale treatment in industrial applications. Thus, medium energy accelerators are the most practical for flue gas and wastewater treatment. For sludge treatment, however, higher energies are required, and the beam power of accelerators used for this purpose reaches 400 kW. Moreover, there are several applications that call for the manufacture of accelerators with a beam power of up to 1 MW. The basic criteria for accelerators for environmental application are:

- High beam power to increase productivity and reduce unit operating costs;
- High electrical efficiency to reduce demand and unit operating costs;
- High beam utilization to increase productivity and reduce unit operating costs.

The basic parameters of selected accelerator designs are given in Table 9. The cost of an accelerator is governed by its beam power. The accelerator with the highest power has the lowest unit cost for power generation and is the most economical in environmental applications.

TABLE 9. BASIC PARAMETERS OF SELECTED ACCELERATORS FOR RADIATION PROCESSING

Manufacturer (accelerator type)	Energy (MeV)	Current (mA)	Power (kW)	Price[a] (10^9 $)	Unit cost ($/W)
IBA, Belgium (UHF)	10	15	150	6.1	40.7
RDI, USA (DC)	5	50	250	4.9	19.6
NHV, Japan (DC)	5	30	150	5.0	33.3
Vivirad, France (DC)[b]	5	200	1000	4.4	4.4
BINP, Russian Federation (UHF)	5	10	50	1.2	24.0
BINP, Russian Federation (DC)	1	400	400	2.0	5.0

[a] As of 2004.
[b] Under development.

TABLE 10. TYPICAL PRICE AND UNIT COST OF ACCELERATORS, BY POWER

Power (kW)	Price (10^9 $)	Unit cost (10^4 $/kW)
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

Table 10 and Fig. 23 present the typical price and unit cost of accelerators according to their power.

5.2.6. Accelerators for flue gas treatment

Radiation processes for the removal of SO_2 and NO_x from flue gas formed by fuel combustion for thermal power production have been successfully demonstrated in many laboratories and pilot plant facilities. Full scale industrial plants are already in operation in China and Poland, and other industrial facilities for flue gas treatment are under construction or consideration. It has been clearly established that industrial implementation of the

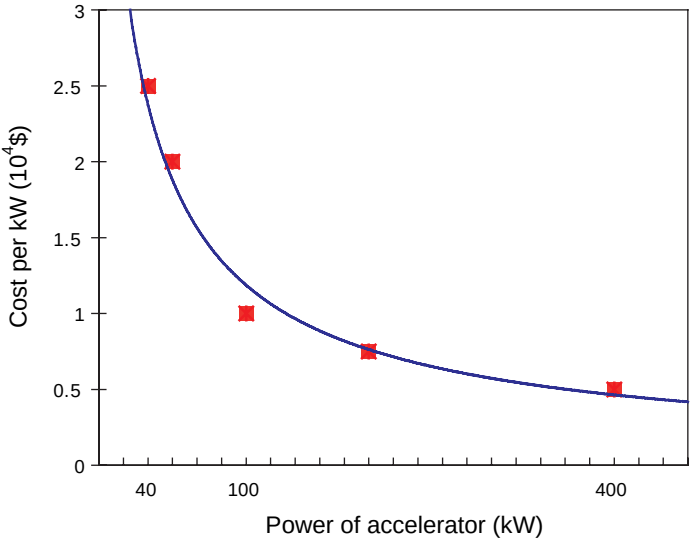

FIG. 23. Unit cost versus power of a typical accelerator.

TABLE 11. BASIC PARAMETERS OF SELECTED PILOT AND INDUSTRIAL FACILITIES FOR FLUE GAS TREATMENT

Parameter	Shin Nagoya pilot plant, Japan	Kawęczyn pilot plant, Poland	Pomorzany electric power station, Poland
Flue gas stream (Nm^3/h)	12 000	20 000	270 000
Removal efficiency (%):			
SO_x	94	96	80
NO_x	80	72	70
Accelerator:			
Energy (keV)	800	700	700
Beam power	3 × 45 mA	2 × 72 mA	4 × 375 mA
Vessel size	2.4 m × 1.9 m × 14 m	∅ 1.6 m × 10 m	2 × ∅ 2.6 m × 14 m
Vessel cross-section (m^2)	4.9	2	2 × 5.3
Beam utilization (%)	66	64	68
Gas velocity (max.) (m/s)	1	2.8	7
Dose (kGy)	10.5	11.5	8
Thickness of first foil (μm Ti)	38	50	50
Thickness of second foil (μm Ti)	30	50	50

electron beam process for flue gas treatment requires accelerators with a beam power of at least 300 kW and electron energy in the range of 0.8–1 MeV. Table 11 shows the basic parameters of selected pilot and industrial facilities. According to the present state of the art of accelerator technology, those accelerators meeting such requirements are based on high power, high voltage transformers.

The economical design of the Pomorzany electron beam flue gas treatment plant was based on a retrofitted electron beam installation for a boiler capacity of approximately 130 MW. The unit investment costs of $160/kW are calculated on the basis of two parallel reaction chambers, requiring four accelerators [5.3]. The unit operating costs are related to the electric generating capacity of the electric power station. Thus, the operating costs for electron beam flue gas treatment are $806/t pollutant removed, and the annual operating costs are $7346/MW installed power.

The investment costs of retrofitting wet FGD installations are usually $120–200/kW, depending on the size of the plant and local conditions; the investment costs of retrofitting SCR system installations are approximately $110/kW, depending on the plant capacity and the difficulty and scope of the retrofit [5.4]. Thus, the investment costs of a combination of wet FGD and SCR

systems are, for a small boiler, approximately $230/kW, which is considerably higher than the investment costs of electron beam technology ($160/kW). The annual operating costs of the wet FGD methods are approximately $2500–3000/MW, while those of SCR methods are $3800–4600/MW [5.4]. Thus the removal of both pollutants using conventional methods costs $6300–7600/MW annually, which is higher than the cost of the electron beam process.

5.2.7. Accelerators for wastewater treatment

Radiation processing has been found to be effective in water and wastewater treatment (decomposition of toxic substances) and wastewater disinfection. Water and wastewater treatment activities have been studied in laboratory and pilot plant facilities in Austria, Brazil, Japan, the Republic of Korea, the Russian Federation, the United States of America and several other countries [5.5–5.8]. Facility throughput can be increased and unit costs of wastewater treatment can be decreased by improving electron beam utilization. Higher process efficiency and lower unit costs can be achieved by using lower dose levels. The minimum dose depends on the origin, specific properties and nature of the contaminant in the wastewater to be treated. It can vary from 0.2 up to 2 kGy. Electron energies of more than 1.0 MeV are useful for industrial scale plants. Such energies provide adequate penetration of accelerated electrons into the wastewater in admissible hydrodynamic regimes of wastewater flow. Therefore, accelerators with an energy range of 1.0–2.0 MeV (middle energy) provide maximum practical use for wastewater treatment.

On the basis of economic evaluation of data obtained by several investigators, it has been found that for the reclamation of effluents from a municipal plant a suitable dose is approximately 0.2 kGy for a flow rate of 100 000 m^3 of effluent per day. The cost assessment of a radiation processing plant using electron beam technology is based on a dose 0.2 kGy and an electron accelerator capacity of 400 kW. The cost of such a high power accelerator is approximately $2 million; the cost of the building, piping and other equipment, and construction is estimated to be $1.5 million. With the additional costs of taxes, insurance and documentation of $0.5 million, the overall capital and operating costs are approximately $4.0 million and $1.0 million, respectively, as shown in Tables 12 and 13.

This estimate does not include the costs of the land and research and development, or the cost of licensing from the regulatory authorities. The expected construction period includes 11 months for civil and installation work and 3 months for trial operation. To estimate the operating costs, the annual electricity use of the accelerator and other equipment is calculated to be

TABLE 12. CAPITAL COSTS OF AN INDUSTRIAL RADIATION PROCESSING PLANT

	Cost (10^6 $)	Remark
Accelerator (1 MeV, 400 kW, double window)	2.0–2.5	Costs of land, R&D and authorization are not included
Water reactor and other raw material	1.0–1.5	
Installation costs (welding, piping inspection, etc.)		
Design		
Shield room and construction works		
Other (transport, taxes, insurance, etc.)	0.5	
Total	4.0–4.5	~$4 million

TABLE 13. OPERATING COSTS OF AN INDUSTRIAL RADIATION PROCESSING PLANT

	Cost increase due to incorporation of electron beam ($)	Remark
Investment	4 000 000	Not included in operating costs
Interest	240 000	6%
Depreciation	200 000	20 a
Electricity	320 000	800 kW
Labour	100 000	3 shifts
Maintenance, etc.	80 000	2%
Total	940 000	~$1 million/a

500 kW (80% efficiency) and 300 kW, respectively, for a total of 800 kW. Based on year-round operation (8000 h/a), the electricity costs are $320 000/a when the cost of electricity is assumed to be $0.05/kW·h. The labour costs of operation are calculated on the basis of three shifts per day and are approximately $100 000/a. Therefore, the actual operating costs of a plant treating 100 000 m^3 of waste per day are approximately $1.0 million/a, including interest and depreciation of investment, and approximately $0.12 for investment and $0.03 for operation for each cubic metre per day of wastewater capacity [5.9].

5.2.8. Accelerators for sewage sludge treatment

The sludge generated by sewage plants is a rich source of agricultural nutrients. However, it also contains a high level of pathogens, which limits its reuse. Thus, there is a need to extend the treatment process to include the removal of the pathogenic bacteria with a high degree of reliability.

High energy radiation processing has the ability to inactivate the pathogens with a very high degree of reliability and in a clean and efficient manner. In India, the Sludge Hygienization Research Irradiator (SHRI), an industrial scale demonstration plant with a ^{60}Co source, is in operation, treating up to 110 m^3 of sewage sludge per day [5.10].

Liquid sludge irradiators can be designed to operate in a batch or continuous flow mode. Irradiation of dewatered or dry sludge is carried out in facilities equipped with a conveyor belt system similar to those used for medical product sterilization. With their present capabilities, electron accelerators are very well suited to continuous treatment of waste, in a dewatered, dried or prepackaged form of the appropriate thickness, beneath the beam of accelerated electrons applied at a steady rate.

In the sewage treatment station for Otwock, Poland, which was designed on the basis of research data [5.11, 5.12], dewatered sewage sludge containing 30% dry matter is spread on the transporter and disinfected by electron irradiation at a dose of 5 kGy. The capacity of the installation is 70 t/d, which corresponds to a wastewater stream of 48 000 m^3/d. The capital costs of the radiation processing unit were estimated to be $4.0 million, primarily related to the cost of the accelerator and the building.

Table 14 lists economic aspects of the irradiation process performed using different accelerators. The table shows that the higher investment costs related to the price of accelerators are compensated for by greater beam power. Low cost, effective, high power and high energy accelerators will become an adequate technical and economic solution for the electron beam treatment of sewage sludge in the near future.

The key to the successful implementation of electron beam treatment in environmental protection is cost–benefit analysis, management and optimization. To compete with other, conventional processes from the economic point of view, electron beam processing has to be designed to operate with a cost effective accelerator that provides sufficient amounts of low doses of radiation.

The most important consideration is the relatively high capital cost of an electron beam system. This means that its capital amortization is a major item in the cost–benefit assessment. To be appropriate, the accelerator has to meet all technical and economic conditions for successful application. The unit costs decrease as the throughput increases. Optimization of electron beam utilization

TABLE 14. ECONOMIC ASPECTS OF ELECTRON BEAM TREATMENT OF SEWAGE SLUDGE
(60% facility utilization, operating 7300 h/a)

Irradiated waste material	30% dry mass	35% dry mass	35% dry mass
Maximum productivity per accelerator (t/h)	27	9	90
Capacity per accelerator (t)			
Per hour	56	12.5	—
Per day	1 120	250	—
Per year	408 800	91 250	912 500
Accelerator type	Rhodotron TT 300	ILU 10	Under construction
Manufacturer	IBA, Belgium	INP, Russian Federation	Vivirad, France
Electron energy (MeV)	10	5	5
Beam power	3 × 150 kW	2 × 50 kW	1 × 1 000 kW
Dose (kGy)	12	12	12
Investment costs ($)	25 500 000	4 364 000	8 800 000
Operating costs ($)	3 662 800	919 500	2 000 000
Unit cost ($)	9.0	10.1	2.2

by properly arranging the irradiation unit increases productivity and reduces unit operating costs. Significant unit cost reductions can be achieved by implementing low dose levels.

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This publication provides a summary of information relevant to radiation processing for environmental applications. It reports on current uses of radiation based technologies for flue gas treatment, wastewater purification and treatment of sewage sludge. This report is expected to serve as a basis for the preparation of guidelines for and feasibility studies of further implementation of radiation processing technologies aimed at mitigating environmental degradation, and to play an important role in promoting these technologies worldwide.

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