Electron Beam Processing – What are the Limits

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Abstract. The radiation emitted from electron accelerators or gamma sources is widely applied in the industry. Up to 200 big gamma irradiators and up to 2000 electron accelerators are used for radiation processing. The radiation sources are installed in service centers or are incorporated as a unit in the technological line. The biggest application of the process concerns sterilization and polymer cross-linking. Other still being a challenge is application of the technology in food irradiation. The biggest applications in line regarding the power of machines used type concern environmental protection. This is a new approach regarding electron beam application the machine installed with process vessel is a new radiation catalytic reactor installed in the process line. All the rules applicable to the chemical and process engineering process integration scheme are valid. Due to the fact that mass flow of the stream (flue gases or wastewater) are huge the high power accelerators are applied in such cases. The important factor concerns reliability of accelerators for in line applications, the 8500 hours continuous operation in the year is required. The radiation has to compete with other technologies applied widely in the environmental protection sector like FGD +SCR (flue gases) or biological treatment (wastewater).

1. Introduction

The radiation technologies found different applications in industry [1,2,3]. One type of radiation sources are gamma irradiators [4] other accelerators[5]. According to the Machi's report [6] up to 2000 electron accelerators and up to 200 big gamma irradiators [7] and are used for radiation processing. The radiation sources are installed in service centers [8] or are incorporated as a unit in the technological line [9]. The biggest application of the process concerns sterilization [10] and polymer cross-linking [11]. The advantage of electron accelerators over gamma sources is due to the high dose rate what shortens the processing time [12]. An electron accelerator of the power 15 kW regarding delivered dose is equivalent to 1 MCi of the Co-60. Electron accelerators of the power up to 700 kW and the energy up to 10 MeV are available on the market [13, 14, 15]. The present situation concerning difficulties in the cobalt delivery due to transport restrictions will promote accelerator applications as a radiation source. However, the penetration range of the gamma rays is much larger than those of electron beam, of accepted processing energy up to elements activation limits [16]. The application of e/X conversion is a technical feasibility [17], however due to the low conversion efficiency this is some limit of this technique application [18]. The most complete review of accelerator technology is presented by Berejka and Cleland [19]. The environmental applications of electron accelerators is a new challenge [20], due to the fact that high power accelerators with high reliability are required in the continuously operated gas or liquid flow installations [21]. The design and operational issues are quite different to observed in commonly used radiation processing plants, therefore the progress in the field may open a new unexplored fields of electron accelerators application.

2.Environmental Issues and Major Pollution Control Requirements

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 the degradation of the environment. For example, fossil fuels, including coal, natural gas, petroleum, shale oil and bitumen, are the primary source of heat and electrical energy generation, and are responsible for emitting a large number and amount of pollutants into the atmosphere via exhaust gases from industry, power stations, residential heating systems and vehicles. During the combustion process, different pollutants such as fly ash, SO_X (including SO_2 and SO_3), NO_X (including NO_2 and NO) and VOCs are emitted.

Water pollution is other major problem in the global context. It has been suggested that it is the leading worldwide cause of deaths and diseases. In addition to the acute problems of water pollution in developing countries, industrialized countries continue to struggle with pollution problems as well. Water is typically referred to as polluted when it is impaired by anthropogenic contaminants and either does not support a human use, like serving as drinking water and/or undergoes a marked shift in its ability to support its constituent biotic communities, such as fish. BOD, COD and SS heavy metals, PCB

2.1. Air Pollution Control

Many countries have introduced a regulations similar to those known as the Clean Air Act in the US which describes one of a number of pieces of legislation relating to the reduction of air pollution [22]. Different measures are undertaken to curb air pollution, control of emission at the source is one off them. The limits regarding maximum allowable concentration in flue gases have been set, they may vary from the country to the country.

2.2.Water Pollution Control

Similar legislation governing water pollution was introduced in most of the countries and is known as the Clean Water Act in the US. The objective of these actions is to restore and maintain the chemical, physical, and biological integrity of the waters by preventing point and dispersed pollution sources. Governments control water pollution by setting stringent regulations for wastewater discharge, extension and improvement of sewerage systems, improvement of river water flow, and by appropriate environmental impact assessment of various projects which discharge wastewater into public water areas. Effluent water quality is regulated based on its daily maximum value. Standard values for BOD, COD and SS are given as the maximum and average values respectively. Other limits concern heavy metals and PCBs. The limits are established by national[23] and international authorities [24].

3.Pollution control process design and integration

Different processes have been applied for air and water pollution control. Regarding the pollutants emitted in combustion processes Flue Gas Desulfurization (FGD) based on the sulfur dioxide absorption in lime or limestone water suspension and Selective Catalytic Reduction (SCR) for nitrogen oxide reduction by gaseous ammonia over a catalyst are mostly used. Polyaromatic Hydrocarbones (PAHs) including dioxins and mercury are mostly adsorbed on active carbon dust or bed.

Wastewater purification is mostly based on mechanical or sedimentation processes for suspended solid removal and biological processes for organic contaminants reduction. In some cases chemical treatment, ion exchange or membrane processes are being applied.

In all cases, including radiation technologies, which will be discussed later the process design is based on unit operations integration. The design starts at a conceptual level and ultimately ends in the form of construction blue prints. Process design is distinct from equipment design, which is closer to the design of unit operations. Processes often include many unit operations. Designers usually do not start from zero, especially for complex projects. Often the engineers have pilot plant data available or data from full-scale operation data. Other sources of information include design criteria provided by published scientific data, laboratory experiments, and general chemical engineering methods of heat and mass transfer calculations input. The following documents are being executed in design phase;

- Block Flow Diagrams (BFD) – it is a very simple diagram composed of rectangles and lines indicating major material or energy flows.
- Process Flow Diagram (PFD) - shows relationships between major components in a system. A PFD also tabulate process design values for the components in different operating modes, typical minimum, normal and maximum values. A PFD does not show minor components, the piping systems, piping ratings and designations.
- Piping and Instrumentation Diagram (P&ID) - shows all of piping including the physical sequence of branches, reducers, valves, equipment, instrumentation and control interlocks. The P&ID are used to operate the process system.
- Basic Engineering(BE) – gives technical specifications of the system components (equipment), balances, estimated investment and operational costs
- Technical and Civil Works Design – covers all technical drawings of equipment integration, positioning, foundations and buildings

Designs have objectives and constraints which requires a trade-off among such factors. Constraints include: capital cost, available space, safety concerns, environmental impact and projected effluents and emissions, solid and liquid waste production, operating and maintenance costs .Other factors that designers may include are: **reliability** i.e. the ability of a system or component to perform its required functions under stated conditions for a specified period of time; **redundancy** i.e. duplication of critical components of a system with the intention of increasing reliability of the system, usually in the case of a backup or fail-safe; **flexibility** i.e. ability to adapt to external changes of stream flow, pollutants concentration in effluent and other treated medium physicochemical parameters.

Nowadays a new methods regarding design are being applied so called process integration. It is a holistic approach to system design which considers the interactions between different unit operations from the outset, rather than optimising them separately. This can also be called integrated process design or process synthesis. Process integration exploits the interactions between different units in order to employ resources effectively and minimize costs The methodology is often employed, in conjunction with simulation and mathematical optimization tools and the main advantage of process integration is to consider a system as a whole (i.e. integrated or holistic approach) in order to improve their design and/or operation. In contrast, an analytical approach would attempt to improve or optimize process units separately without necessarily taking advantage of potential interactions among them.

The above mentioned procedures and requirements are applied for the processes in which an accelerator installed on the process vessel is just one of the component of the flow system and this is a biggest differences between service centers and radiation technologies applied for pollution control.

3.Radiation technologies for pollutants control

The possibility of radiation technologies for the wastewater treatment has been reported many years ago [25,26]. However these were just preliminary works and many laboratory and pilot scale tests were performed till in the beginning of XXI century these technologies found the first industrial applications [27,28].

3.1.Electron beam flue gas treatment

The development of the process has followed the stages which were discussed earlier, the laboratory installations were tested in JAERI, Takasaki, Japan; EBARA, Japan; KFK, Karlsruhe, Germany; INCT, Poland; MINT, Malaysia and other centres all over the world. Then pilot installation were constructed in Indianapolis, USA; Badenwerk, Karlsruhe, Germany; Kaweczyn , Poland; Nagoya, Japan; Mianyan and Beijing China and other centers. Finally industrial plants were constructed in Chengdu, China and EPS Pomorzany, Szczecin, Poland. The history of the developments is discussed by Chmielewski in [29].

The process itself consists of spray cooler (different solutions were tested; dry bottom scrubber and scrubber with water recirculation). The dry bottom scrubber seams to be most feasible solution, however in the case of high fly ash concentration the recirculation of the water may reduce particulate load to assure purity of the by-product. Ammonia injection installation is a standard unit used in may technologies. The other important unit operation important is filtration of fine aerosol which is sticky and hygroscopic. Bag filters, gravel bed wet filters and ESP were tested at pilot plant in EPS Kaweczyn [30]. The most important is construction of the process vessel with the accelerators. The different designs were developed as illustrated in Figure 1[31] – however finally the solution tested on the pilot plant at EPS Kawęczyn found its way to Pomorzany industrial plant applications [32].

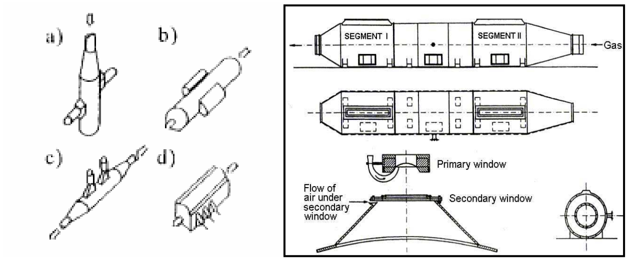

Fig.1.Process vessels applied at pilot plant; a - Indianapolis, b - Badenwerk, c –Kaweczyn, d – Nagoya (right).Process vessel applied at Kaweczyn – longitudinal gas irradiation, double window system and air curtain (left).

Two accelerators 50 kW, 700keV each were applied in the case of Kaweczyn pilot plant, the size and number of accelerators increased up to four units 268 kW, 700keV each (two power supplies) in the case of Pomorzany industrial plant. This is a challenge and some problems aroused regarding power supply insulation (in 800keV operation mode) and accelerator window construction.

The high power accelerator requirements are listed by Zimek [33]:

- Low electron energy to reduce investment and operational costs
- High beam power to achieve high throughput and lower unit operational cost
- High electrical efficiency to reduce exploitation costs
- High energy beam-gas transfer efficiency to reduce exploration costs.

These requirements can be met in the case of flue gases treatment where penetration of electrons due to the fact that density of the fluid is ca.1,2 kg/m^3 is rather high. However the losses at the double window (two titanium foils with 70 mm air gap) rise from 36 to 48% when electron beam energy decrease from 700 to 500keV. The accelerators of the size 1000 kW, 1MeV seams to most optimal for these applications. The high reliability and long intervals between maintenance are required since the plant must continuously operate 8500 hours annually.

3.2.Electron beam wastewater treatment

Electron beam wastewater treatment investigations have started many years back and were pioneered by Pikaev [34], Cooper [35] and others. In recent years a new achievements were

reported by Han regarding pilot [36] and full scale operation [37]. Possible applications of the process are discussed by Sampa and IAEA CRP participants [38].
The system for wastewater treatment consist in all cases from pre-treatment unit (screens, clarifiers etc) and in conventional case from biological treatment. However in many cases organic pollutants could be hardly biodegradable. In such a case combination of electron beam process and biological treatment seams feasible. Additional operational costs reduction can be achieved by ozone/eb application.
The main engineering problem concerns high density of the treated medium, liquid density is almost 1000 higher than gas density and electron penetration range is low. Therefore special process vessels are being applied (Fig. 2).

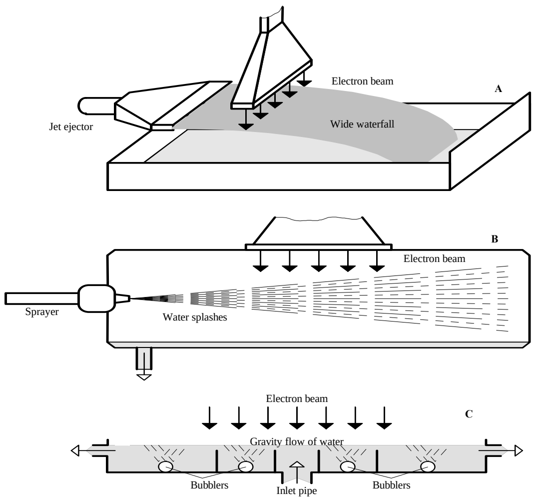

Fig.2. Solutions applied for irradiation of wastewater.

The irradiation in water waterfall was proposed by group of Cooper [35] and farther developed by Han [36], Pikaev's idea concern irradiation in spray [39], while Sampa et al. [40] invented liquid up flow system most classical from process engineering point of view.
The most promising solution not applied in commercial scale is sludge irradiation for microbiological control and agricultural applications [41].

4.Electron accelerators for pollution control technologies.

There is quite high potential regarding possible accelerator application in pollution control technologies. Beside the applications discussed above, the new possibilities concern organic pollutants destruction in gaseous phase [42], oil fired boiler flue gases treatment [43] and others.
Selection of the accelerators depends on the parameters of the process, in the case of environmental protection , the main parameters are:

- Density of the medium treated [kg/m^3]
- Mass flow of the stream [kg/s]
- Required dose [J/kg].

In the case of flue gas treatment density of the medium is close to 1.2 kg/m^3 , in the case of wastewater treatment density of liquid is close to 1000 kg/m^3 , and in the case of biological sludge with 30% solid phase content density is close to density of water.
The mass flow in the case of electron beam flue gas treatment varies from 15x10^3 kg/h (medical waste incinerator) up to 500x10^3 kg/h and more in the case of coal/oil fired boiler.
Regarding wastewater the flows are in range from 100x10^3 kg/h up to 1000x10^3 kg/h.

Output of sludge electron beam hygenization unit treating biological sediment from wastewater treatment plant operating for $100x10^3$ habitants town is about 70 tons of dewatered solid (30% dry mass) a day.
Dose required for flue gases treatment is close to 9 kGy, sludge hygenization and wastewater treatment require dose close to 3kGy.
Therefore most feasible from technical and economical point of view for flue gas treatment are accelerators of 800 keV – 1 MeV electrons energy and 500 – 1000 kW beam power. For special applications (medical and municipal waste incinerators) smaller units are required.
For dewatered sludge treatment accelerators of electron energy 10 MeV seems to be most feasible, since the required beam power is not too high.
Wastewater treatment require optimization of the whole system; electron accelerator – process vessel.

5.Conclusions

The flue gas electron beam treatment technology has been developed to the full industrial scale. The advantages over conventional processes has been illustrated. The development of high power, low cost , reliable accelerators is necessary for the broader implementation of the process (coal and oil fired boilers, municipal and medical wastes incinerators).
Wastewater electron beam technology is economically feasible in combined processes with biological stage, the high power accelerator installed in the first industrial plant has demonstrated good operational parameters.
Application of accelerators for sludge hygenization is very feasible and economical process , especially for the countries which required organic matter for sandy soil improvement, and the accelerators present at the market fits well technology needs.

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