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#### **Electron Accelerators for Environmental Protection**

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This article gives an overview of existing and possible electron accelerator applications for environmental pollution control. Laboratory and pilot plant tests and industrial applications have illustrated the possibility of applying this technology for purification and treatment of gaseous, liquid, and solid wastes. Examples of ionizing radiation application to protect the environment and human health are discussed.

Keywords: Electron accelerator; flue gas treatment; wastewater treatments; biological sludge disinfection.

# 1. Introduction

The powerful tools of ionizing radiation, electron accelerators, have been used for radiation processing of materials for more than half a century [1-4]. However, the possibility of radiation applications for environmental pollution control was realized in the 1970s, when environmental protection agencies were established and standards for pollutant emission limits were set. The pioneer in these applications was the Japan Atomic Energy Research Institute, Takasaki [5]. The special input for application of the technology was the development of new high power electron accelerators which can be used for on-line processing of huge flow streams of liquid or gaseous pollutants. The accelerators were employed for off-gas and wastewater treatment [6-8] and biological sludge from wastewater treatment plant disinfection testing [9], and have higher throughput in comparison with gamma sources applied to the last-mentioned technology as well [10]. Technologies which apply particle accelerators are considered important for further high-tech processes in different fields of national economies: material processing, sterilization of medical products, environmental protection, medicine (patient treatment and diagnosis; manufacturing of radiopharmaceuticals), cargo inspection, chemical analysis, nuclear power (ADS and transmutation), and so on [11]. Electron

beam processing of materials was introduced during the 1950s, and this technology has been continually evolving since then. A variety of industrial electron accelerators can now provide electron energies from 0.3 MeV to more than 10 MeV, with average beam power capabilities of up to 300 kW [12]. Nowadays higher power accelerators are available as well.

## 2. Industrial Off-Gas Purification

Pollutants are emitted to the atmosphere along with off-gases from industry, power stations, residential heating systems, and municipal waste incinerators. Fossil fuels, which include coal, natural gas, petroleum, shale oil, and bitumen, are the main source of heat and electrical energy. Recently, biomass has also been a main fuel for renewable energy production in heat boilers. Besides the major constituents (carbon, hydrogen, oxygen), all these fuels contain metal, sulfur, and nitrogen compounds.

During the combustion process, different pollutants, such as fly ash, sulfur oxides (SO<sub>2</sub> and SO<sub>3</sub>), nitrogen oxides (NO<sub>x</sub> = NO<sub>2</sub> + NO), hydrochloride (HCl), and volatile organic compounds, including chlorinated species, are emitted. Ninety-five percent of emitted NO<sub>x</sub> is NO, an insoluble and nonreactive compound that is difficult to remove. Fly

ash contains different trace elements (heavy metals). Mercury is emitted in adsorbed or free forms.

Gross emission of pollutants is tremendous in most countries, all over the world. These pollutants are present in the atmosphere in conditions in which they can affect man and his environment. Air pollution caused by particulate matter and other pollutants acts not only directly on the environment but also by contamination of water and soil, leading to their degradation. Wet and dry deposition of inorganic pollutants leads to acidification of the environment. These phenomena affect the health of people, increase corrosion, and destroy forest and plants.

Different air pollution control technologies are sought. The conventional technologies most often used for air pollution control are: wet FGD (flue gas desulphurization), based on SO<sub>2</sub> absorption in lime or limestone slurry; and SCR (selective catalytic reduction), based on NO<sub>x</sub> reduction over a catalyst to atmospheric nitrogen with ammonia as a reductor. However, technologies which treat different pollutants in one step are of special interest. Electron beam treatment technology (EBFGT) is such a process.

# 2.1. Interaction of electrons with flue gas, components

After irradiation of polluted gas, fast electrons interact with gas, creating various ions and radicals, and 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 vapor concentration, the oxidizing radicals •OH and •HO<sub>2</sub> and excited ions such as  $O(^{3}P)$  are the most important products. The SO<sub>2</sub>, NO, NO<sub>2</sub>, and NH<sub>3</sub> present cannot compete with the reactions because of very low concentrations, but react with N, O, OH, and HO<sub>2</sub> radicals. Ammonia, as mentioned above, is added to the gas to neutralize acids formed in reactions, with aerosol of ammonium sulfate and nitrate being the final products of the reaction. The interaction of electrons with gas forms visible cold plasma (Fig. 1).

# 2.2. $SO_x$ and $NO_x$ removal from fossil fuel combustion flue gases

The method of sulfur and nitrogen oxide removal is based on the oxidation of both pollutants and their reaction with water to form acids. The acids



Fig. 1. The visible glow indicates that cold plasma is formed inside the process vessel, in which gas is irradiated.

are neutralized with gaseous ammonia to form the solid aerosol, a mixture of ammonium nitrate and sulfate, which is the popular nitrogen-bearing component of NPK (nitrogen, phosphor, potassium) fertilizer. There are several pathways of NO oxidation known. In the case of EBFGT the most common are as follows [13]:

$$\begin{split} \mathrm{NO} &+ \mathrm{O}(^{3}\mathrm{P}) + \mathrm{M} \rightarrow \mathrm{NO}_{2} + \mathrm{M}, \\ \mathrm{O}(^{3}\mathrm{P}) &+ \mathrm{O}_{2} + \mathrm{M} \rightarrow \mathrm{O}_{3} + \mathrm{M}, \\ \mathrm{O}(^{3}\mathrm{P}) &+ \mathrm{O}_{2} + \mathrm{M} \rightarrow \mathrm{O}_{3} + \mathrm{M}, \\ \mathrm{NO} &+ \mathrm{O}_{3} + \mathrm{M} \rightarrow \mathrm{NO}_{2} + \mathrm{O}_{2} + \mathrm{M}, \\ \mathrm{NO} &+ \mathrm{HO}_{2}^{\bullet} + \mathrm{M} \rightarrow \mathrm{NO}_{2} + {}^{\bullet}\mathrm{OH} + \mathrm{M}. \end{split}$$

After the oxidation  $NO_2$  is converted to nitric acid in the reaction with  ${}^{\bullet}OH$ , and  $HNO_3$  aerosol reacts with  $NH_3$ , giving ammonium nitrate. NO is partly reduced to atmospheric nitrogen.

$$NO_2 + OH + M \rightarrow HNO_3 + M,$$
  
 $HNO_3 + NH_3 \rightarrow NH_4NO_3.$ 

There can also be several pathways of  $SO_2$  oxidation, depending on the conditions. In the EBFGT process the most important are radiothermal and thermal reactions. Radiothermal reactions proceed through radical oxidation of  $SO_2$  and  $HSO_3$ , which creates ammonium sulfate in the following steps [14]:

$$\begin{split} \mathrm{SO}_2 + {}^{\bullet}\mathrm{OH} + \mathrm{M} &\to \mathrm{HSO}_3 + \mathrm{M}, \\ \mathrm{HSO}_3 + \mathrm{O}_2 &\to \mathrm{SO}_3 + \mathrm{HO}_2^{\bullet}, \\ \mathrm{SO}_3 + \mathrm{H}_2\mathrm{O} &\to \mathrm{H}_2\mathrm{SO}_4, \\ \mathrm{H}_2\mathrm{SO}_4 + 2\mathrm{NH}_3 &\to (\mathrm{NH}_4)_2\mathrm{SO}_4. \end{split}$$

The thermal reaction is based on the following process:

$$\begin{split} &\mathrm{SO}_2 + 2\mathrm{NH}_3 \to (\mathrm{NH}_3)_2 \mathrm{SO}_2, \\ &(\mathrm{NH}_3)_2 \mathrm{SO}_2 \xrightarrow{\mathrm{O}_2,\mathrm{H}_2\mathrm{O}} (\mathrm{NH}_4)_2 \mathrm{SO}_4, \end{split}$$

The total yield of  $SO_2$  removal consists of the yields of thermal and radiothermal reactions, and can be written as follows [15]:

$$\eta_{\mathrm{SO}_2} = \eta_1(\phi, T) + \eta_2(D, \alpha_{\mathrm{NH}_3}, T),$$

where  $\eta, \phi, T, D$ , and  $\alpha_{\rm NH_3}$  are process efficiency, gas humidity, gas temperature, dose deposited (amount of energy transferred to gas by means of irradiation), and ammonia stoichiometry  $(NH_3 \text{ concentration in})$ relation to the stoichiometric value), respectively. The yield of the thermal reaction  $(\eta_1)$  depends on the temperature and humidity, and decreases with the temperature increase. The yield of the radiothermal reaction  $(\eta_2)$  depends on the dose, temperature, and ammonia stoichiometry. The main parameter in  $NO_x$  removal is the dose. The rest of the parameters play a minor role in the process. The high dose is required for high concentrations of  $NO_x$  removal, while  $SO_x$  is removed under proper conditions at low energy consumption.  $SO_x$  removal efficiency equal to 95% is easily achieved, while at the nitrogen oxide concentrations observed in coal- or oil-fired boilers a removal efficiency of 70-80% is observed. This level of pollutant removal is requested by power plant operators due to the existing standards of air pollution control. The scheme of the flue gas treatment process is presented in Fig. 2.

Recent tests have illustrated the possibility of using process applications to treat the polyaromatic



Fig. 2. Scheme presenting sequences of the physicochemical reactions which lead to acidic pollutant removal and solid fertilizer particle formation.

(PAH) and volatile organic hydrocarbons (VOC) present in off-gases in trace concentrations [16], and the process has some features which may allow its application to mercury control as well [17]. The applications of this technology to treatment of municipal and medical flue gases, where the gas flow rate is not so high, would be very economical and feasible from the environmental point of view [18, 19].

# 2.3. Technical applications of the process

The above mechanism of the process, studied in laboratory conditions, was a basis for the technical implementation of the technology. However, in real, industrial conditions, dose distribution and gas flow patterns are important from the technological point of view [20]. These parameters influence the electrons' energy, mass, and heat transfer before, after, and in the process vessel. After humidification and lowering of the temperature, flue gases are guided to the reaction chamber, where irradiation by electron beam takes place. The electrons are introduced into the process vessel via thin  $50\,\mu\text{m}$  titanium foil. NH<sub>3</sub> is injected upstream of the irradiation chamber. The ammonia is used to neutralize the sulfuric and nitric acids, and to form the solid particle aerosol. The size of the aerosol particles is about one micron and the by-product is sticky; therefore high efficiency dust collectors have to be applied downstream of the chemical reactor. Electrostatic precipitators (ESPs) are equipped in the screw conveyers installed at the heated bottom and hammering systems at the electrodes and other filter components. The insulators are protected by air jets. The solid by-product is a high class fertilizer.

In 1970–71, Japanese scientists [5] demonstrated the removal of SO<sub>2</sub> using an electron beam from a linear accelerator (2–12 MeV, 1.2 kW). A dose of 50 kGy at 100°C led to the conversion of SO<sub>2</sub> to an aerosol of sulfuric acid droplets, which were easily removed. Ebara Co. employed an electron accelerator (0.75 MeV, 45 kW) to convert SO<sub>2</sub> and NO<sub>x</sub> into a dry product containing (NH<sub>4</sub>)<sub>2</sub>SO<sub>4</sub> and NH<sub>4</sub>NO<sub>3</sub> which could be used as a fertilizer. By the "Ebara process," two larger scale pilot plants were constructed in Indianapolis, USA, and Karlsruhe, Germany. The Indianapolis plant was equipped with two electron beam accelerators (0.8 MeV, 160 kW)

and had a capacity of  $1.6-3.2 \times 10^{+4} \,\mathrm{m^3/h}$ , with gas containing 1000 ppm SO<sub>2</sub> and 400 ppm NO<sub>x</sub>. In Karlsruhe, two electron accelerators (0.3 MeV, total power 180 kW) were used to treat  $1-2 \times 10^{+4} \,\mathrm{m^3/h}$  flue gas containing 50–500 pm SO<sub>2</sub> and 300–500 ppm NO<sub>x</sub>.

However, the final engineering design technology for industrial applications was achieved at the pilot plants operated in Nagoya, Japan [21], and Kaweczyn, Poland [22]. In the case of the latter, new engineering solutions were applied: doublelongitudinal gas irradiation, an air curtain separating the secondary window from corrosive flue gases, and modifications of the humidification/ammonia system (high enthalpy water or steam injection, ammonia water injection), and others. The obtained results have confirmed the physicochemistry of the process discussed earlier. In Fig. 3 the applied process vessel is presented. The double window was applied to protect the window of the accelerator from the corrosive



Fig. 3. Scheme of the flue gas irradiation vessel applied at the EPS Kaweczyn, Poland, pilot plant and the EPS Pomorzany, Poland, industrial plant.

flue gas atmosphere, and the air curtain protects a secondary window from such effects as well.

These new solutions led to improvements in economic and technical feasibility and final industrial scale plant construction.



Fig. 4. Scheme of an industrial plant for electron beam flue gas treatment: EPS Pomorzany, Szczecin, Poland.



Fig. 5. Removal efficiency of  $SO_2$  and  $NO_x$  as a function of energy absorbed in gas.

Ebara Corporation has constructed a full scale plant in Chengdu, China, mostly for  $SO_x$  removal, and therefore the power of accelerators applied is 320 kW for treatment of  $270,000 \text{ Nm}^3/\text{h}$  of the flue gas. The reported efficiency is 80% for  $SO_x$  and 20%for  $NO_x$  [23].

The flue gas treatment industrial installation is located in EPS Pomorzany in Szczecin, in the north of Poland (Fig. 4) [24]. The installation purifies flue gases from two Benson boilers of 65 MW<sub>e</sub> and 100 MW<sub>th</sub> each. The maximum flow rate of the gases is 270,000 Nm<sup>3</sup>/h and the total beam power exceeds 1 MW. There are two reaction chambers with nominal flow gas rates of 135,000 Nm<sup>3</sup>/h. Each chamber is irradiated by two accelerators (260 kW, 700 keV) installed in series. The applied dose is in the range of 7–12 kGy. The removal of SO<sub>2</sub> approaches 80– 90% in this dose range, and that of NO<sub>x</sub> is 50–60% (Fig. 5). The by-product is collected by the electrostatic precipitator and shipped to the fertilizer plant.

Other laboratory tests have proven the applicability of the technology to the treatment of flue gases from oil-fired boilers [25] and the feasibility of applying the process for the treatment of mercury (in flue gas). A review of the process vessel construction for accelerator-based continuous processing flow systems was given by Berejka [26].

# 2.4. Electron accelerators applied for flue gas treatment

In Table 1 accelerators applied in different laboratory, pilot, and industrial size installations are listed; early project lists are taken from the paper

Table 1.	List of	accelerators	applied	$\operatorname{at}$	$\operatorname{test}$	sites
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Site	Accelerator (MeV; kW)	$\frac{\rm System}{\rm (Nm^3/h)}$	
Tokyo 1970–71	2–12; 1.2 linear	20 l, batch	
Takasaki 1972–74	-; 15 Cocroft–Walton	60	
Ebara 1974–77	0.3-0.75; 30	1000	
Univ. Tokyo 1974–78	1; 90–120 Dynamitron	36-84	
Ebara 1977–78	$0.6-0.75; 2 \times 10-45$	3000 - 10,000	
JAERI 1981	1.5; 30	0.09	
Res. Cottrell 1984–85	$2 \times 0.8; 80$	5300	
Indianapolis 1984–88	$2 \times 0.88; 160$	8000-24,000	
Karlsruhe 1984	190-220; 22	100 - 1000	
Karlsruhe 1984	150-300; 36	60 - 1000	
Badenwerk 1985	$260-300; 2 \times 90$	10,000-20,000	
Warsaw 1990–now	1; 20	1 - 400	
Kawęczyn 1990–94	$700; 2 \times 50$	20,000	
Fujisawa 1991	500; 15	1500	
Matsudo 1992	900; 15	1000	
Nagoya 1992	$800; 3 \times 36$	12,000	
Tokyo 1992	$500; 2 \times 12.5$	50,000	
Mianyang 1999	800; 36	3000 - 12,000	
Chengdu 1999–2004	$800; 2 \times 320$	270,000	
Bejing 2000			
Pomorzany 2002–now	700; $4 \times 268$	270,000	
Maritza East 2003–05	700; $3 \times 12.5$	10,000	

by Frank [27]. The small units were widely used in R&D and industry accelerators with low and medium power. The power supplies had power of up to 100 kW and appropriate window dimensions (due to the cooling requirements).

The most popular accelerators, due to the electron energy requirements (up to 1 MeV), were transformer accelerators like the one presented in Fig. 6. These are the most economical units with high energy efficiency, and their preferability for applications is connected with the fact that the density of flue gas is close to  $1.25 \text{ kg/Nm}^3$ , which assures good penetration of the medium treated by electrons, much deeper in comparison with the liquid or solid phases.

The series of accelerators presented in the picture is produced by the Budker Institute of Nuclear Physics in Novosibirsk and similar ones are produced by the D. V. Efremov Scientific Research Institute of Electrophysical Apparatus in Saint Petersburg, Russia. The ELV units are available from EB Tech Co., Republic of Korea, as well. Vivirad S.A., France,

are made for electron energies in the range of  $70-300 \,\mathrm{kV}$ . However, in this case the single window has to be used in the system, which causes difficulties in the application of these units for treatment of corrosive media. Accelerators of this type are produced by Energy Sciences Inc., USA, and Nissin High Voltage Corporation, Japan.

Very low energy, self-shielded accelerators which can be applied for VOC treatment are manufactured by Advanced Electron Beams (AEB), USA, and the BroadBeam division of PCT Engineering, Davenport, USA. The typical parameters of accelerators which may be applied in emission control technologies are given in Table 2 [28].

The lessons learned from pilot and industrial installations have shown that the technology itself is superior and very competitive with more conventional flue gas treatments.

However, problems reported at industrial plants in China were connected with the failure of accelerators, and similar problems occurred at the installation in Poland. The technology requires application of accelerators of very high power. Therefore the accelerators applied in the mentioned installations were of power higher than  $250\,\mathrm{kW}$  and the power to two of them was supplied from a single high power supply. This was a breakthrough in the technology, since the required availability of the system is equal to 92% of boiler operation time (7000-8000 h a year). The problems related to the accelerators can be avoided by proper system design, manufacturing, and quality control. One reason for some of the quality issues may be that the equipment manufacturers themselves have not performed sufficient research in the development of very high power accelerators.

Table 2. P	arameters	of	selected	electrons	accelerators.
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Accelerator type parameter	EPS-800-375	Dynamitron	ELV-12
Nominal energy	$800\mathrm{keV}$	$1{-}5{\rm MeV}$	$0.6{-}1.0\mathrm{MeV}$
Energy stability	_	$\pm 2\%$	$\pm 1\%$
Nominal beam current	$375\mathrm{mA}$	$50 \mathrm{mA}$	$500\mathrm{mA}$
Beam current stability	-	$\pm 2\%$	$\pm2\%$
Beam power	$300\mathrm{kW}  imes 2$	$250\mathrm{kW}$	$400 \mathrm{kW}$ (three heads)
Scan width	$225\mathrm{cm}$	$200\mathrm{cm}$	$200 \mathrm{~cm}$
Dose uniformity	$\pm 5\%$	$<\pm5\%$	$<\pm5\%$
Mode of operation	Continuous	Continuous	Continuous
Number of scanners	Two heads	One head	Three heads
Total beam power	$600  \mathrm{kW}$	$250\mathrm{kW}$	$400\mathrm{kW}$
Power consumption	$682\mathrm{kW}$	$350\mathrm{kW}$	$500 \mathrm{kW}$
Electrical efficiency	88%	71%	80%
Manufacturer	NHV, Japan	IBA(RDI), USA	BINP, Russia; EB Tech., Rep. of Korea



Fig. 6. An example for an accelerator used for EBFGT. An ELV-3a accelerator (two units) has been applied at the EPS Kaweczyn pilot plant.

offers insulated core transformer accelerators with a cable-connected arrangement  $(500-750 \,\mathrm{keV})$  and integral tank  $(0.8-5 \,\mathrm{MeV})$ . In the above-presented design, a pencil-shaped beam of electrons from a point source is accelerated through a multistage accelerator tube and then scanned by an electromagnetic field through the window into the gas. This type of equipment is normally used for applications requiring accelerating voltages above 300 kV. The second type of EB equipment uses one or more filaments arranged inside a vacuum chamber across the product or the web to be treated or several filaments in parallel to the web direction. These filaments generate a curtain of electrons over the entire product width, without the need for scanning of the electron beam. Its small dimensions make this type of EB equipment especially suited for installation on small mobile units. Modern "low energy EB systems"



Fig. 7. General view of the EBFGT plant for the Sviloza Power Plant, Bulgaria [31].

New developments in the field of electron beam linear accelerators have been reported by PAVAC Industries, Canada. Future use of the electron beam treatment of flue gases technology is predicted. Reliable and moderately priced accelerators are a key factor for progress in the field. Guidelines for feasibility studies have been elaborated [29]. The scheme of a new planned unit at the Svilosa Power Plant, Bulgaria, is presented in Fig. 7 [30].

The plant capacity is  $580 \,\mathrm{MW_t}$  (four coalfired boilers). The flow rate of flue gases will be  $600,000 \,\mathrm{Nm^3/h}$  and the estimated installation construction cost is equal to 26 million euros.

#### 3. Wastewater Treatment

Because of the increasing levels and complexity of polluted effluents from municipalities and industry, current wastewater treatment technologies are often not successful for the remediation of polluted waters and disinfection. Development and implementation of alternative technologies for the cleanup of industrial wastewater, municipal water, groundwater, and drinking water are critical to sustainability in many countries [32]. Very important R&D work on a large scale wastewater treatment application has been performed at the Miami Electron Beam Research Facility [33].

The water purification process uses a product of water radiolysis (Fig. 8) in pollutants degradation [34].

Some of these free radicals formed are oxidative species (•OH), and the others are reductive (H,  $e_{aq}^-$ ) ones. Thus there is competition between oxidation and reduction processes in the system, and the application of synergy with ozone may improve



Fig. 8. Water radiolysis; the main active species formed.

the overall efficiency of destruction of organic pollutants. In this case the ozone reaction with the strong reductive species leads to the formation of hydroxyl radicals (Fig. 9) [35].

Aqueous effluents that have been treated by irradiation include polluted drinking water and liquid industrial and agricultural wastes. However, attention must be paid to the toxicity of the by-products formed in the process, which is the main limitation on its implementation. An industrial plant has been constructed in the Republic of Korea. Based on



Fig. 9. Reactive species formation by radiation in the presence of ozone.



Fig. 10. Three scanners of the accelerator over jet type of wastewater treatment system (left) and the scheme of the accelerator (right) [38].

the data obtained in the laboratory and pilot plant experiments, suitable doses were determined to be around 0.2 kGy for the flow rate of  $10,000 \text{ m}^3$  of effluent per day [36, 37]. In this case a high power accelerator (1 MeV, 400 kW) manufactured by EB Tech Co., Republic of Korea, has been applied (Fig. 10).

High energy electron disinfection of sewage wastewater in flow systems was proposed and tested very early [39] and the hybrid application (COD and microbiological load) is the most promising for future implementations.

A review of the different usages of radiation for wastewater treatment has been given by in Ref. 40. Decoloration and degradation of aqueous solution of reactive azo dye, namely Reactive Red-120 (RR-120), was carried out by electron beam irradiation. The  $BOD_5/COD$  ratio increased upon irradiation and it indicated the transformation of non-biodegradable dye solution into biodegradable solution. This study showed that electron beam irradiation could be a promising method for treatment of textile wastewater containing RR-120 dye [41]. Ionizing radiation such as electron beam irradiation was utilized in the decoloration and degradation of the remaining dye waste after dyeing cotton fabrics with four different dyestuffs based on azo and anthraquinone structures. The results showed that higher decoloration and degradation of dyes was obtained when using an electron beam than when using gamma irradiation under the same conditions. The TOC and COD reductions for all dye solutions were approximately 72-91% and 71-93%, respectively [42]. Other applications of ionizing radiation concern removal of heavy metals from water [43].

In developing wastewater systems it is necessary to consider the penetration range of electrons in the

Table 3. Penetration of electrons of different energies in water (cm) [44].

Energy (MeV)	1.5	3.0	5.0
$R(opt)^*$	0.45	1.05	1.86
$R(50e)^{\dagger}$	0.53	1.21	2.13
2 R(50e)	1.06	2.42	4.26

\*R(opt) — exit dose equals entrance dose;  ${}^{\dagger}R(50e)$  — exit dose equals half entrance dose.

medium, and data for different energies are given in Table 3 [44].

Due to the limited penetration range of electrons in a medium of density equal to  $1000 \text{ kg/m}^3$ , special construction of the irradiation vessel has to be carried out (Fig. 11).

These types of solutions assure the treatment of the entire water stream.

New developments in environmental applications of accelerator-generated electron beams concern degradation of antibiotics and leftover drugs released in liquid effluents. Regarding the use of antibiotics for animal husbandry, administered drugs, metabolites, or degradation products penetrate the ecosystem via the application of manure or slurry to areas used for agricultural purposes or from pasture-reared animals that excrete directly on the land. Degradation of ampicillin in pig manure slurry and an aqueous ampicillin solution has been studied using electron beam irradiation. The results demonstrate that the technology is an effective means of removing antibiotics from manure and bodies of water [45].

Ion exchangers and other substances used in the wastewater treatment process are synthesized employing electron beam grafting or polymerization. Liquid phase polymerization of acrylamide–acrylic acid to form polyelectrolytes used in wastewater



Fig. 11. Process vessels for wastewater irradiation; jet, spray, and upflow.

cleaning was examined employing accelerated electron beam and microwave irradiation methods. Quality indicators such as total suspended matter (TSM), chemical oxygen demand (COD), biological oxygen demand (BOD), and fat, oils, and grease (FOG) were measured before and after the treatment with polymeric flocculants and compared with the results obtained from classical treatment with  $Al_2(SO_4)_3$ . It was found that the combined treatment with polymers and  $Al_2(SO_4)_3$  increases the degree of purification of both wastewaters up to 99% [46].

## 4. Biological Sludge Disinfection

The problem of water contamination by chemical and biological matter is well known. Due to the fact that in many regions deficits of water for municipal, agricultural, and industrial use are observed, water from reservoirs, mainly rivers, is reused many times. Therefore perfect purification and disinfection are necessary for protecting the health of consumers; even so, bottled water and household filters are very popular as a source of good quality drinking water.

The most popular and efficient wastewater purification systems are biological treatment plants.

As a result of the process, these plants are a source of biological sludge, which is a waste (which contains approximately 3% solids; to obtain a higher concentration of solids, a dewatering process is applied). Unfortunately, the sludge of municipal wastewater origin is biologically contaminated by viruses, bacteria, and eggs of parasites. In the case of landfill storage these contaminants survive for many years, due to the fact that even in regions with severe winters the sludge undergoes continuous fermentation and the temperature is much higher than the freezing point of water. Some years ago, different countries solved the problem by dumping at sea, which is prohibited nowadays. The sludge is a good organic fertilizer and is especially good for sandy soil applications, so some countries are applying injection under the soil level, which is not so safe from a health point of view if the field is used for cultivation of food industry crops. Therefore, in the EU, sludge incineration is the main direction taken to solve the problem; however, all combustion processes emit pollutants and greenhouse gases to the atmosphere.

Different methods of disinfection are proposed: heat pasteurization, mixing with lime, and ionizing radiation treatment. Radiation treatment is used



Fig. 12. Scheme of the installation for electron beam sludge treatment.

for hygienization of food items [47] and sterilization of medical products [48]. In the first case low doses are applied to control microbiological contamination of the consumable product (e.g. spices) and in the second case much higher doses are applied to obtain microbe-free products. The destruction of the microbes is achieved by direct and indirect DNA double and single strand breaks and other damage to cell components. Again, due to the high concentration of water in a living organism, the free radicals formed play a most important role in the indirect damage to the living organism's structure. The indirect action of ionizing radiation, which is very similar to that discussed earlier for nonliving physicochemical matter, is connected with water radiolysis and the effect of active species on the DNA strain. Research has shown that sewage sludge can be disinfected successfully by exposure to high energy radiation. Doses of 2-3 kGy destroy more than 99.9% of bacteria present in sewage sludge [10]. Higher doses (up to  $10 \,\mathrm{kGy}$ ) are required to inactivate more radiation-resistant organisms. Both gamma sources (Co-60, Cs-137) and electron accelerators can be used for the irradiation of sewage sludge. Gamma source radiation has better penetration, allowing thicker layers of sludge to be irradiated, although they are less powerful and require a longer irradiation time than electron sources. The irradiated sludge, being pathogen-free, can be beneficially used as manure in agricultural fields as it is rich in nutrients required for the soil. Initial field trials of sludge as manure in agricultural fields of winter wheat crops as well as summer green gram crops have been very encouraging. Since the irradiated sludge is free from bacteria, it can also be employed as a medium for growing bacteria useful for soil like rhizobium and azotobacter produce biofertilizers, which can be used to enhance crop yields. In the case of sludge or soil irradiation,

the high energy accelerators are preferable [49]. The efficiency of sludge disinfection by irradiation was investigated using an electron beam accelerator, with Ascaris ovum as a model. The  $D_{10}$  values obtained for irradiation of residual sludge contaminated with ova depended on the source of the ova: the  $D_{10}$ values were  $788 \pm 172 \,\text{Gy}$  for suspensions of ova extracted from slaughterhouse sludge and 1125  $\pm$ 145 Gy for suspensions freshly prepared by dissection. Ovum suspensions freshly prepared by dissection were more irradiation-proof. Similarly, the  $D_{10}$ value was affected by storage: it was  $1125 \pm 145$  Gy for freshly produced ovum suspensions and  $661 \pm 45 \,\mathrm{Gy}$ for suspensions of ova stored for two months at 48°C in deionized water [50]. An accelerator of  $10 \,\mathrm{MeV}$  at 10 kW is able to irradiate 70 tonnes of sludge a day at a dose of 5–6 kGy, and the concept of such a plant is presented in Fig. 12. The estimated cost of installation is US\$ 4 million [9].

The feasibility study was performed for an Elektronika accelerator (10 MeV, 10 kW) NPO "Torij," Moscow, Russia. In the USA, high power linacs for radiation processing are manufactured by IBA Industrial, USA (formerly known as Radiation Dynamics, Inc.). The company has the longest record of continuous operation in the business of making industrial electron accelerators.

EB Tech Co. Ltd., Republic of Korea, which is very active in the field of accelerator applications for environmental pollution control, tested and prepared a feasibility study for 1 MeV beam application in sludge irradiation (Fig. 13).

Application of this technology may play a very important role in the reclamation of desert land to increase food production and ensure food security. For example, cultivated areas in Egypt existing around the Nile valley and delta represent only 4% of the total area, with the remaining 96% being barren



Fig. 13. Technical solution for sludge treatment on a conveyer [51].

deserts. Reclamation of such desert lands requires water and fertilizer input [52]. The electron beam was studied to enhance the biodegradability of sewage sludge. Changes in the physicochemical characteristics of the sludge were examined with various irradiation doses, sludge thicknesses, and exposure times. Irradiation thickness was suggested as the key factor in the efficiency of solubility of solid organic matter, whereas exposure time would be the most critical parameter in inducing cell lysis in sewage sludge. In addition, biogas production was improved by as much as 22% when the sludge thickness was 0.5 cm with a dose of 7 kGy [53].

Economics is a driving force in accelerator technology environmental applications, and the economic feasibility of irradiation-composting plants for sewage sludge was presented by Hashimoto *et al.* [54]. Some aspects of comparison of feasibility of electron accelerators versus a gamma source are discussed in Ref. 55.

## 5. Conclusions

The electron beam is an important technique for environmental protection applications. Thousands of electron accelerators based on different principles have been constructed and used in the field of radiation chemistry and radiation processing. The progress in accelerator technology means that not only a growing number of units but also lower cost, higher dose rate, more compact size suitable for production lines, beams shaped adequately for the process, reliability, and other parameters that are important in radiation-processing application can be realized. Modern industrial accelerators can provide electron beams with an average power up to several hundred kilowatts, with an energy range suitable for radiation processing  $(0.15-15 \,\mathrm{MeV})$  [56]. The major producers of accelerators are located mainly in the USA, Japan, Russia, Korea, France, and Germany. Several other countries are capable of producing accelerators, among them China, Poland, Canada, and Sweden, but their instruments are usually prototype constructions and are used rather as pilot and R&D installations. Although the present level of accelerator development can satisfy most commercial requirements, this field continues to expand and stimulate radiation-processing activity. On the other hand, the specific demands of the growing field of industrial applications including environmental protection have a strong impact on R&D in accelerator technology.

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