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Organic Pollutants Treatment from Air Using Electron Beam Generated Nonthermal Plasma – Overview

Yongxia Sun¹ and A. G. Chmielewski^{1,2}

¹*Institute of Nuclear Chemistry and Technology, Warsaw,*

²*University of Technology, Warsaw, Poland*

1. Introduction

The municipal and industrial activities of man lead to environment degradation. The pollutants are emitted to the atmosphere with off-gases from industry, power stations, residential heating systems and vehicles. Organic pollutants, mainly volatile organic compounds (VOCs), which are emitted into atmosphere cause stratospheric ozone layer depletion, ground level photochemical ozone formation, and toxic or carcinogenic human health effects, contribute to the global greenhouse effect, accumulate and persist in environment. Regulation on organic pollutants emission into atmosphere has been enforced in many countries. Electron beam (EB) generated nonthermal plasma technology is one of the most promising technologies which has been successfully demonstrated on industrial scale coal fired power plants to remove SO₂ and NO_x from waste off-gases; Meanwhile EB technology has been tested in pilot scale to remove dioxins and Polycyclic aromatic hydrocarbons (PAHs) from off-gases generated from solid waste incinerators and coal fired power plants, good results were obtained. It is a very promising technology to treat multiple pollutants including SO₂, NO_x and organic pollutants simultaneously from industrial off-gases. The principle of EB process to decompose pollutants is following. When the energy of the fast electrons is absorbed in the carrier gas, it causes ionization and excitation processes of the nitrogen, oxygen or water molecules in the carrier gas. Primary species and secondary electrons are formed, and the latter are thermalized within 1 ns in air at 1 bar pressure. These primary species such as ions, radicals or other oxidizing species and the thermalized secondary electrons react with pollutants by a series of reactions to cause pollutants decomposition.

Organic pollutants treatment using EB technology has been studied intensively in recent 30 years mainly in laboratory scale. However less work has been done to review this technology development on organic pollutants treatment. This chapter aims a comprehensive description of organic pollutants treatment using EB generated nonthermal plasma technology. General description of EB generated nonthermal plasma technology will be given in section 2, organic pollutants treatment from air and its recent development will be overviewed in sections 3 & 4, and general mechanism of organic pollutants decomposition in air will be discussed in section 5.

2. Electron beam (EB) generated nonthermal plasma technology

2.1 History

Wet flue gas desulphurisation (FGD) and selective catalytic reduction (SCR) can be applied for flue gas treatment and SO₂ and NO_x emission control. VOCs are usually adsorbed on active carbon, but this process is rarely used for lean hydrocarbon concentrations up to now. All these technologies are complex chemical processes and waste, like wastewater, gypsum and used catalyses, are generated (Srivastava et al., 2001). EB technology is among the most promising advanced technologies of new generation. This is a dry-scrubbing process of simultaneous SO₂ and NO_x removal, where no waste except the by-product is generated. EB technology for air treatment was first used by Japanese scientists in 1970-1971 to study SO₂ removal using an electron from linear accelerator (2-12 MeV, 1.2kW). A dose of 50 kGy at 100°C led the conversion of SO₂ to an aerosol of sulphuric acid droplets, which were easily removed (Machi, 1983). In 1981, Slater (1981) used EB technology to study the decomposition of low concentrations of vinyl chloride (VC) in different base gas (air, nitrogen, and argon).

2.2 EB accelerator

In physics and chemistry, plasma is a state of matter similar to gas in which a certain portion of particles are ionised. Nonthermal plasma means only a small fraction (for example 1%) of the gas molecules are ionized. The most common method for plasma generation is by applying an electric field to a neutral gas. Electrons emitted from electron beam accelerators can ionize gas mixture and generate nonthermal plasma. More than 1000 accelerators have been used in the field of radiation chemistry and radiation processing (Zimek, 1995). The reduction of SO₂ and NO_x pollutants from flue gases, emitted during fuel combustion in electrical power and heat production, is one of the radiation processes which were successfully demonstrated in industrial scale in electric power station (EPS) Pomorzany, Szczecin, Poland (Chmielewski et al, 2004a). A basic principle of an accelerator is that the electric field acts on electrons as charged particles and gives them energy equal to the voltage difference across the acceleration gap. The accelerator types are mainly determined by the method by which electron field is generated. There are three types of accelerators used in air pollutants treatment: transformer accelerator, UHF accelerator and linear microwave accelerator (Zimek, 2005). High power accelerators have been developed to meet specific demands of environmental application and high throughput processes to increase the capacity and reduce unit cost of operation. Table 1 lists accelerators for radiation processing (Zimek, 2005).

Accelerator type	Direct DC	UHF 100-200 MHz	Linear 1.3-5.8GHz
Beam current	< 1.5A	< 100mA	< 100 Ma
Energy range	0.1-5 MeV	0.3-10 MeV	2-10MeV
Beam power	400kW	700 kW	150kW
Efficiency	60-80%	25-50%	10-20%

Table 1. Accelerator for radiation processing (recent development)

2.3 Terminology

In radiation application in environmental protection, there are three terms to be mentioned, **dose**, **G-value** and **removal efficiency (Re)** or **decomposition efficiency(De)**.

In radiation process, it is very important to consider energy consumption for decomposition of pollutants, how much energy (unit: kJ) is consumed/absorbed to decompose amount of pollutants in the base gas (unit: kg). Energy absorbed by per amount of gas is defined as a term of **dose**, unit is **kGy**. $1 \text{ kGy} = 1 \text{ kJ} / \text{kg}$

G-value is defined as the number of molecules of product formed, or of starting material changed, for every 100 eV of energy absorbed. The G value is related to the ionic yield (M / N) by the expression (Willis and Boyd, 1976):

$$G (\text{molecules} / 100 \text{ eV}) = (M / N) \times (100 / W)$$

Where W (measured in electronvolts) is the mean energy required to form an ion pair in the material being irradiated. G value of **1 molecule /100 eV** is equal to a radiation chemical yield of **0.1036 $\mu\text{mol} \cdot \text{J}^{-1}$** .

Removal efficiency (Re) or decomposition efficiency (De) of organic pollutants is defined as below:

$$\text{Re} = (C_0 - C_i) / C_0$$

where C_0 is initial concentration of organic pollutants, unit: ppm (v/v);

C_i is concentration of organic pollutants at i kGy absorbed dose, unit: ppm (v/v).

3. EB treatment organic pollutants

There are two systems applied to study organic pollutants in laboratory scale by using EB generated nonthermal plasma - flow system and batch system. Flow system contains one step: preparation and irradiation of the gas mixture which contains organic pollutants are carried out in on-line system. Batch system contains two steps: first step is to prepare gas mixture which contains organic pollutants into a sealed container, the second step is to put this sealed container under electron beam accelerator for irradiation.

3.1 Aliphatic organic pollutants degradation in flow system under EB-irradiation

3.1.1 Concentration of aliphatic organic pollutants vs. dose

There have been some previous studies of chlorinated hydrocarbons' decomposition in plasma reactors. Slater(1981) studied the decomposition of low concentrations of vinyl chloride(VC) in air, nitrogen, and argon in an electron beam generated plasma reactor. It was found that VC can be effectively removed by electron-beam irradiation at concentrations 3-500 ppm from room-temperature host-gas streams of argon, nitrogen and air. And at low dose the specific energy required fell in the range $2.5 < G < 10$ molecules removed per 100 eV. HCl was one of main products.

Vitale et al.(1997a) studied decomposition low concentration of ethyl chloride (EC) and vinyl chloride(VC) in atmospheric air streams by an electron beam generated plasma reactor. The gas was prepared by mixing dry air with standard VC (3925ppm VC in air) or EC (3717ppm EC in air). The gas entered the reactor at atmospheric pressure and ambient temperature. The electrons entered the front of the reactor, and VOC contaminated gas entered the rear of the reactor. The VOC contaminated gas thus flowed counter-current to

the electron beam. A Hewlett-Packard 5890 gas chromatograph and a HP-5971-A mass spectrometer were used to analyze VOCs concentration. The energy requirements for 90% decomposition of VC and EC were reported as a function of inlet concentration. VC requires less energy for decomposition than EC.

Similar experiments were carried out to decompose 1,1-dichloroethane (1,1-DCA), 1,1-dichloroethylene (1,1-DCE), 1,1,1-trichloroethane (1,1,1-TCA), trichloroethylene (TCE) using EB by the same research group (Vitale et al., 1996, 1997b–d). It was found that decomposition efficiency of chlorinated compounds was: TCE > 1,1-DCE > 1,1-DCA, 1,1,1-TCA.

Won et al. (2002) studied the decomposition of perchloroethylene (PCE), trichloroethylene (TCE), dichloroethylene (DCE) in dry air. An electron accelerator of ELV type, with electron energy 0.7 MeV, maximum beam current 35 mA, maximum power 25 kW was used for irradiation. Over 80% TCE was decomposed at 20 kGy dose at initial concentration below 2000 ppm. The order of decomposition efficiency of these compounds was: TCE > PCE > DCE. Hirota et al. (2004) studied dichloromethane decomposition under EB irradiation and found that it was very difficult to treat dichloromethane.

For non-chlorinated organic compounds, 20 VOCs divided into five groups were investigated by Hirota et al. (2004), among them, 13 VOCs were aliphatic organic compounds. The order of decomposition VOCs in air was: cyclohexadiene > cyclohexane > benzene (group I); trans-hexane > 1-hexane (group II); heptane > hexane > pentane (group IV); and trichloroethylene > methanol >> acetone > CH₂Cl₂ (group V). Organic substances with long carbon chains readily succumbed to electron-beam treatment.

3.1.2 Different base gas mixtures influence on the decomposition efficiency of aliphatic organic pollutants

Won et al. (2002) studied TCE decomposition in different gas mixtures and found that the order of decomposition efficiency of TCE in different gas mixtures was: oxygen > air > H₂ > He.

3.1.3 Water concentration

In order to clarify OH radical influence on the chlorinated hydrocarbons (Cl-HC) decomposition, Won et al. (2002) tested TCE and PCE decomposition of air mixtures with different water vapor concentrations, and found that the decomposition efficiency of TCE and PCE increased less than 10% in the presence of water vapor compared with that in the dry air.

3.1.4 Irradiation products

The irradiation products of DCE, TCE and PCE in dry air under EB-irradiation were investigated by Won et al. (2002) and it was found that CO and CO₂ were the irradiation products. For PCE, CO₂ formation was above 40% at 15 kGy absorbed dose. Vitale et al. (1997a) also reported that CO, CO₂ and HCl as main irradiation products when they studied ethyl chloride and vinyl chloride decomposition in air. Prager et al. (1995) studied DCE, TCE and PCE degradation in dry or humidified synthetic air, they identified HCl, CO, chloromethanes, chloroacetyl chloride and phosgene as main products.

3.2 Aliphatic organic pollutants degradation in batch system under EB-irradiation

3.2.1 Concentration of aliphatic organic pollutants vs. dose

Chloroethylene can be effectively decomposed by EB irradiation in the order of PCE > TCE > trans-DCE > cis-DCE (Hakoda et al., 1998a, 1998b, 1999, 2000, 2001; Hashimoto et al.,

2000). Sun et al. (2001, 2003) and Sun and Chmielewski (2004) studied 1,1-DCE, cis-DCE, trans-DCE decomposition under EB irradiation and found that the order of decomposition DCEs in air was 1,1-DCE > trans-DCE > cis-DCE. Decomposition efficiency of chloroethylene increases with the absorbed dose increase. The initial concentration of chlorinated ethylene was in below 2000 ppm and the water concentration in the air mixture was 200–300 ppm. Son et al. (2010a) studied decomposition of butane in EB irradiation in batch system, it was found that removal efficiencies of butane were 40% at 2.5 kGy and 66% at 10 kGy, when the initial concentration of butane was 60 ppm.

3.2.2 Different base gas mixtures influence on the decomposition efficiency of aliphatic organic pollutants

Different base gases influencing on the decomposition efficiency of butane were studied (Son et al, 2010a), it was found that decomposition efficiency of butane was extremely low when the background gas was He, in contrast to the efficiencies with background gases of N₂ and air. Decomposition efficiencies of butane was 23% in He, 63% in N₂ and 70% in air at 10 kGy absorbed dose.

3.2.3 Water concentration

Water influence on the TCE decomposition and irradiation products of TCE under EB-irradiation were studied by Hakoda et al. (2000). It was found that when water concentration was below 1000 ppm, there was no big difference between process efficiency for dry and humid air for TCE decomposition and dichloroacetyl chloride, carbon monoxide, carbon dioxide, phosgene and small amount of chloroform irradiation products' formation.

This result agrees well with that water vapor effect on the TCE decomposition under EB-irradiation in a flow system (Won et al., 2002). Sun et al. (2001) made a computer simulation of 1,1-DCE decomposition in air in a batch system and found that a reaction pathway of OH radical contributes less than 10% for 1,1-DCE decomposition. When water vapor concentration increased to 2.5%, yield of gaseous products decreased, that means the aerosol products are possibly formed (Hakoda et al., 2000).

3.2.4 Ozone

Hakoda et al. (1999, 2000, 2001) investigated O₃ influence on the trans-DCE, cis-DCE and TCE decomposition by using EB-irradiation, it was found that O₃ enhanced decomposition of trans-DCE only, cis-DCE and TCE were not affected.

3.2.5 Irradiation products

From environmental protection point of view, it is very important to identify by-products formation from Cl-HC degradation. Radiolytic products of trans-DCE, cis-DCE, TCE, PCE under EB-irradiation were reported by Hakoda et al. (1999, 2000, 2001). Chloroacetyl chloride and dichloroacetyl chloride were the main organic products for DCE (trans and cis) and TCE degradation; CO and CO₂ were inorganic products and their formations were below 25% based on carbon balance. Chmielewski et al. (2004b), Sun et al. (2003) and Sun & Chmielewski (2004) studied cis-DCE and trans-DCE degradation under EB-irradiation. Chloroacetyl chloride was not observed as degradation products, but it was a degradation product for 1,1-DCE (Sun et al., 2001). Son et al. (2010a) studied butane decomposition under EB irradiation and identified CO₂, acetaldehyde, acetone, 2,3-butanedione, 2-butanone, and 2-butanedinitrile as degradation products of butane.

3.3 Aromatic organic pollutants degradation in flow system under EB-irradiation

In this section, besides aromatic chlorinated hydrocarbons degradation, decomposition of some nonchlorinated aromatic organic compounds will be discussed, too.

3.3.1 Concentration of aromatic organic pollutants vs. dose

Xylene and chlorobenzene decompositions in a flow system under EB-irradiation (Hirota et al. 2000, 2002) were studied, it was found that decomposition efficiency of xylene was higher than that of chlorobenzene, and about 50% chlorobenzene was decomposed at an absorbed dose of about 10 kGy at the initial concentration of chlorobenzene being 10–40 ppm. Kim (2002) studied decomposition of benzene and toluene, it was found that the decomposition efficiency of toluene was higher than benzene, and about 80% benzene was decomposed at 16 kGy when the initial concentration of benzene was smaller than or equal to 130 ppm. Han et al. (2003) studied toluene, ethylbenzene, o-, m-, p-xylenes and chlorobenzene decomposition in air. The order of the decomposition efficiency of selected VOCs from high to low was : toluene > ethylbenzene > benzene; p-xylene > m-xylene > o-xylene. About 44.7 % toluene and 43.2% ethylbenzene was decomposed at 10 kGy, while 85% chlorobenzene was decomposed. The decomposition efficiency of ethylbenzene and toluene was significantly increased about 50% with the addition of chlorobenzene comparing without chlorobenzene addition.

3.3.2 NH₃ influence on the decomposition efficiency of aromatic organic pollutants

Effect of ammonia on the decomposition of PAHs was observed when an electron beam process was applied to treat multiple pollutants (SO₂, NO_x, PAHs) with NH₃ addition from industrial off-gases emitted from EPS, Kawęczyn, Poland (Chmielewski et al., 2002). NH₃ addition enhanced PAHs removal efficiency. Hirota et al. (2000) studied chlorobenzene decomposition in air mixture, it was found that addition of NH₃ enhanced the dechlorination of chlorobenzene. About 65% of chlorine in reacted chlorobenzene was dissociated from carbon with electron beam at doses of 4 and 8 kGy. Ammonia addition enhanced the dechlorination to 80%.

3.3.3 Water concentration

Effect of water vapor on the decomposition of toluene was investigated by Kim (2002). It was found that the water vapor injection leads to 15–20% removal efficiency increase for toluene compared to the process without water injection. Water influences decomposition of toluene higher than TCE and PCE, OH radical plays an important role for aromatic hydrocarbon decomposition.

3.3.4 Irradiation products

Degradation products of chlorobenzene and xylene in an air mixture were studied by Hirota et al. (2000, 2002). The gaseous products of xylene degradation were identified to be formic, acetic, propionic, and butyric acids and/or the corresponding esters with CO and CO₂. Approximately 30% of the reacted xylene was the gaseous products at a dose of 8 kGy. Organic and inorganic chlorine presence in gaseous, aerosols and residues were investigated by same authors for chlorobenzene degradation. Inorganic chlorine was mainly presented in gaseous products, while organic chlorine was presented in aerosols and residues aerosols and residues. Some aerosol products were formed from chlorobenzene degradation, and 3% of the

aerosol products were identified to be carboxylic acids or esters (Hirota et al., 2000). Aerosols, benzaldehyde, dipropyl 1,2-benzenedicarboxylic acid, nitromethane were reported as toluene main degradation products in dry air under EB irradiation; while methyl chloride, dipropyl 1,2-benzenedicarboxylic acid, toluene, nitromethane were reported as main degradation products of ethylbenzene/chlorobenzene mixture in dry air. Trace amount of acetone, hexane, benzene was also observed (Han et al., 2003).

3.4 Aromatic organic pollutants degradation in batch system under EB-irradiation

3.4.1 Concentration of aromatic organic pollutants vs. dose

Decomposition of aromatic organic compounds in a batch system vs. dose under EB-irradiation was studied by Hirota et al. (2002, 2004), Ostapczuk et al. (1999), Sun et al. (2008) and Hashimoto et al. (2000). Decomposition efficiency of these compounds increase with the absorbed dose increase, 4-chlorotoluene (4-CTO) decomposition as an example was presented in Fig. 1. The order of decomposition efficiency of these compounds was: xylene > chlorobenzene > benzene > hexane > cyclohexane. For 4-chlorotoluene (4-CTO) and 1,4-dichlorobenzene (1,4-DCB), no apparent decomposition efficiency of these two compounds was observed (Fig.2).

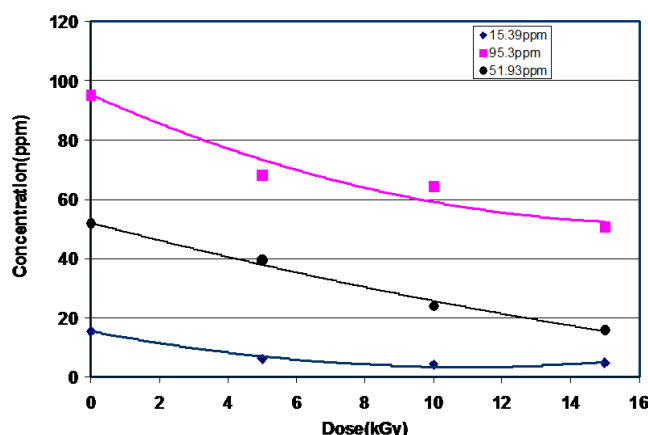


Fig. 1. 4-Chlorotoluene decomposition in air mixture in an electron beam generated non-thermal reactor.

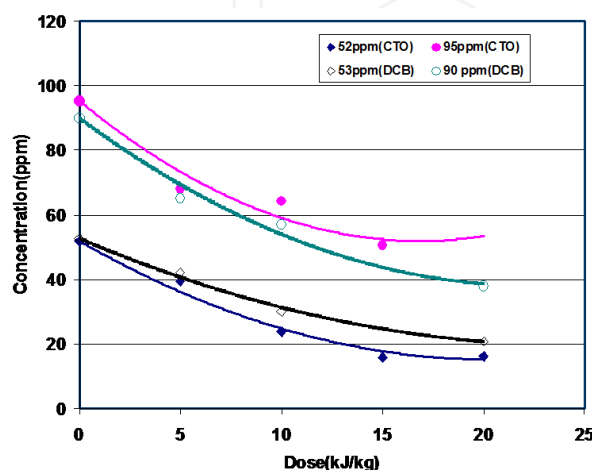


Fig. 2. Decomposition efficiency comparison between 4-chlorotoluene and 1,4-dichlorobenzene.

The decomposition efficiency of chlorinated aliphatic hydrocarbons using EB irradiation is more efficient than that of chlorinated aromatic hydrocarbons. 25.0 kGy is sufficient to remove over 97% 1,1-DCE and 98.0% trans-DCE at initial concentration of 1,1-dichloroethylene (DCE) and trans-dichloroethylene being 903.8 ppm and 342.0 ppm, respectively; while 60% 1,4-dichlorobenzene (DCB) at initial concentration being 90ppm was removed at 57.9 kGy. This result is comparable with decomposition of chlorobenzene. Hakoda et al. (1998b) and Hashimoto et al. (2000) studied degradation of chlorobenzene/air using EB irradiation in batch system, it was found that 40% chlorobenzene was removed under EB-irradiation at 37.7 kGy dose (calculated by N₂O gas dosimeter) for initial concentration of chlorobenzene being 102 ppm. Sun et al. (2007a) studied 1-chloronaphthalene and found that over 80% 1-chloronaphthalene was removed at 57.9 kGy under EB-irradiation for low initial concentration of 1-chloronaphthalene (12~30 mg/m³) in air mixture. Energy consumption for decomposition 1,4-dichlorobenzene was lower than that of 1-chloronaphthalene. Therefore, the observed order in easily decomposition chlorinated hydrocarbons is: 1,1-DCE > trans-DCE > cis-DCE > 1,4-DCB > 1-chloronaphthalene. Based on this work and other's work, we learn that: For chlorinated aliphatic hydrocarbons, the more chlorinated compounds is, the more it is easy to be decomposed.

Aliphatic hydrocarbons is more easily decomposed than aromatic hydrocarbons. For aromatic hydrocarbons, compounds with less benzene ring are easily to be decomposed.

3.4.2 Different base gas mixtures influence on the decomposition efficiency of aromatic organic pollutants

Toluene decomposition at different background gases in a batch system was studied by Kim (2002). The order of decomposition efficiency of toluene in different background gases is: N₂ > air > O₂ > He. This order is different from the order for TCE decomposition in a flow system (Won et al., 2002).

We studied 1,4-dichlorobenzene (1,4-DCB) decomposition in different base gas mixtures at the initial concentration of 1,4-DCB being 50 ppm, the similar phenomenon was observed (Sun et al, 2006). The decomposition efficiency of 1,4-DCB in nitrogen is higher than that in air and much more higher than in 1.027% NO-N₂ mixture(N₂ as balance gas) (Fig.3) , this phenomenon agrees well with toluene decomposition in different gases (Kim, 2002).

3.4.3 Water concentration

Effect of water vapor on the decomposition of aromatic compounds in a batch system under EB-irradiation was investigated by Kim (2002). Four percent water vapor injection leads to 5–10% increase of VOC removal efficiency for both toluene and benzene, and effect of water vapor influence on the decomposition of toluene under EB irradiation in a flow system is higher than that in a batch system.

3.4.4 Irradiation products

Benzaldehyde and phenol were reported as products when Ostapczuk et al. (1999) studied styrene decomposition in air under EB irradiation, the removal efficiency of styrene was ranged from 83-95%. The humidity in air mixture was ranged from 0.3% to 1.6%. In order to obtain information of by-products produced from toluene destruction, we carried out experiment at higher inlet concentration of toluene at higher absorbed dose. More than 97%

toluene was removed from gas phase at 53.6 kGy absorbed dose when inlet concentration of toluene was 151.9 ppm. A GC-MS spectrum of toluene/air mixture after EB-irradiation is presented in figure 4. A trace amount of benzaldehyde was eluted at retention time 7.735 min in figure 4 and was identified by our carefully comparing mass spectrum of this compound with a reference mass spectrum of benzaldehyde provided by Wiley library (figures 5a & 5b). Trace amount acetone was also found in our experimental condition (Sun et al., 2009a).

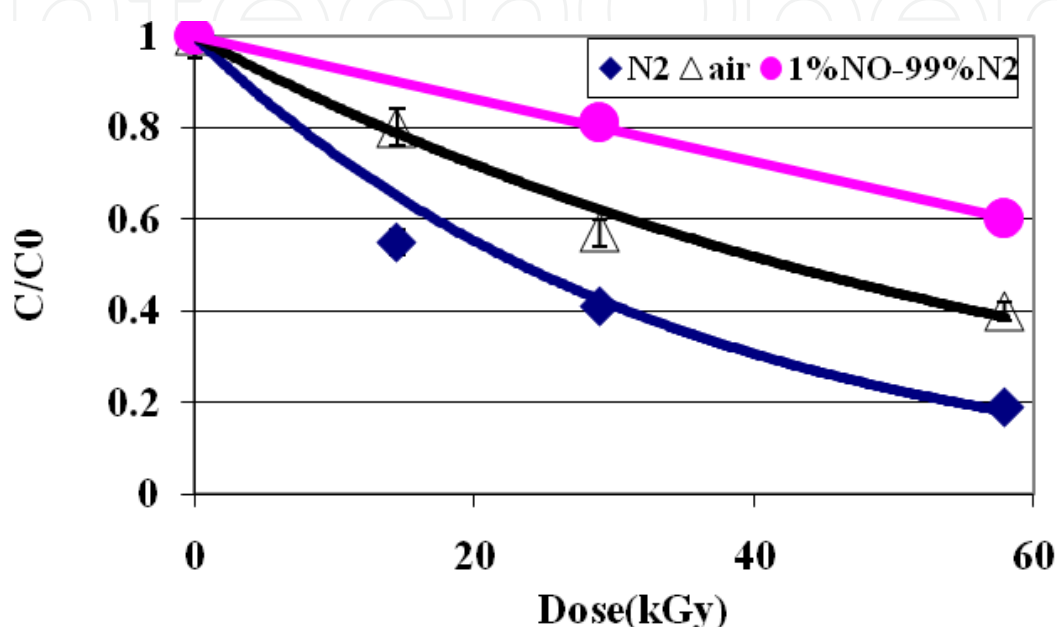


Fig. 3. 1,4-Dichlorobenzene decomposition in different gas mixture



Fig. 4. A GC-MS spectrum of toluene/air mixture after EB-irradiation (inlet concentration of toluene was 151.9 ppm, dose was 53.6 kGy).

Benzaldehyde as by-product of degradation of toluene was also reported (Han et al., 2003; Kim et al., 2005). Trace amount of acetone was found based on Han et al.'s work (2003). Besides these, Aerosols and benzene were reported as by-products in both works (Han et al.,

2003; Kim et al., 2005). Han et al.(2003) also identified dipropyl 1,2-benzenedicarboxylic acid, nitromethane and trace amount of hexane as by-products of degradation of toluene. For degradation of 4-chlorotoluene in air mixture, chlorobenzene (C_6H_5Cl , retention time was 4.910 min) and 4-chlorobenzaldehyde (ClC_6H_4CHO , retention time was 12.502 min) were identified as by-products. A GC-MS spectrum of 4-chlorotoluene/air mixture after EB-irradiation was presented in figure 6, a compound eluted at retention time 7.590 min of the GC-MS spectrum was identified as 4-chlorotoluene (Sun et al., 2008).

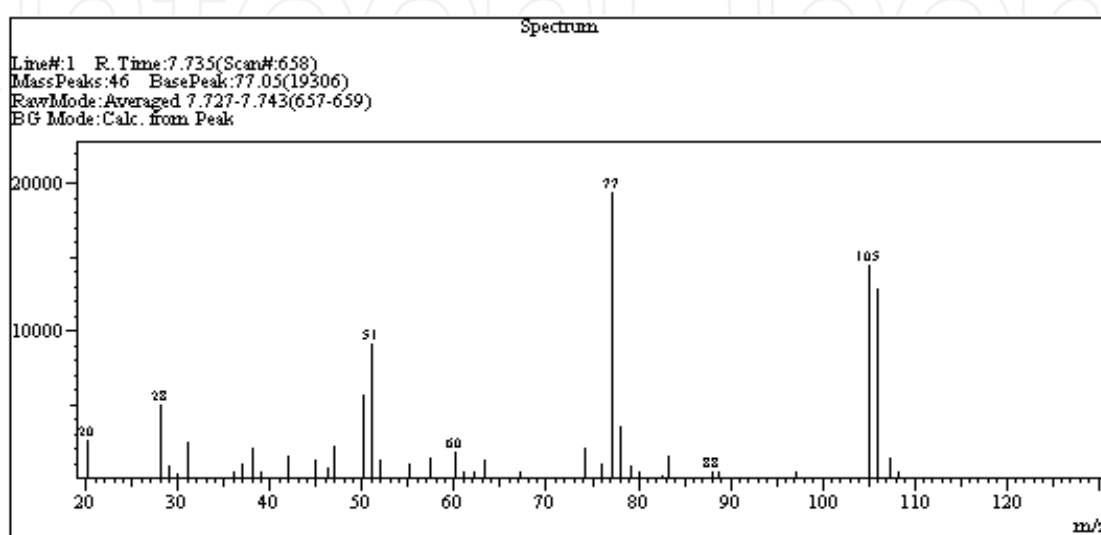


Fig. 5a. A mass spectrum of by-product which eluted at 7.735 min retention time

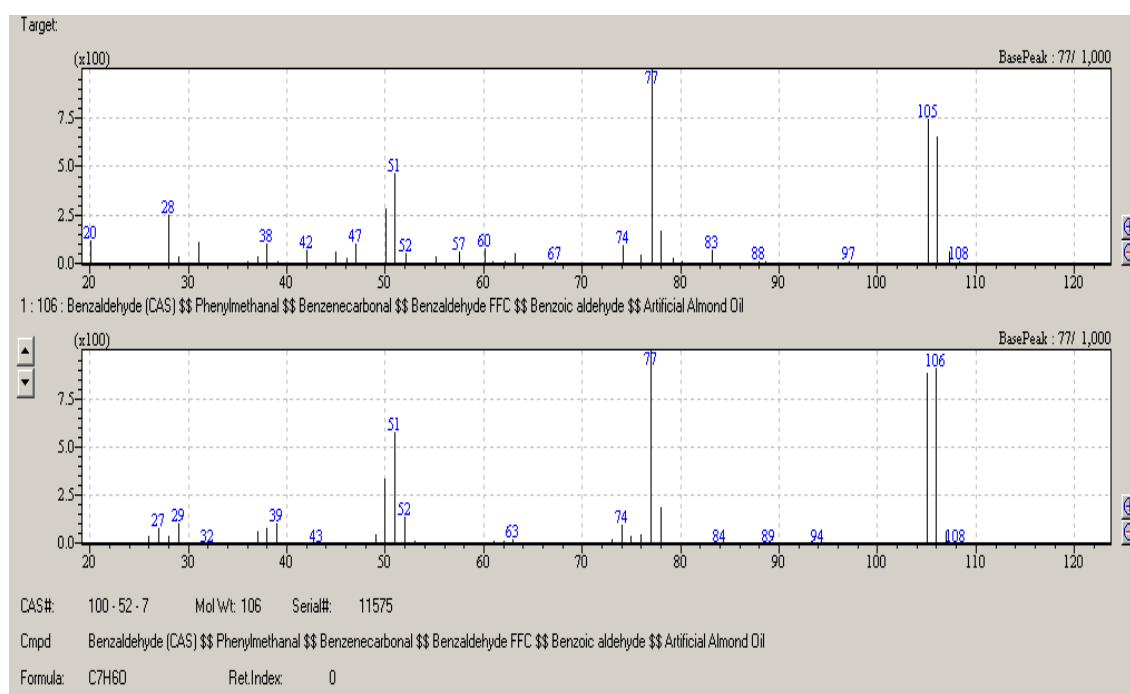


Fig. 5b. A mass spectrum of the compound which eluted at 7.735 min retention time and its reference mass spectrum of benzaldehyde

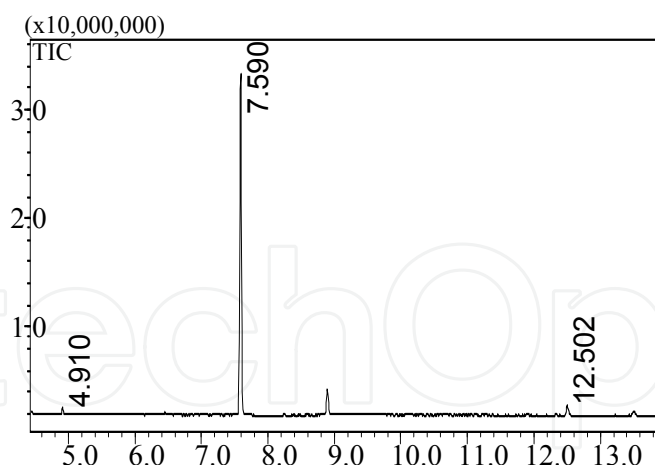


Fig. 6. A GC-MS spectrum of 4-chlorotoluene/air mixture after EB-irradiation

3.5 PAHs and Dioxin removal from waste off-gas under EB-irradiation

Dioxins reduction from waste incinerator was studied using EB technology in Japan (Hirota et al., 2003) and in Germany (Paur et al., 1998). Hirota et al. (2003) studied reduction the emission of polychlorinated dibenzo- p-dioxins (PCDD) and polychlorinated dibenzofurans (PCDF) in a flue gas of 1000 m³N/h from the municipal solid waste incinerator (MSWI), located at Takohama Clean Center which treats 450 t (150 t * 3 furnaces) of solid waste in 1 day, at a temperature of 200 °C. they found that more than 90% PCDD/Fs was removed at 14 kGy when initial concentration of PCDD was in the range of 0.22-0.88 ng-TEQ/m³N and PCDF in the range of 0.35-12.4 ng-TEQ/m³N. Paul et al. (1998) also reported that over 90% PCDD was removed at 12 kGy dose for initial concentration of PCDD being 21-110 ng/m³N (AGATE-M plant, Germany).

16 kinds of toxic PAHs were investigated under electron beam irradiation in the pilot plant in Electric Power Station Kawęczyn, Poland (Chmielewski, et al., 2003). The investigation was carried out under the following experimental conditions: flue gas flow rate 5000 Nm³/h; humidity 4.5%; inlet concentrations of SO₂ and NO_x that were emitted from the power station were 192 and 106 ppm, respectively; ammonia addition was 2.75 Nm³/h; alcohol addition was 600 l/h, the absorbed dose was 8 kGy. The results was presented in Fig.7. It was found that under these experimental conditions the concentrations of naphthalene (NL, C₁₀H₈), acenaphthene (AC, C₁₂H₁₀), fluorene (C₁₃H₁₀), phenanthrene (C₁₄H₁₀), anthracene (C₁₄H₁₀) were decreased, while the concentrations of acenaphthylene (C₁₂H₈), fluoranthene (C₁₆H₁₀), pyrene (C₁₆H₁₀), benzo(a)anthracene (C₁₈H₁₂), chrysene (C₁₈H₁₂), benzo(b p k)fluoranthene (C₂₀H₁₂), benzo(e)pyrene (C₂₀H₁₂), benzo(a)pyrene (C₂₀H₁₂), perylene (C₂₀H₁₂), dibenzo(a; h)anthracene+indeno(1,2,3-cd) pyrene (C₂₂H₁₄), benzo(g; h; l)perylene (C₂₂H₁₂) were increased. Removal efficiencies of SO₂ and NO_x were 61.6% and 70.9%, respectively. The concentration of hydrocarbons of small aromatic ring (PAHs, like naphthalene (C₁₀H₈), acenaphthene (C₁₂H₁₀), fluorene (C₁₃H₁₀), anthracene (C₁₄H₁₀)) was reduced, while the concentration of fluoranthene was increased remarkably after irradiation.

Similar experiments were carried out in EPS Kawęczyn with ammonia presence but without alcohol addition (Chmielewski et al, 2002; Ostapczuk et al, 2008a). It was found that removal efficiency of PAHs ranges from 40% up to 98%.

Callén et al. (2007) studied PAH removal from lignite-combustion flue gas from Bulgarian Maritza-East thermal power plant (TPP) and obtained that PAHs concentration after EB

irradiation resulted in ~ 10 fold decrease in studied PAHs emissions. The removal efficiency of PAH removal at the dose of 4 kGy was 85% (weight/weight). High PAH removal efficiency was obtained especially for 2 and 3 rings PAH, this result was similar to that obtained in our previous work (Chmielewski et al., 2003).

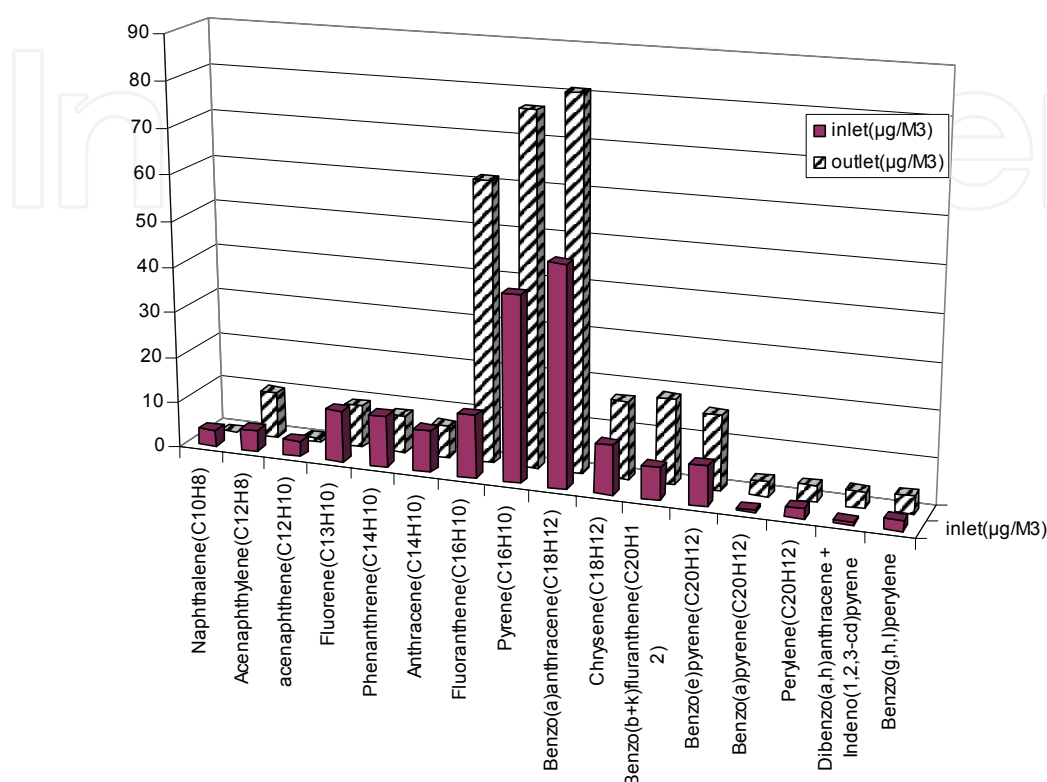


Fig. 7. EB irradiation influence on PAHs removal.

The concentration of PAHs in by-product was also examined. It was relatively low, varied from few up to 12 µg per kg of fertilizer for the experimental work carried out in EPS Kawęczyn, Poland. Less than 3% of PAH were removed in adsorption on the by-product surface (Ostapczuk, et al., 2008a). The study in Maritza-East TPP allowed PAH assessment in solid by-products obtained from EB lignite-combustion flue gas. The determined PAH content was reasonable, ~ 60 microg/kg and was lower than PAHs background in Bulgarian soils. These results demonstrated the insignificant role of adsorption for PAHs removal (Callén et al., 2007).

Naphthalene (NL) and acenaphthene (AC) decomposition in gas mixture was studied in lab scale experiment (Ostapczuk, et al., 2008b). It was found that NL was more easily decomposed than AC, G-values for these two compounds were 1.66 and 3.72 mol/100 eV for NL and AC at the dose of 1 kGy, respectively. Humidity influencing on NL and AC decomposition was studied. About 26% and 50% NL were decomposed at 1 kGy dose in dry air (90 vol% N₂; 10 vol% O₂ and 160 ppmv NO) and in humid air (84 vol% N₂; 10 vol% O₂ and 6 vol% H₂O), respectively. NL concentration in both mixtures was on the level of 10–11 ppmv. For AC, about 45% (in O₂) and 82% AC (in humid O₂, 94% O₂ + 6% H₂O) were decomposed at 1 kGy dose for the initial concentration of AC being 160 ppm. Two-ringed naphthol and nitronaphthalene; one-ringed 2,6-diethylbenzoquinone, indane, 1,2- and 1,4-dimethoxybenzenes and carbon oxides were identified as by-products of NL decomposition.

4. A novel hybrid EB-catalyst technology to treat organic pollutants

Electron beam (EB) irradiation is one of the most promising technologies for purification of dilute pollutants, mostly VOCs, with high flow-rate gas stream released from industrial off-gases. Under EB irradiation, VOCs are oxidized into irradiation by-products as well as CO₂ and CO. However, some of these irradiation by-products have adverse effect on environment and human beings.

A new technology which combines EB and catalyst together to treat aromatic VOCs, e.g., toluene (Kim et al., 2004, 2005; Jeon et al., 2008) styrene (Kim, 2005), o-Xylene (Hakoda et al., 2008a, 2008b) and ethylbenzene (Son, et al, 2010b) was developed in the aim of enhancing higher oxidation efficiencies of VOCs into CO₂. Removal efficiency of toluene, styrene and ethylbenzene increased by 10%, 20%, and 20% in an EB-catalyst hybrid system in comparison with that achieved in catalyst-only method at approximately 10 kGy absorbed dose (Kim, et al, 2005). Removal efficiency of ethylbenzene in the EB-catalyst hybrid was 30% higher than that of EB-only treatment. Ethylbenzene was decomposed more easily than toluene by EB irradiation. The G-values for ethylbenzene increased with initial concentration and reactor type: the G-values vary in the range of 7.5-10.9 (EB-only) and 12.9-25.7 (EB-catalyst hybrid) by reactor type at the initial concentration of ethylbenzene being 2800 ppm. Son et al. (2008) and Jeon et al. (2008) also studied different catalysts (Pt, Pd, Cu and Mn) and humidity influence on removal efficiency of toluene using EB-catalyst hybrid system. It was found that removal efficiency of toluene was increased by 36.9%, 35.3% and 22% in the presence of Pt, Pd, Mn and Cu catalysts comparing with EB only for initial concentration of toluene being 1500 ppm, the selectivity to CO₂ with Pt and Pd coupling were relatively higher than those of Cu and Mn. Especially the CO₂ selectivity of EB-Pt coupling was significantly high at a relatively low absorbed dose. The catalytic activity for EB-catalyst coupling system was in the order of Pt, Pd, Mn and Cu. There was no significant difference of removal efficiency of toluene among 0.1, 0.5 and 1.0 wt% loading of catalyst. No significant water effect was observed in EB-catalyst hybrid system (Son et al, 2008).

Other type of catalysts such as TiO₂ (Hakoda, et al., 2008a) was used to study xylene decomposition under EB irradiation in lab scale experimental work. It was found that removal efficiency of xylene and CO₂ formation were increased with the presence of TiO₂ catalyst, the similar phenomenon was observed when Kim studied toluene decomposition using Pt as catalyst (Kim et al., 2005).

Hakoda, et al.(2008b) also studied xylene decomposition using MnO₂ (an O₃ decomposition catalyst), γ -Al₂O₃ was selected as a base material of the catalyst. The combination process at temperatures of about 100°C using MnO₂ placed downstream enhanced the oxidation of the by-products of xylene into CO₂ by active oxygen produced from the O₃ decomposition when the MnO₂ bed was placed downstream of an irradiation space. Furthermore, EB-irradiated γ -Al₂O₃ surface was found to be active, and the oxidation of organics was enhanced by primary electrons. The combination process using γ -Al₂O₃ reduced dose to 33% of a single EB process to obtain the same conversion of xylene to CO₂.

Ighigeanu (et al., 2008) studied VOCs (Toluene, hexane + toluene mixture diluted in air) decomposition by using combination of three different technologies (EB, microwave (MW) and catalysts): (EB + MW+ catalyst); (MW + catalyst) and (EB+catalyst). They found that decomposition efficiency (De) and oxidation efficiency (Eo) of toluene increased significantly for the (EB+MW+catalyst) treatment as compared with (MW + catalyst) and (EB + catalyst) treatments, at initial concentration of toluene being in the range of 180 ppm –

523 ppm; and CO₂ and CO concentrations after treatment were higher for the (EB+MW+catalyst) treatment than for (MW+catalyst) and (EB+catalyst) treatments. De and Eo of toluene were, respectively, as follows: 59.5% and 82.2% for the (MW + catalysis), 77.2% and 87.1% for the (EB + catalyst) and 92.8% and 90.5% for the (EB + MW + catalyst). For air mixture contained toluene and hexane, removal efficiency of toluene and hexane in (EB + MW + catalyst) system was higher than that in (MW + catalyst) system or in (EB + catalyst) system, about 88.5% toluene and 87.8% hexane were decomposed for initial toluene and hexane concentration being 250 ppm, respectively.

5. Mechanism of organic pollutants degradation by using EB technology

In order to obtain high decomposition efficiency of organic pollutants and less toxic by-products, it is very important to understand mechanism of organic pollutants degradation under EB irradiation. In this section, we will discuss mechanism of two groups (chlorinated and nonchlorinated) organic pollutants. General mechanism of organic pollutants decomposition in gas phase under EB irradiation is illustrated in Fig. 8.

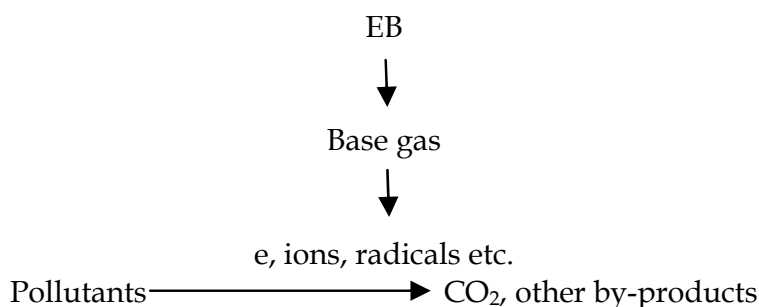


Fig. 8. General mechanism of organic pollutants decomposition under EB irradiation

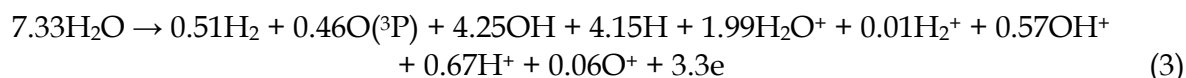
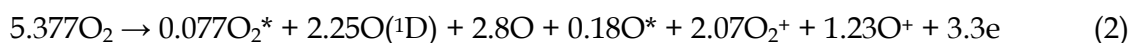
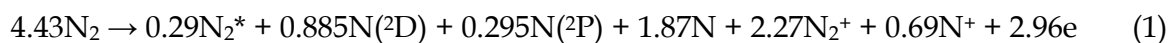
5.1 Chlorinated organic compounds (Cl-HC)

5.1.1 General mechanism of chlorinated aliphatic hydrocarbon decomposition in air mixture

Computer simulations of chlorinated aliphatic hydrocarbons' decomposition in air mixture were carried out and discussed in details (Nichipor et al., 2000, 2002, 2003, 2008; Sun et al., 2001, 2007b, 2009b). The general mechanism of aliphatic hydrocarbon decomposition in an air mixture is described below:

When fast electrons from electron beams are absorbed in the carrier gas, they cause ionization and excitation processes of the nitrogen, H₂O and oxygen molecules in the carrier gas. Primary species and secondary electrons are formed. The secondary electrons are thermalized fast within 1 ns in air at 1 atmosphere.

The G-values (molecules/100 eV) of main primary species are simplified as follows (Mätzing, 1989):



Where G-values of molecules decomposed are listed in the left side of the arrows, and G-values of species formed from the pure k type molecules that absorb an energy of 100 eV are listed in the right side of the arrows. These primary species and thermalized secondary electrons cause Cl-HC decomposition. Based on our and others published work , we know that several type reactions cause Cl-HC degradation.

Positive ions charge transfer and particle dissociation reactions

It is well known that : when air component and molecule are ionized and excited, a large amount of N₂⁺, O₂⁺ , N⁺, O⁺, H₃O⁺(if water concentration is high) are formed, and their ionization potential energy (IE) is higher than that of Cl-HC (see Table 2). The positive charge transfer reaction, positive ion cluster reaction, or particle dissociation reaction occur (Spanel, et al., 1999a, 1999b).

Molecule	IE(eV)	EA(eV)	PA(kJ.mol ⁻¹)
N ₂	15.58		493.8
O ₂	12.07	0.45	421
H ₂ O	12.62		691
NO	9.26	0.03	531.8
O ₃	12.53	2.10	625.5
CCl ₄	11.47	0.80	
CHCl ₃	11.37	0.62	650.6
CH ₂ ClCH ₂ Cl	11.07		
CHCl ₂ CH ₃	11.04		
C ₂ H ₃ Cl	9.99		
1,1-C ₂ H ₂ Cl ₂	9.81	0.1	
cis-C ₂ H ₂ Cl ₂	9.65		
trans- C ₂ H ₂ Cl ₂	9.64		
C ₂ HCl ₃	9.46	0.40	
C ₂ Cl ₄	9.32	0.64	
1,4-dichlorobenzene	8.92		

Table 2. Ionization energy (IE,eV) , electron affinity(EA, eV) and proton affinity(PA, kJ.mol⁻¹) data.

In general, the H₃O⁺ reactions with the aliphatic chloride more varies in their rate constants and products, and in some reactions H₃O⁺.M ions (M=Cl-HC) are formed. The NO⁺ reaction with the aliphatic compounds is generally slow association reactions and form NO⁺.M ions (for e.g., NO⁺ + CHClCCl₂ = NO⁺.CHClCCl₂). The O₂⁺ reactions are fast mainly proceeding via nondissociative charge transfer reactions to produce the cations M⁺ only (for e.g., C₂HCl₃ + O₂⁺ = C₂HCl₃⁺ + O₂), but in some of these reactions minority dissociative charge transfer reactions take place to eliminate Cl/HCl and leave hydrocarbon ion (for e.g., CH₂ClCH₂Cl + O₂⁺ = C₂H₃Cl⁺(95%)+HCl+O₂; and CH₂ClCH₂Cl + O₂⁺ = C₂H₄Cl⁺(5%)+Cl+O₂).

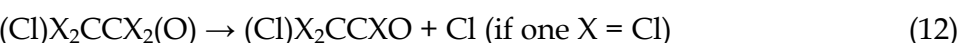
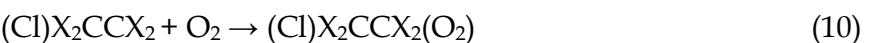
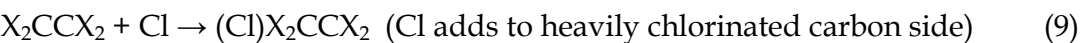
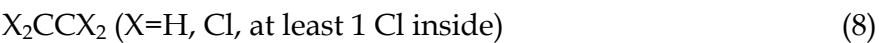
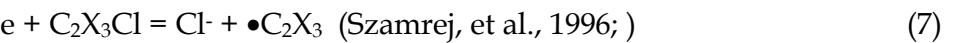
From the calculation results of dichloroethylene (Sun et al., 2001, 2007b), trichloroethylene (Nichipor et al, 2008) and tetrachloroethylene (Sun et al, 2009b), we learn that positive charge transfer reactions contributing to chlorinated ethylenes decomposition less than 10%

Secondary electron attachment, Cl dissociated reaction and negative ions charge transfer reactions

Reaction pathway of secondary electron attachment, Cl dissociative reactions followed by peroxy radical reactions is the main reaction pathway which causes Cl-HC decomposition. The rate constants of electron and Cl with Cl-HC , and the products of these reactions are listed in table 3, respectively (Atkinson, R., 1987a). For chlorinated methane, the products formed by Cl with CH₃Cl, CH₂Cl₂, CHCl₃, reactions are: CH₂Cl, CHCl₂, CCl₃ , and HCl; for chlorinated methane CCl₄, CCl₃ and Cl₂ are formed. If we assume CH₂Cl, CHCl₂, CCl₃ as radical R, generalized mechanism of peroxy radical reactions could be written as follows:



This is a main reaction pathway for Cl-HC decomposition (Bryukov et al., 2002;). For chlorinated aliphatic ethylene, the mechanism of its degradation in air mixture can be generalized as follows:



Molecule	Electron [cm ³ .s ⁻¹]	Cl [cm ³ .s ⁻¹]
CH ₃ Cl	6.1 × 10 ⁻¹¹ , Cl ⁻ + CH ₃	4.78× 10 ⁻¹³ , CH ₂ Cl+ HCl
CH ₂ Cl ₂	1.6 × 10 ⁻¹⁰ , Cl ⁻ + CH ₂ Cl	3.5× 10 ⁻¹³ , CHCl ₂ + HCl
CHCl ₃	4.9 × 10 ⁻⁹ , Cl ⁻ + CHCl ₂	1.2× 10 ⁻¹³ , CCl ₃ + HCl
CCl ₄	1 × 10 ⁻⁷ , Cl ⁻ + CCl ₃	1.4× 10 ⁻¹⁰ , CCl ₃ + Cl ₂
C ₂ H ₃ Cl	(1~8) × 10 ⁻¹⁰ , Cl ⁻ + C ₂ H ₃	1.27× 10 ⁻¹⁰ , CH ₂ ClCHCl
C ₂ H ₂ Cl ₂	1 × 10 ⁻⁹ , Cl ⁻ + C ₂ H ₂ Cl	1.4× 10 ⁻¹⁰ , CH ₂ ClCCl ₂
C ₂ HCl ₃	(0.29~1) × 10 ⁻⁸ , Cl ⁻ + C ₂ HCl ₂	9.3× 10 ⁻¹² , CCl ₃ CHCl
C ₂ Cl ₄	1 × 10 ⁻⁷ , Cl ⁻ + C ₂ Cl ₃	(4~6)× 10 ⁻¹¹ , C ₂ Cl ₅
C ₂ H ₅ Cl	(2~7) × 10 ⁻¹³ , Cl ⁻ + C ₂ H ₅	6.8 × 10 ⁻¹² , CH ₃ CHCl + HCl

Table 3. Rate constants and products for the reactions of electron, Cl with chlorinated aliphatic compounds.

The decomposition efficiency of Cl-HC mainly depends on the rate constants of secondary electron attachment, and Cl addition reaction followed by peroxy radical reactions (Knox et al., 1966, 1969; Thüner et al., 1999). This decomposition pathway has been confirmed experimentally (Hirota et al., 2002).

O₂⁻ cause Cl-HC decomposition, O₂⁻ + M = O₂ + M⁻ (M= Cl-HC) (14)

O atom, OH radical , and other radical reactions with Cl-HC

Other decomposition pathways for chlorinated aliphatic hydrocarbons are: O atom decomposition pathway (Sanhueza et al., 1974a, 1974b; Teruel, et al., 2001), OH radical decomposition pathway (Atkinson, R., 1987b; Howard, et al., 1976; Liu et al., 1989; Chandra et al., 1999; Chang et al., 1977) and other species decomposition pathway, such as O₃ and NO₃. The rate constants of O and OH with Cl-HC , and the products of these reactions are listed in table 4, respectively (<http://kinetics.nist.gov/kinetics/index.jsp>). By-products of irradiation vary with reactants.

Molecule	O [cm ³ .s ⁻¹]	OH [cm ³ .s ⁻¹]
CH ₃ Cl	1.18 × 10 ⁻¹⁶ , OH + CH ₂ Cl	4.2 × 10 ⁻¹⁴ , CH ₂ Cl+ H ₂ O
CH ₂ Cl ₂	6.48 × 10 ⁻¹⁶ , OH + <i>CHCl₂</i>	1.4 × 10 ⁻¹³ , CHCl ₂ + H ₂ O
CHCl ₃	1.02 × 10 ⁻¹⁵ , OH + CCl ₃	1.0 × 10 ⁻¹³ , CCl ₃ + H ₂ O
CCl ₄	1.89 × 10 ⁻¹⁶ , ClO + CCl ₃	< 4 × 10 ⁻¹⁶ , CCl ₃ + HOCl
C ₂ H ₃ Cl	5.96 × 10 ⁻¹³ , products	8.06 × 10 ⁻¹² , CHClCH ₂ OH
1,1-C ₂ H ₂ Cl ₂	9.8 × 10 ⁻¹³ , products	8.10 × 10 ⁻¹² , CH ₂ OHCCl ₂
C ₂ HCl ₃	1.4 × 10 ⁻¹³ , products	2.2 × 10 ⁻¹² , products
C ₂ Cl ₄	1.9 × 10 ⁻¹³ , products	1.7 × 10 ⁻¹³ , products
C ₂ H ₅ Cl	1.12×10 ⁻¹⁵ , OH+ other products	6.42×10 ⁻¹³ ,H ₂ O + other products

Table 4. Rate constants and products for the reactions of O, OH with chlorinated aliphatic compounds

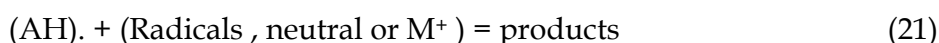
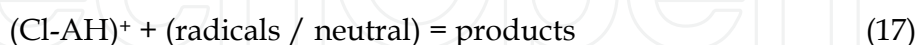
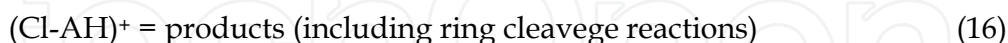
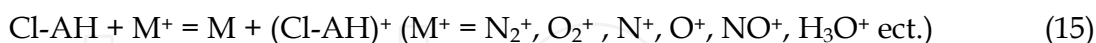
The mechanism of decomposition of chlorinated aliphatic hydrocarbons under EB irradiation could be described as follows: Cl⁻ dissociative secondary electron attachment followed by peroxy radicals reaction is a main path for Cl-HC decomposition, positive and negative charge transfer reactions with Cl-HC, O atoms and other radicals reactions with Cl-HC cause Cl-HC degradation too.

5.1.2 General mechanism of chlorinated aromatic hydrocarbons (Cl-AH) decomposition under EB-irradiation

Similar to the mechanism of chlorinated aliphatic hydrocarbons under EB-irradiation, the mechanism of chlorinated aromatic hydrocarbons go through secondary electron attachment and positive charge transfer reactions at the beginning stage of irradiation. At the late stage of irradiation, radical reactions play very important role for chlorinated aromatic hydrocarbon decomposition. Because rate constants of Cl radicals with chlorinated aromatic hydrocarbons (usually 1.0 × 10⁻¹⁵ ~ 1.0 × 10⁻¹⁶) (Shi & Bernhard, 1997) are much smaller than those of OH radicals (1.0 × 10⁻¹² ~1.0 × 10⁻¹³), Cl radical addition reaction followed by peroxy radical reaction pathway is not so important for chlorinated aromatic hydrocarbon decomposition in air mixture; on the contrary, OH radical reaction pathway is more important for chlorinated aromatic hydrocarbon decomposition in low or high humidity air mixture (Sun et al., 2007c).

Some positive charge transfer reactions, such as N_2^+ , cause benzene ring cleavage of chlorinated aromatic hydrocarbons. Aliphatic byproducts are formed. Hirota et al.(2000) and Han et al. (2003) observed some aliphatic organic compounds formed from chlorobenzene decomposition in air mixture under EB-irradiation.

The generalized chemical reactions could be written as follows:



5.2 Nonchlorinated organic compounds

For nonchlorinated aromatic organic compounds, VOCs decomposition mainly go through:

- Positive ions' charge transfer reactions:

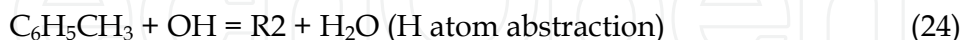
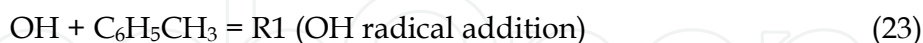


Because RH has lower ionisation energy (IE) (for eg., $IE_{\text{benzene}} = 9.24 \text{ eV}$; $IE_{\text{PAHs}} < 10 \text{ eV}$) than most primary positive ions ($IE > 11 \text{ eV}$), such as N_2^+ , O_2^+ formed from radiolysis of base gas, part of VOC will be decomposed by rapid charge transfer reactions.

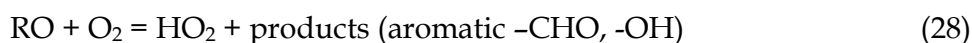
- Radical-neutral particles reactions

OH radicals play very important role for VOC decomposition, especially when water concentration is above 1%. OH radicals react with VOC in two ways:

OH radicals addition to the aromatic ring or H atom abstraction (e.g. toluene)



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



6. Conclusion

Electron beam technology to treat organic compounds has been studied for many years. Based on experiments of lab scale in batch system and flow system and experiments of pilot scale, it was shown that aliphatic organic compounds ($C \leq 4$) are easily to be decomposed by electron beam technology, the energy necessary to decompose aliphatic hydrocarbons in the order of lower to higher: chlorinated unsaturated hydrocarbons, chlorinated saturated hydrocarbons, hydrocarbons. For aromatic hydrocarbons decomposition in gas phase, energy is much higher than that used to decompose aliphatic hydrocarbons. About 70% aromatic VOCs are decomposed at 20 kGy absorbed dose for most single ring aromatic hydrocarbons.

Organic compounds in gas mixture can be decomposed by EB-irradiation, and the decomposition efficiency of organic pollutants increases with the absorbed dose. For chlorinated aliphatic hydrocarbons, the decomposition efficiency of unsaturated (with double $C=C$ bond) hydrocarbons is higher than that of saturated hydrocarbons, and the decomposition efficiency of chlorinated compounds with higher numbers of chlorine groups is higher than observed for the compounds with lower number of chlorine groups. Decomposition efficiency of chlorinated aromatic hydrocarbons is lower than chlorinated unsaturated (with double $C=C$ bonds) aliphatic hydrocarbons.

Different matrix gas and some additives influence the organic pollutants decomposition. For chlorinated aliphatic hydrocarbons, the decomposition efficiency of Cl-HC in oxygen or air is higher than that observed in nitrogen; and for chlorinated aromatic hydrocarbons (such as 1,4-DCB) the decomposition efficiency of Cl-HC in nitrogen is higher than that in air. The reason for this can be explained by their different decomposition mechanisms. Water vapor injection and NH_3 addition increase decomposition efficiency of organic pollutants.

Removal efficiency of organic pollutants in hybrid system (EB + catalyst) is higher than that in EB or catalyst system only.

Mechanism of organic pollutants decomposition is composed of following steps. At the early stage of EB irradiation, secondary electrons interact with the base gas mixture components and positive and negative charge transfer reactions play important roles for organic pollutants decomposition. At the latter stage of EB-irradiation, radical reactions play important roles for organic pollutants decomposition.

7. Acknowledgment

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Ten years after coming into force of the Stockholm Convention on Persistent Organic Pollutants (POPs), a wide range of organic chemicals (industrial formulations, plant protection products, pharmaceuticals and personal care products, etc.) still poses the highest priority environmental hazard. The broadening of knowledge of organic pollutants (OPs) environmental fate and effects, as well as the decontamination techniques, is accompanied by an increase in significance of certain pollution sources (e.g. sewage sludge and dredged sediments application, textile industry), associated with a potential generation of new dangers for humans and natural ecosystems. The present book addresses these aspects, especially in the light of Organic Pollutants risk assessment as well as the practical application of novel analytical methods and techniques for removing OPs from the environment. Providing analytical and environmental update, this contribution can be particularly valuable for engineers and environmental scientists.

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51000 Rijeka, Croatia
Phone: +385 (51) 770 447
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Phone: +86-21-62489820
Fax: +86-21-62489821

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