# The Current State and Issues of Electron Beam Flue Gas Treatment

電子ビームによる排ガス処理技術の現状とその課題

<u>Teruyuki Hakoda</u> <u>箱田照幸</u>

Japan Atomic Energy Agency, Quantum Beam Science Center 1233, Watanuki, Takasaki, Gunma 370-1292, Japan 日本原子力研究開発機構 量子ビーム応用研究センター 〒370-1292 群馬県高崎市綿貫町1233

An electron beam flue gas treatment is introduced for removal of nitrogen and sulfur oxides (NOx, SOx) in a flue gas from a coal-fired boiler, decomposition of dioxins in a flue gas from a municipal solid waste incinerator, and oxidation of volatile organic compounds (VOCs) in a flue gas from painting factories as typical application of EB technologies for environmental protection. Their current issue is also discussed for an industrial and commercial scale plant.

## 1. Introduction

Electron beams (EBs) are one of the advanced technologies for treating environmental pollutants. This technology is based on oxidation reactions with active species produced by the irradiation to media. When irradiated to air, many active species such as OH radicals, O atoms, and  $N_2^+$  are produced. Among the species, OH radicals are most important active species that can oxidize dilute pollutants in a gas stream. In the present report, the EB treatment of nitrogen and sulfur oxides (NOx, SOx), dioxins, and volatile organic compounds (VOCs) in flue gases are introduced as an application of EB technology for environmental protection. Their current issue is also discussed for an industrial and commercial scale plant.

# 2. Removal of NOx/SOx

Nitrogen and sulfur oxides (NOx and SOx) in flue gases from a coal-fired boiler will cause acid rain. When the flue gases are irradiated with EBs, NOx and SO<sub>2</sub> are simultaneously oxidized into HNO<sub>3</sub> and H<sub>2</sub>SO<sub>4</sub> in the gas phase. These acids are changed into particulate (HN<sub>4</sub>)NO<sub>3</sub> and (NH<sub>4</sub>)<sub>2</sub>SO<sub>4</sub> through the neutralization with ammonia (NH<sub>3</sub>) injected into the flue gases before EB irradiation. These particulate products are removed from the gas phase at an electrostatic precipitator (ESP) and used as an agricultural fertilizer.

Pilot-scale test was performed for a flue gas of 12,000 Nm<sup>3</sup>/h in Japan, where sufficient removal efficiency of NOx and SOx was obtained, as shown in Fig. 1. Depending on initial concentrations and gas temperature, approximately 80% of NOx and more than 94% of SOx were removed at a dose of 8.3 kGy(=kJ/kg) [1].

Based on these results, industrial-scale plants are now working at Chengdu  $(300,000 \text{ m}^3\text{N/h})$  and

Hangzhou (305,400 m<sup>3</sup>N/h) in China using accelerators (800 kV/400 mA  $\times$  2), Pomorzany Thermoelectric Power Station (270,000 m<sup>3</sup>N/h) in Poland using accelerators (700 kV/370 mA  $\times$  4) [2].



Fig. 1. Removal efficiency of NOx and SOx [NOx]₀= 220-240 ppm, [SOx]₀=650-950 ppm, Temperature= 62-67°C

## **3. Decomposition of Dioxins**

Dioxins are a very toxic to human body from the respect to endocrine disrupting character, carcinogenicity, and mutagenicity. They include polychlorinated dibenzo-p-dioxins (PCDDs) and polychlorinated dibenzofurans (PCDFs). Municipal solid waste incinerators (MSWIs) are the main source for the dioxin emission. The reduction of their emissions has been required for incineration.

Pilot-scale test was conducted in Japan to decompose/detoxify dioxins in a flue gas from the MSWI at a temperature of 200°C [3]. The test plant was constructed at a site of the Takahama Clean Center treating 450 t (150 t  $\times$  3 furnaces) of solid wastes in 1 day. The incineration of 150 t of wastes generated approximately 40,000 m<sup>3</sup> N/h flue gas. A flue gas of 1,000 m<sup>3</sup>N/h for the test plant was obtained downstream of ESP and then irradiated

with EBs using an electron accelerator (300 kV/40 mA). As shown in Fig. 2, more than 90% decomposition of PCDD/Fs was obtained at a dose of about 14 kGy. An annualized cost for EB systems was estimate to be 7% lower than that for bag-filter systems.



Fig. 2. Decomposition efficiency of dioxins

#### 4. Oxidation of VOCs

Volatile organic compounds (VOCs) have been used in various industries as solvents, washing reagents and basic materials for chemical synthesis. The emission of VOCs to the atmosphere causes toxic photochemical smog. Several efforts have been made to reduce VOC emission by installing treatment technologies, reducing the quantities used, and recycling. VOCs can be treated by two methods: they can be recovered by adsorption, absorption, and condensation, or can be eliminated by thermal incineration, catalytic incineration, and nonthermal plasma (NTP). Electron beam (EB) and electric discharge techniques are used to create NTPs that can treat VOCs with concentrations less than 100 ppmv at a flow rate of  $10^{-1}$ – $10^5$  Nm<sup>3</sup>/h [4].

Under EB irradiation, VOCs are attacked by OH radicals to produce organic acids and aldehydes as gaseous and particulate intermediate irradiation byproducts. Since these products will cause photochemical smog, they should be oxidized into COx (CO<sub>2</sub>, CO) through mineralization reactions. Some studies on the EB treatment combined with catalytic oxidation process have demonstrated the enhancement of their mineralization. We observed that an ozone decomposition catalyst was one of appropriate catalysts combined with EBs at a laboratory study [5].

A pilot-scale test for decomposition of VOCs in a gas stream (500 m<sup>3</sup>N/h) was performed using an EB/MnO<sub>2</sub> system using an electron accelerator (160 kV/50 mA) [6]. A combined MnO<sub>2</sub> bed enhanced the removal of VOCs and the mineralization of VOCs and its irradiation organic byproducts as

shown in Fig. 3. The yield of  $CO_2$  to  $CO_3$  increased from 52–60% to 83–89% by the catalytic treatment.

Based on the obtained results, the catalytic oxidation was profitable to reduce total initial installation cost for purification of VOC gas stream.



Fig. 3. Removal ratio of toluene  $(\bullet, O)$  and xylene  $(\blacktriangle, \bigtriangleup)$  and the mineralization ratio  $(\blacksquare, \Box)$  with and without the catalytic treatment as a function of dose for 5-ppmv toluene/xylene/air mixture.

#### 5. Current issue of EB treatments

An electron accelerator is a key component of EB flue gas technology. The installation of a few industrial-scale plants were cancelled, because of fatal accidents happened at high-voltage parts of used accelerators. Accordingly, high-power and stable accelerators are necessary for the application to industrial and commercial flue gas treatment.

The price of the accelerator is also important factor to realize the EB flue gas treatment. The cost of self-shielding accelerators is about 1/3 of that of house-shielding accelerators at the same power. The use of such self-shielding accelerator is one of solutions to reduce the capital cos of EB flue gas treatment.

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