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New “wet type” electron beam flue gas treatment pilot plant

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HIGHLIGHTS

- Presentation of a novel EBFGT pilot plant.
- Very efficient design incorporating an electron beam.
- “Wet type” flue gas scrubber.
- An “All-in-one type” treatment system.

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ABSTRACT

We describe a new pilot plant for flue gas cleaning by a high energy electron beam. The special feature of this pilot plant is a uniquely designed reactor called VGS® (VIVIRAD Gas Scrubber, patent pending), that allows oxidation/reduction treating flue gas in a single step. The VGS® process combines a scrubber and an advanced oxidation/reduction process with the objective of optimizing efficiency and treatment costs of flue gas purification by electron accelerators. Promising treatment efficiency was achieved for SO_x and NO_x removal in early tests (99.2% and 80.9% respectively). The effects of various operational parameters on treatment performance and by-product content were investigated during this study.

1. Introduction

Fossil fuels like coal or heavy fuel oil are commonly used for producing energy in thermal power plants, refineries and industrial plants. The combustion of such fuels results in massive emissions of sulfur oxides ($SO_x = SO_3 + SO_2$) and nitrogen oxides ($NO_x = NO + NO_2$) that produce significant air pollution responsible for acidic rain and contribute to the greenhouse effect (Turhan et al., 2001).

Different conventional and widely commercialized processes are used for removing these pollutants from flue gas. There are two types of processes: desulfurization and denitrification. Concerning desulfurization, current methods can be subdivided into wet, semi-dry and dry processes. Similarly, denitrification systems can be classified as selective catalytic reduction and selective non-catalytic reduction (Sonde, 2011). Desulfurization and denitrification of flue gas involve therefore the use of two different processes which induce higher treatment costs, a large space requirement and a large workforce. In addition, most of these processes generate other pollution like acidic wastewater, gypsum, carbon dioxide and exhausted catalyst. In this context, plasma technology represents a promising alternate solution for flue gas treatment.

Electron Beam Flue Gas Treatment (EBFGT) consists of introducing a high energy electron beam into a flue gas in the presence of ammonia and water in order to convert pollutants into a potentially valuable by-product. During irradiation, high energy electrons interact with the major flue gas components (N_2, O_2, H_2O and CO_2) creating various ions and radicals such as $O^•$, $OH^•$, $N^•$ and $HO_2^•$ (Chmielewski, 2007). These active species rapidly react with SO_x and NO_x to form sulfuric acid (H_2SO_4) and nitric acid (HNO_3) respectively. A stoichiometric input of ammonia converts these acids into a mixture of ammonium sulfate $(NH_4)_2SO_4$ and ammonium nitrate (NH_4NO_3) that can be used as a soil fertilizer.

Concerning NO_x removal, even if a part of NO is reduced to gaseous nitrogen, this radiation-induced process can be

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summarized as follows (Chmielewski et al., 2004):

$$NO \xrightarrow{O,O_3,HO_2} NO_2 \xrightarrow{OH^\bullet} HNO_3 \xrightarrow{NH_3} NH_4NO_3 \quad (1)$$

Regarding SO_2 and SO_3 removals, the two major degradation pathways are thermal (2) and radio-thermal (3) mechanisms and they each lead to the formation of ammonium sulfate:

$$SO_2 \xrightarrow{NH_3,H_2O} (NH_4)_2SO_3/(NH_4)HSO_3 \xrightarrow{O_2} (NH_4)_2SO_4 \quad (2)$$

$$SO_2 \xrightarrow{OH^\bullet} HSO_3 \xrightarrow{O_2,OH^\bullet} SO_3 \xrightarrow{H_2O} H_2SO_4 \xrightarrow{NH_3} (NH_4)_2SO_4 \quad (3)$$

Since the first EBFGT experiments in Japan in the early 1970s, pilot plants and industrial EBFGT units were usually operated with dry or semi-dry processes and were generally multistaged as follows:

(i) Flue gas conditioning (pre-heater, electrostatic precipitation, etc.).
(ii) Input of ammonia to a mixing chamber.
(iii) Electron beam treatment in a reactor.
(iv) Recovery of processing by-product (electrofilter).

Many scientific studies and pilot or industrial operating units have demonstrated EBFGT performances: in Chengdu (China) (Chmielewski et al., 2004), Pomorzany (Poland) (Chmielewski, 2007), Nagoya (Japan) (Namba et al., 1995), Sviloza (Bulgaria) (Kim et al., 2011), among others. These studies have usually pointed out the main advantages of this technology as follows:

- Simultaneous denitrification and desulfurization.
- Removal of pollutants like volatile organic compounds, dioxins, HX (where X=F, Cl, Br, I), Hg, etc.
- Useful by-product (fertilizer).
- Reliable process.
- Simple operating mode.
- Lower capital and operating costs than conventional technologies.

The good performance reported by some of these facilities has attracted VIVIRAD S.A. into reconsidering the EBFGT process in order to make it more compact, increase its efficiency and reduce fouling and corrosion associated with dry by-product generation.

Since the formation of reactive species such as hydroxyl radicals or aqueous electrons during electron beam treatment relies on the presence of water molecules in the treatment zone, researchers have shown that humidity improves EBFGT efficiency for pollutant removal (Basfar et al., 2010; Genuario and de Kat, 2009; Calinescu et al., 2013; Ostapczuk et al., 1999). The idea behind the present development is therefore to introduce an electron beam and a suitably large amount of liquid water into a single reaction chamber. This increases the electron energy deposited in the liquid phase compared to that deposited in the gaseous phase of a wet but unsaturated gas stream at atmospheric pressure.

This gas processing concept named VGS® (VIVIRAD Gas Scrubber) was implemented in 2011 at the Sarayköy Nuclear Research and Training Center (SANAEM) of the Turkish Atomic Energy Authority (TAEK) with the collaboration of the Turkish Petroleum Refineries Corporation (TÜPRAŞ).

This paper presents the TAEK-TÜPRAŞ EBFGT pilot plant fitted with VGS® and early experimental results conducted on the treatment of flue gas generated by heavy fuel oil combustion.

2. Experimental

2.1. Pilot plant

The TAEK-TÜPRAŞ EBFGT pilot plant consists of a 10 kW ICT type self-shielded electron accelerator (500 keV and 20 mA) fitted with a 50 × 5 cm^2 extraction window connected to a stainless steel reaction chamber (VGS® reactor) (Fig. 1). In its upper part, the VGS® receives flue gas from a burner (5861520 kJ/h) that is able to provide up to 2000 normal cubic meters per hour (Nm^3/h) of flue gas at a temperature ranging from 80 to 140 °C. Due to space limitations, the burner and its related facility are located outside the electron beam building.

Electron beam treatment of flue gas using VGS® is a wet type "all in one" process as it brings together the gas stream, the liquid inputs (water and ammonia), the electron beam and the by-product recovery in a single device.

The VGS® reactor is composed of three parts (Figs. 2 and 3)

- Flue gas stream and electron beam are injected into the upper part of the reactor.
- In the central part of the reactor, flue gas is simultaneously exposed to a dense fog of water, ammonia and by-product solution and to high energy electrons. The dense fog is produced by spray nozzles located throughout the interior of the reactor. Most reactions with radicals and the neutralization by ammonia take place in this central volume.
- Liquid and gas phases are separated by gravity in the lower part of the reactor

(1) Treated flue gas is exhausted via peripheral ducts to the stack. Each flue gas exhaust pipe is equipped with water curtains to prevent any ammonia loss.
(2) By-product solution is recovered in a pool at the bottom of the reactor where neutralization reactions can still occur. Concentrated by-product solution is extracted from the VGS® at the bottom of this tank.

This pilot plant is fully controlled via an on-line computer linked to gas analyzers located upstream and downstream of the VGS® for measurement of pollutant concentrations (NO_2 and NO for NO_x analysis, SO_2 for SO_x measurement since SO_2 is the main

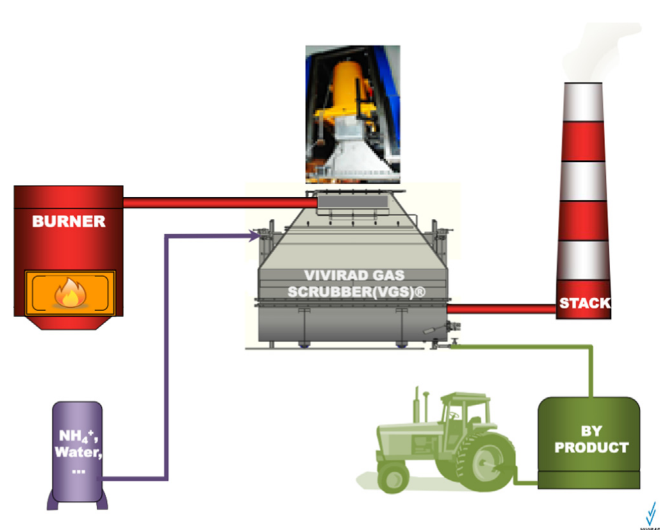

Fig. 1. Schematic view of TAEK-TÜPRAŞ EBFGT Pilot Plant.