

THE REDUCTION OF GAS PHASE AIR TOXICS FROM COMBUSTION AND INCINERATION SOURCES USING THE GE-MITSUI-BF ACTIVATED COKE PROCESS

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Introduction

This paper provides information on the GE-Mitsui-BF dry DeSO_x/DeNO_x/Air Toxics removal process which can be utilized by utility, chemical, oil refineries and other industries. This process, originally researched and developed during the 1960's by Bergbau-Forschung (BF), now called Deutsche Montan Technologies, was licensed to Mitsui Mining Company (MMC) in 1982 to investigate, test and adapt the system to Japanese regulations which are more stringent than those in the United States. General Electric Environmental Services, Inc. (GEESI) has licensed the Mitsui-BF process for flue gas cleaning applications in North America. The process is installed on four coal-fired boilers and fluidized catalytic cracker (FCC) units. These units were constructed by MMC in Japan and Uhde GmbH in Germany. MMC has also developed a technology to produce the Activated Coke (AC) used in the removal process based on their own metallurgical coke manufacturing technology.

Research & Development of AC

AC is a formed, carbonaceous material designed for a dry DeSO_x/DeNO_x/Air Toxics process used for flue gas cleaning. AC has a high mechanical strength against attrition and crushing during the circulation and handling process.

Laboratory tests of the AC have been conducted by MMC since 1980, including selection of raw materials, methods of producing high mechanical strength pellets, procedures for carbonization, activation and chemical treatment methods to enhance adsorptive effectiveness.

In October 1996, a 3,000 ton/year capacity plant for AC production began operation in MMC's Kitakyushu works.

Characteristics of MMC's activated coke vs. activated carbon are shown below.

Items	MMC's	
	Activated Coke	Activated Carbon
BET Surface Area (m ² /g)	150-250	850
Mechanical Strength (%)	95	85
SO ₂ Adsorption Capacity (Mg-SO ₂ /g)	60-120	220*170**
NO _x Removal Efficiency (%)	80-85	60-70
Price Ratio (-)	1/4 - 1/3	1
* with fresh material		
** with used material		

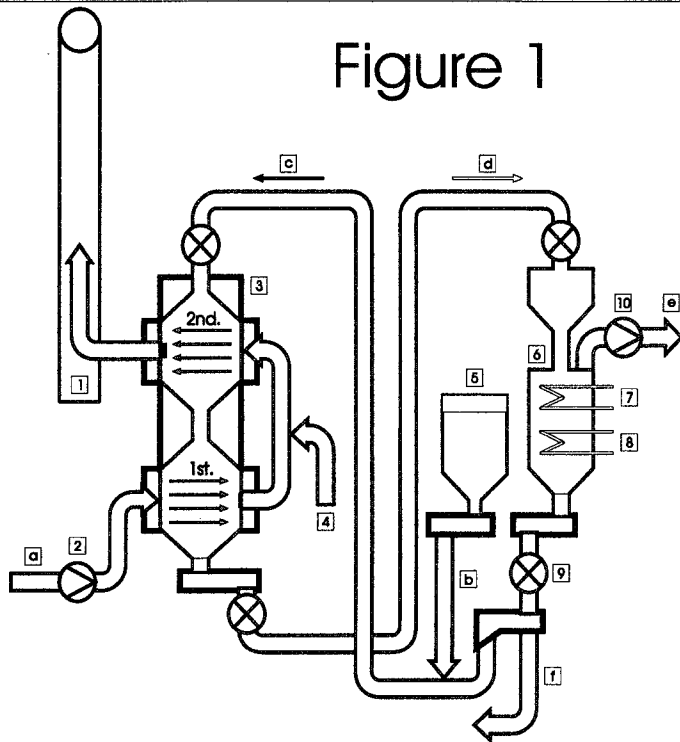
GE-Mitsui-BF Dry Simultaneous DeSO_x/DeNO_x/Air Toxics Removal Process

MMC's activated coke is utilized in a process which accomplishes desulfurization, denitrification and air toxics removal simultaneously in a single system consisting of three sections: adsorption, AC regeneration and byproduct recovery. A schematic of the process is provided in Figure 1.

ADSORPTION: The adsorption section consists of two stages. AC moves continuously from top to bottom through the adsorber. First AC enters in the top of the second stage, where NO_x reduction with the addition of NH₃ occurs. The discharged AC from the bottom of the second stage enters the top of the first stage, where the majority of the SO_x and Air Toxics adsorption occurs. The SO_x/Air Toxics-filled AC is discharged from the bottom of the first stage and sent to the regeneration stage. If only DeSO_x or DeNO_x along with Air Toxics removal separately is required, a single stage process can be designed.

AC REGENERATION: AC discharged from the bottom of the first stage is sent to a two-stage regeneration vessel where sulfuric acid, its ammonia salts and air toxics are thermally decomposed into an SO₂ concentrated gas.

Figure 1



1. Stack
2. Flue Gas Booster Fan
3. Adsorber
4. NH₃ Injection
5. Activated Coke Bin
6. Desorber
7. Regeneration Heater
8. Regeneration Cooler
9. Vibrating Screen
10. Blower

- a. Flue Gas
- b. Activated Coke (granular)
- c. Activated Coke (regenerated)
- d. Activated Coke (saturated)
- e. SO₂-rich Gas
- f. Fines (return to boiler)

During the first stage, the AC is indirectly heated by an external furnace's combustion air. This SO₂-rich gas (SRG) is sent to the byproduct section. During the second stage, the cooled, regenerated AC is filtered to remove fine dusts, then recycled back to the adsorber. The AC fines can be recycled and disposed of by burning as fuel.

BYPRODUCT RECOVERY: SRG generated in the AC regeneration section contains approximately 20-25% SO₂. It can be converted into either salable elemental sulfur (purity >99%) either liquid or solid, sulfuric acid (purity >98%) or liquid SO₂.

Process Applications

In 1987, a commercial plant started operation at Idemitsu Kosan, Aichi Oil Refinery which treats flue gas from the catalyst regeneration section of a Residue Fluid Catalytic Cracking Unit (RFCC) (236,000 Nm³/hr design). Performance of this DeSO_x/DeNO_x plant has been very successful. A removal efficiency of 100% for SO_x and over 80% for NO_x has been constantly achieved at operating temperature of 180 C.

Additional applications include two power plants in Germany, one burning lignite and one burning hard coal.

Conclusion

The GE-Mitsui-BF dry, low temperature DeSO_x/DeNO_x/Air Toxics removal system has been proven capable of removing over 99% of the SO_x, including SO₃, up to 99% of selected air toxics and over 80% of the NO_x in coal-fired and fluidized bed boilers and RFCC units. In addition, the process is effective in reducing vaporous, elemental mercury by 99%+ and dioxins/furans by 70% to 98% from combustion flue gases.

References:

1. K. Knoblauch, E. Richter, and H. Juntgen. "Application of Active Coke in Processes of SO_x and NO_x Removal from Flue Gases", Fuel, September 1981, Vol. 60, p. 832.
2. Y. Komatsubara, M. Yano, I. Shiraishi and S. Ida, "Preparation of Active Coke for the Removal of SO_x and NO_x in the Flue Gases", Proceedings of 16th Biennial Conference on Carbon, 1983, p. 235
3. I. Mochida, M. Ogaki, H. Fujitsu, Y. Komatsubara and S. Ida, "Catalytic Activity of Coke Activated with Sulfuric Acid for the Reduction of Nitric Oxide", Fuel, July 1983, Vol. 62, p. 867.
4. S.M. Dalton, "Current Status of Dry NO_x-SO_x Emission Control Processes", Proceedings of the 1982 Joint Symposium on Stationary Combustion NO_x Control, 1982, p. 32-1.