Development of the PPCP Technology in IEPE

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Abstract: The development of the PPCP technology in the IEPE has been introduced in this paper, including process technologies, pulsed powers, plasma reactor, activated vapor and ammonia, additional catalyzer, by-product catcher, match between the pulsed power and the reactor, and so on. According the experimental results, the feasibilities of the industrial application has been proved and there is a plan on building an industrial demonstration set.

Keywords: PPCP, $DeSO_2$ and $DeNO_x$, Pulsed power

1 INTRODUCTION

In 1980 s, a Japanese scholar named Masuda proposed that pulsed powers replaced the E-beam accelerators for flue gases treatment. [1] Due to the advantages of this PPCP (Pulsed corona induced Plasma Chemical Process) technology, for example, low cost, simple process line, simultaneously scrubbing SO_2 and NO_x and other poisonous substances, and so on, more and more attentions have been drawn to study on the advanced technology. There are a lot of experts in China, Korea, Russia, Canada, Japan, Italy, Poland, etc. to research on it and more inspiring results have been achieved. Now, several middle scale facilities were built in China, Korea, etc, to prove the feasibilities of industrial application. [2-8]

For last decades, supported by ministry of science and technology, China, Institute of Environmental protection Engineering (IEPE) and Dalian Science and Technology University have focused on this subject. A middle scale industrial set with 50,000 Nm^3/h flue gas has been built at the thermal plant in Sichuan Science City. With the operation of this set, the key technologies and equipments have been grasped.

There is a plan on building an industrial scale set with over 300,000 Nm^3/h flue gases flow in a 100 MW power plant.

2 PROCESS TECHNOLOGIES

In 1998, a pilot plant with 1,000 Nm^3/h–3,000 Nm^3/h was built in Dalian Science and Technology University [8]. In this pilot plant, two types of pulsed powers were tested. One was based on the spark-gap switch, other was based on the magnetic pulsed compressor. These pulsed powers will be introduced in the 3rd section.

According these results from the above-mentioned pilot plant, a middle scale industrial set with maximum 20,000 Nm^3/h flue gases flow was built at the thermal plant in Sichuan Science City. A 50 kW–100 kW pulsed power based on the magnetic compressor had been tested on this set, meanwhile, the influential factors such as temperature, humidity, residual time, ratio of the NH_3 and SO_2, energy exhausted, concentration of the inlet SO_2 had been tested [6]. The process line is shown as Fig. 1. The flue gases came from a boiler with an ash catcher, then got into a cooling tower in which the water showered from the top. The flue gases with 60 ℃–80 ℃ in temperature and 8%–13% (%Vol) in humidity went to the entrance of the plasma reactor where the NH_3 was sprayed into the flue gases, then the NH_3 reacted with SO_2 and NO_x in the flue gases in the reactor, where the plasma was induced by the pulsed power, meanwhile, the solid by-products such as $(NH_4)_2SO_4$ and NH_4NO_3 became, finally, the by-products in flue gases were caught by a catcher behind the plasma reactor, and the cleaned flue gases went into the air through a stack. The scene of the middle scale industrial set is shown as Fig. 2.

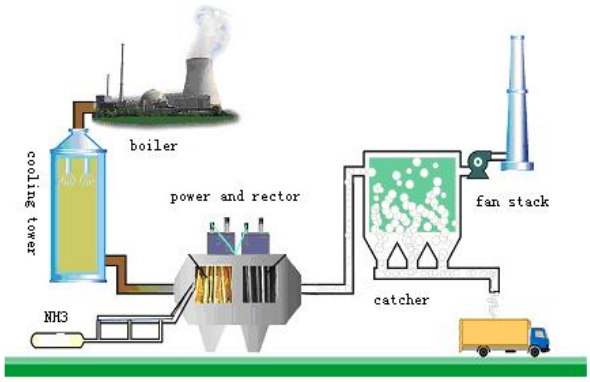


Fig. 1 The diagram of the process line of PPCP

Fig.2 The scene of the middle scale industrial set

In 2004, the middle scale industrial set had been modified to meet 50,000 Nm^3/h flue gases flow. The technologies of activated vapor and ammonia and additional catalyzer were applied in this set.

A 200 kW pulsed power based on BPFN (Blumlein Pulse Forming Network) was test on it, and the continuous operation was conducted.

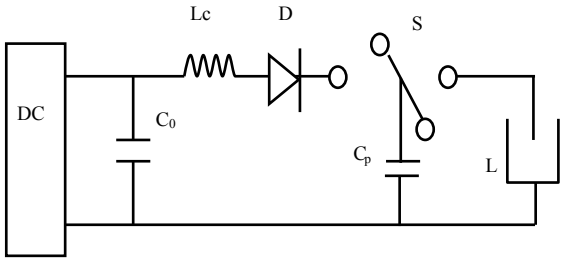
On the conditions of 40,000 Nm^3/h flue gases flow, temperature in 75℃–80℃, humidity in 8 %–10 % (%Vol), residual time in 4 s–5 s, mol ratio of the NH_3 and SO_2 in 1.8:1–1.85:1, energy exhausted in 3 Wh/Nm^3–4 Wh/Nm^3, the results were achieved as follows: The removal efficiencies of the SO_2 and NO_x were over 90% and 40% respectively without additional catalyzer.

3 KEY EQUIPMENTS

3.1 Pulsed Powers

The pulsed power is one of the key equipments of the PPCP technology. It is very difficult to develop of these pulsed powers because there are several requirements of the PPCP technology as follows: high average power, high energy efficiency, continuous operation for long time, narrow pulse width and fast rise time and so on. It is worst that the pulsed powers must match to the reactors whose impedance is capacitive and variable.

For meeting these requirements, four types of pulsed powers had been developed. The first one was based on a spark-gap switch, which average power was about 15 kW. The schematic diagram of the circuit is shown as Fig.3.

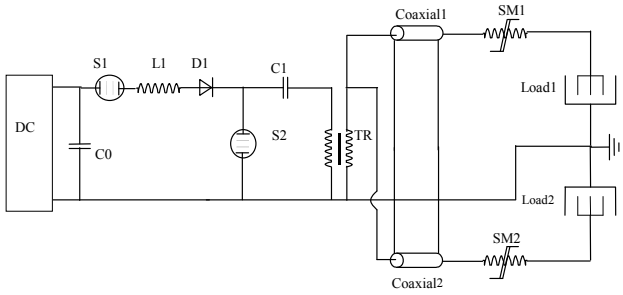


DC- direct power C_0-energy storage capacitor Lc-inductance D-diode stack S-rotary spark-gap switch Cp-pulesd capacitor L-reactor Load

Fig. 3 diagram of pulsed power based on the rotary spark-gap switch

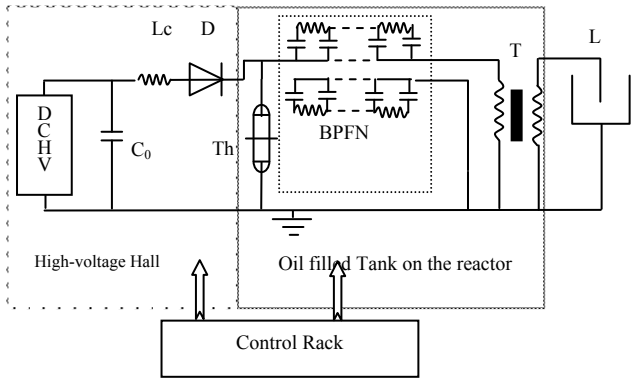
Because of the bad performance of the spark-gap switch for continuous operation, the second type pulsed power had been developed, which was based on the magnetic pulsed compressor. There were two sets, one was 10 kW and other was 50 kW–100 kW. The schematic diagram of the circuit is shown as Fig.4. They had been tested on the pilot plants with 1,000 Nm^3/h–3,000 Nm^3/h flue gases flow and 20,000 Nm^3/h flue gases flow respectively.

In order to increase average power of one set, a 200 kW pulsed power had been developed, which was based on BPFN. The schematic diagram of the circuit is shown as Fig. 5. This power had been tested on the middle scale industrial set with 40,000 Nm^3/h–50,000 Nm^3/h flue gases flow. The continuous operation was over 2000 h. The main characteristics of this pulsed power were as follow: 120 kV peak voltage, 3 kA peak current, 400 Hz–600 Hz repeating frequency, 300 ns rise time, 800 ns pulse width and the maximum average power up to 150 kW. The equipments and the output waveforms are shown as Figs .6 and 7 respectively.



DC-power supply; C0-energy storage capacitor; S(1,2)-thyratrons Lc-inductance; D1-diode stack; C1-pulsed capacitor TR-pulsed transformer; Coaxial(1,2)-coaxial cables SM(1,2)-magnetic pulsed compressors Load(1,2)-reactor loads

Fig. 4 Diagram of the pulsed power based on magnetic pulsed compressors



DCHV□high-voltage DC; C_0□storage capacitors; Lc□charge inductor; D□high-voltage diode stacks; Th□thyratron; BPFN—Blumlein pulse forming network; T□Pulse transformer; L□Load of the reactor

Fig. 5 The schematic diagram of the pulsed power

Fig. 6 The equipments in the high voltage hall

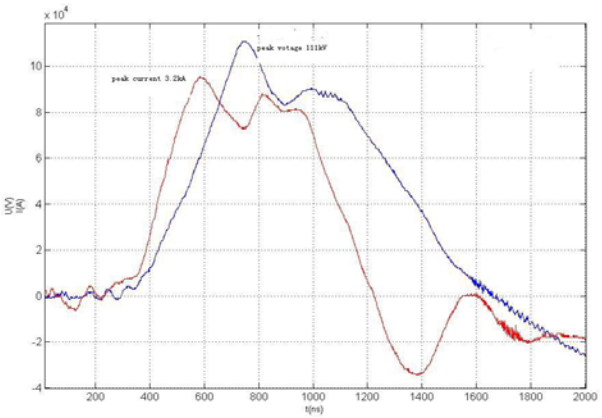


Fig. 7 The voltage (1) and current (2) waveforms of the pulsed power

To meet the requirements of the high efficiency and long lifetime, a pulsed power based on the semiconductor switches and magnetic pulsed compressors is being developed. In this subject, the connection in series of the semiconductors, which is controlled by a special photoelectric control system that can be adjusted to suit for the dispersing characteristics of the semiconductors, is considered. Other high voltage magnetic compressor and a saturable pulsed transformer are used in system too.

At first step, the pulsed power is set to be as follows: 10 kW–30 kW average power, 100 kV peak voltage, 4 kA peak current, 250 ns pulse width, 100 Hz–300 Hz repeating frequency. A 200 kW pulsed power based on this method has been proposed.

3.2 Plasma Reactors

There are several types of plasma reactors such as wire-cylinder reactor, wire-plate reactor, dielectric barrier discharge reactor, and so on. In the middle scale industrial set, there were three parts in the plasma reactor. First part was the activated vapor and ammonia set, which detail will be introduced in the next section. The second part was the pulsed electric field to induce plasma and caused many the energetic electrons and the radicals such as OH, O, HO_2 etc. to oxide the poisonous substances. The wire-plate structure was employed, which is always applied in the electrostatic precipitators. The distance between the plates was about 260 mm and the distance between the wires was about 100 mm. There were 10 channels and it was 3m in height and 3 m in length. The 200 kW pulsed power provided energy into reactor. The static capacitor of the reactor was about 5.6 nF and the impedance of the reactor was about 30 Ω–40 Ω at the peak voltage. But the impedance was various due to the status of the flue gases changes. When the humidity was 8%–10% (vol %), it was helpful to insert energy into the reactor, if the humidity was over 14 % (vol %), the sparks took place frequently and it was difficult to insert energy into the reactor because that the vapor absorbed the electrons prevented the electron avalanches taking place. The third part of the reactor was a pre-collector for the by-products to research on the integration of the reactor and the collector. On the condition of the 12,000 Nm^3/h flue gases, the collection efficiency was up to 85% by the pre-collector.

3.3 By-product Catcher

In the process, an electrostatic precipitator with wire-plate electrodes used for the by-product catcher, in which there were electric fields with 10 m^2 in their sections, located behind the plasma reactor. As for the by-products of the $(NH_4)_2SO_4$ and NH_4NO_3, they were very small powder with less 1 μm in diameter and they were very hygroscopic.

If the humidity was too high, the powder would cling to the electrodes and it couldn't be separated from the electrodes, then the wires became bigger and the corona disappeared there, so the powder couldn't be caught well. In this case, the steel brushes were set to separate the powder from electrodes. Otherwise, if the humidity was too low, the powder flew with the gases flow when the cleanness took place there. So a suitable humidity was very important for by-product catcher.

Through operation of the middle scale industrial set, that humidity is 8%–13% is OK for the by-product catcher. The photograph of the by-product is shown as Fig.8.

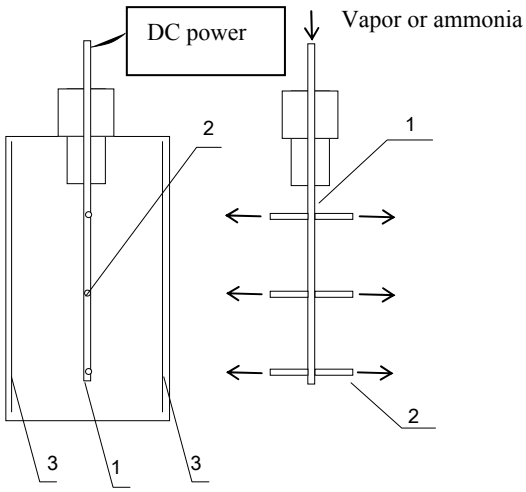
Fig. 8 The photograph of the by-product

4 DEVELOPING PROCESS TECHNOLOGIES

4.1 Activated Vapor and Ammonia

The geometry of the activated vapor and ammonia set is shown Fig. 9. A direct current power provided energy to the point electrodes. At the tips of the electrodes, intensity of the electric field was very strong. So a lot of energetic electrons and free radicals appeared there, when the gases such as vapor and ammonia passed the hole in the electrodes.[11]

The energy was focused on the reactant such as vapor and ammonia, so the efficiency of energy was better than that of energy inserted into flue gases directly. In a small pilot plant, this technology increases 10% removal efficiency of the SO_2. In the middle scale industrial set, the 20%–30% of SO_2 could be removed by the first part of the activated vapor and ammonia set, meanwhile, some of by-product could be collected by the DC electric field.



1. Main pipe 2. Pin electrodes with hole 3. Grounded electrodes

Fig. 9 The diagrammatic sketch of pipe-nozzle electrode

4.2 Additional Catalyzers

To increase the removal efficiency of the NO_x and decrease the energy exhausted, there are many experts to study on additional catalyzers such as CH_4, $C2H_4$, C_3H_6. Results of the research have been grasped, it is very valid that a little of additional catalyzer is added into flue gases. When the concentration of the C_3H_6 in the flue gases is a few tens ppm, the removal efficiency of NO_x is up to 70 % on the condition of the inserted energy less 2.0 Wh/Nm^3[7]. The formulas are shown as follows:

$$C_2H_4.C_3H_6+O\rightarrow R^*+RCO^*$$
$$C_2H_4.C_3H_6+HO^*\rightarrow R^*$$
$$C_2H_4.C_3H_6+HO^*\rightarrow RO^*+RCO^*+\text{ aldehyde}$$
$$2NO_2+2NH_3\rightarrow N_2+H_2O+NH_4NO_3$$

To grasp this technology, a small pilot plant with 200 Nm^3/h flue gases flow had been built in the Dalian Science and Technology University. Through operation this pilot plant, the inspiring results have been gained. Then the experiment was conducted in the middle industrial scale set. The maximum removal efficiencies of NO and NO_x were up to 75% and 55% respectively when the inserted energy was about 2.0 Wh/Nm^3 with 30 ppm additional catalyzer. The curve is shown as Fig. 10.

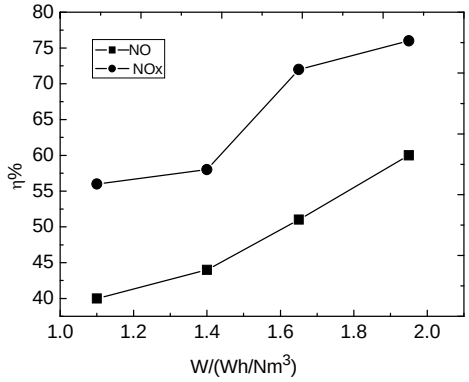


Fig. 10 The removal efficiency of the NO and NO_x dependent the inserted energy

The additional catalyzer perhaps is pollution, so further study must be conducted before the industrial application.

5 OTHER EQUIPMENTS

In the process line, there were other equipments such as the cooling tower, the additional ammonia station, the additional SO_2 station, the fan, analyzers in the line and so on.

For testing the influence of the variable SO_2 concentration at inlet of the entry of the reactor in wide range, so the additional SO_2 station was built in the process line. The fan was driven by a frequency converter to changes the flue gases flow easily. The analyzers could monitor the concentration of the SO_2 and NO_x at the both of the inlet and outlet of the reactor at same time.

The data in the process were collected by the computer in the control room, including the flue gases flow, concentration of the SO_2 and NO_x, and so on. And amount of the both of the NH_3 and SO_2 added into flue gases were controlled by the computer too. So the data was recoded in the computer clearly and correctly.

6 MATCH BETWEEN PULSED POWER AND REACTOR

The match between the pulsed power and the reactor is very important in this technology, So there are a lot of experts such as Y. Zhu, K. Yan, and so on [9-10, 12]. In some small-scale pilot, experts can match between pulsed power and the reactor well according to their experience, but in the industrial application, it is high cost and complicated work to adjust the pulsed power or the reactor, so we need a lot of knowledge to instruct engineers in the design of the industrial set.

According to a small scale pilot plant made by the IEPE and the Zhejiang University, the results of experiment are shown as follows: The relation of the η(energy efficiency) and the EP (peak intensity of the electric field) was approximately linearity. The curve is shown as Fig. 11.

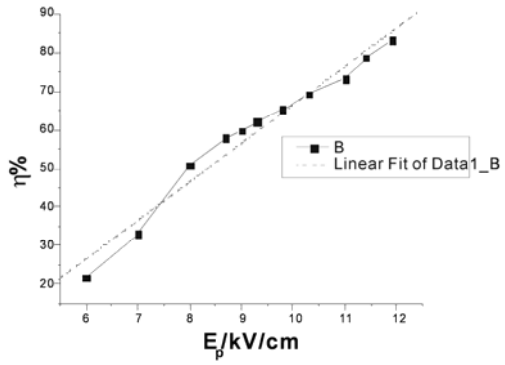


Fig. 11 Relation of η and E_p

In the middle industrial scale set, the network of the resistors and inductors was applied to match the pulsed power to the reactor. At the pulse voltage's rise time, the network couldn't exhaust the energy, so the pulse voltage could rise without the influence of the network. But at the tail of the pulse voltage, the network can exhaust some of the energy in the reactor, so the tail of the pulse voltage falls rapidly. The pulse width could be shortened to avoid sparks in the reactor by this method, so the peak voltage can be rise higher to get higher energy efficiency. In the middle scale industrial set, the maximum energy efficiency was up to 70% under good conditions of the flue gases.

7 FURTHER STUDY AND SUGGESTIONS

(1) The long-life, stable and high average power pulsed power should be further studied to meet the requirements of the industrial application completely.

(2) To decrease the exhausted energy and increase average power of the pulsed power, the matching between pulsed powers and the reactors should be conducted to build a systemic method.

(3) The industrial application set in an over power 100 MW plant should be conducted for the illustration of the PPCP technology.

(4) At the start of the market of $DeSO_2$ and $DeNO_x$ in China, the PPCP technology perhaps can become a competitive technology in the next decade. So the economic feasibilities should be study deeply.

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