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Research Article

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Experimental study of urea on SNCR removal of NO_X

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ABSTRACT

Nitrogen oxide is one of the main pollutants in the atmosphere, so it is very important to study the technology of nitrogen oxide emissions. In this paper, urea being used as reducing agent, the denitration effect and law of various experimental conditions on SNCR technology were studied , by changing the reaction temperature, NSR, O_2 concentration and the other conditions,. Experimental results showed that the reaction temperature, NSR and O_2 concentration had a great influence of urea on SNCR removal of NO_X; the optimum denitration temperature of SNCR was about 925℃, at the same time the maximum efficiency was 81%; the optimal temperature window of reduction was 875-1025℃, within this interval efficiency over 50%; the optimal NSR was 1.5; denitration efficiency achieved the maximum value(83%) at 1100℃ in anaerobic conditions; in aerobic conditions, the maximum efficiency was achieved at about 925℃; the optimum O_2 concentration was between 1% and 4%.

Keywords: Nitrogen oxide, urea, SNCR, denitration efficiency.

INTRODUCTION

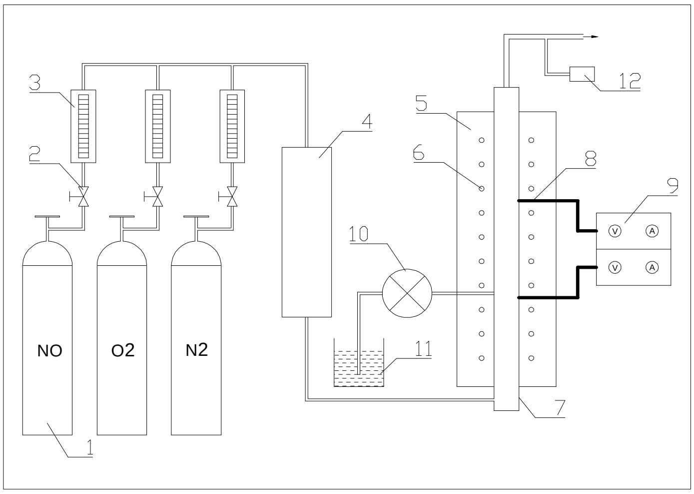
Nitrogen oxide (NO_X) is a compound composed of nitrogen, oxygen, with the common nitrogen oxides NO, NO_2, N_2O, N_2O_5, etc. NO_X in the air is often NO and NO_2, of which NO accounts for 90% or above . NO_X, one of the main pollutants in the atmosphere, could lead to acid rain, photochemical smog, haze and a series of urban environmental problems, and has a huge hazard on human health and ecological environment[1,2]. Therefore, it is very important to study the NO_X removal technology.

Selective non-catalytic reduction (SNCR) technology is a NO_X removal technology, which injects reducing agent with amino-group (ammonia, urea, ammonium bicarbonate) into a suitable temperature range (850-1050℃), and reverts NO_X in flue gas to N_2 and H_2O without catalyst[3，4]. Because urea is a stable and nontoxic solid, easy to store and transport, NO_XOUT process[5] for using urea (NH_2CONH_2) as nitrogen reducing agent is widely applied and studied. Research by scholars of SNCR technology showed that denitration effect on SNCR was related to many factors, such as reaction temperature, ammonia and nitrogen ratio(NSR), oxygen concentration and additive[6]. In this paper, urea being as a reducing agent, by changing the experimental conditions (reaction temperature, NSR, O_2 concentration), this article took a research on the impact and rule of various experimental conditions on SNCR denitration effect in NO_XOUT process, and found experimental conditions and parameters, which made the SNCR denitration effects optimum.

EXPERIMENTAL SECTION

Experimental equipment

Experiments were carried out on a self-made vertical experimental station. The experimental system was composed of gas distribution section, electric furnace reactor, reducing agent injection device, flue gas analyzer and other components. The experimental apparatus was shown in Fig. 1.



1- Gas collecting bottle; 2- Flow control valve; 3- Glass rotor flowmeter; 4- Mixer; 5- Electric heating furnace; 6- Silicon carbide; 7- Corundum tube; 8- Thermo-couple; 9- Temperature controller; 10- Peristaltic pump; 11- Urea solution; 12- Flue gas analyzer
Fig. 1 Experimental system schematic

The reactor, vertically arranged corundum tube (∅60mm×700mm), was arranged in an electric heating furnace. Electric heating furnace with silicon carbide as a heating element, through temperature controller and thermocouple formed temperature feedback system, which could effectively control and measure the furnace temperature, with the control accuracy of ±5℃ and the highest temperature of1350℃. Three kinds of gases, NO, O_2, N_2, through flow control valve and glass rotor flowmeter, got into mixer with certain proportion and formed a simulated flue gas, which replaced NO_X. Simulated flue gas entered corundum tube reactor, then reacted with urea for injection into furnace under certain conditions. The urea solution injection into position from the corundum tube bottom was 210mm. Flue gas was measured by the KM9106 portable flue gas analyzer of British KANE company and America IST series IQ-1000 multifunction gas detector.

Experimental method

Experiments used urea as reducing agent, with mass fraction of urea solution 2% and reaction temperature range from750℃ to1150℃. Following the initial experimental conditions, the total flow of simulated flue gas was constant 2L/min (standard condition), the initial NO concentration was 300 μ L/L, NSR was 1.5, the O_2 concentration was 2%, and N_2 was balance gas. In this paper, the effects of temperature, NSR and O_2 concentration on denitration efficiency and ammonia leakage of SNCR were studied.

RESULTS AND DISCUSSION

$$\text{denitration efficiency } \eta = \frac{\varphi_{(NO,in)} - \varphi_{(NO,out)}}{\varphi_{(NO,in)}} \times 100\%$$

Effect of temperature on SNCR

Because the temperature had a great influence on NO_X reduction, the effects of temperature on denitration efficiency, ammonia leakage and N_2O concentration were focused on.

As was shown in figures 2,3 and 4, the effects of temperature on denitration efficiency, ammonia leakage and N_2O concentration in NO_X removal of urea were obvious. From Fig.2, under different NSR conditions, $DeNO_X$ efficiency had an optimal temperature, about 925 ℃. When temperature was below 800 ℃, the concentration of OH, H, O active radicals was low to inhibit reduction of NO, so that denitration reaction was difficult to carry out and efficiency was less than 10%.When temperature was gradually increased, reaction rate of denitration increased, which caused reaction of urea reduction NO dominant and removal efficiency increasing rapidly. When temperature was 925℃, NO reduction efficiency reached a maximum value of 81% (NSR=1.5), that was to say that optimum denitration temperature was 925℃.When temperature continued to increase, NH_3 oxidation reaction rate was higher than NO reduction reaction rate, resulting in efficiency decrease with the increase of temperature. When temperature exceeded 1150℃, efficiency was lower than 10%. Only in 875-1025℃, efficiency was more than 50%, and it was the optimal temperature window.

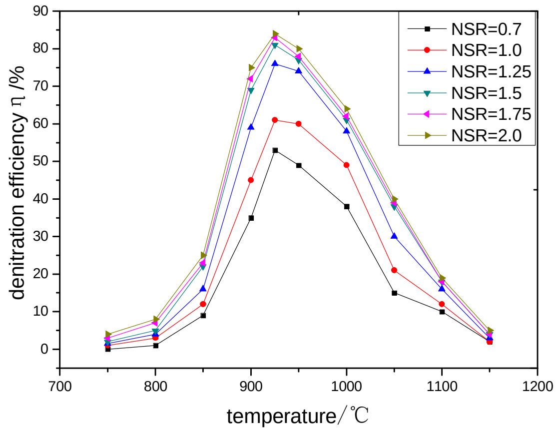


Fig.2 effect of temperature on denitration efficiency

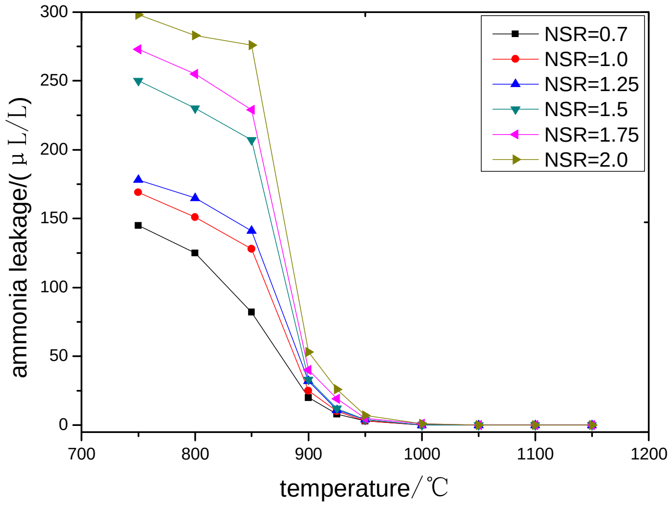


Fig.3 effect of temperature on ammonia leakage

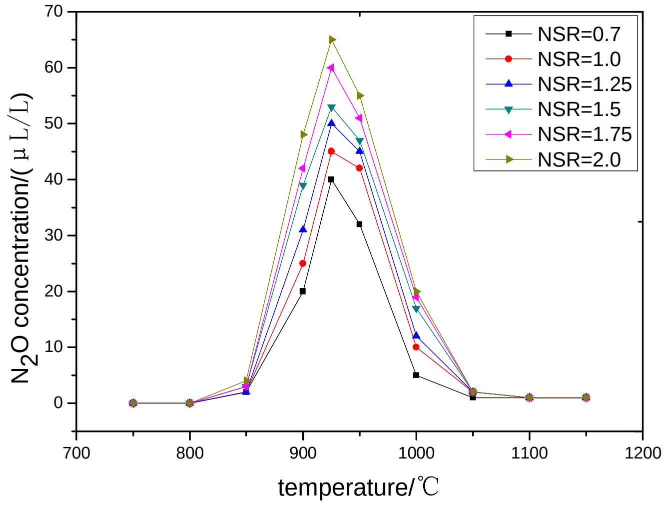


Fig.4 effect of temperature on N_2O concentration

We could see from Fig. 3, the effect of temperature on ammonia leakage was obvious. Ammonia leakage was large at low temperature, because large ammonia decomposition by urea couldn't react on oxidation or reduction reaction to be consumed at low temperature. When the temperature was higher than 800℃, with the increase of temperature, the reduction of NO and the oxidation of NH_3 were accelerated, and the two reactions were consumption of NH_3, so ammonia leakage concentration in flue gas decreased rapidly. At 950℃, ammonia leakage concentration was below 10μL/L. When temperature continued to rise, ammonia leakage was almost zero. Fig. 4 showed that N_2O concentration was low, less than 5μL/L, when temperature was below 850℃. With increasing temperature, the reaction of $NH+NO \rightarrow N_2O+H$ increased, so N_2O concentration increased rapidly. When temperature was 925℃, N_2O concentration reached a maximum value of 53μL/L(NSR=1.5). When temperature continued to increase, that N_2O began to decompose made N_2O concentration decrease rapidly. when temperature reached 1050℃, N_2O concentration was less than 5 μL/L.

According to Fig. 2 and Fig. 3, when temperature was higher than 925℃, ammonia concentration was low, and denitration efficiency decreased continually, which indicated that NH_3 was consumed largely and produced NO, also proved that oxidation of NH_3 played a main role. According to Fig. 2 and Fig. 4, denitration efficiency and N_2O concentration reached the maximum value at 925 ℃.

Effect of NSR on SNCR

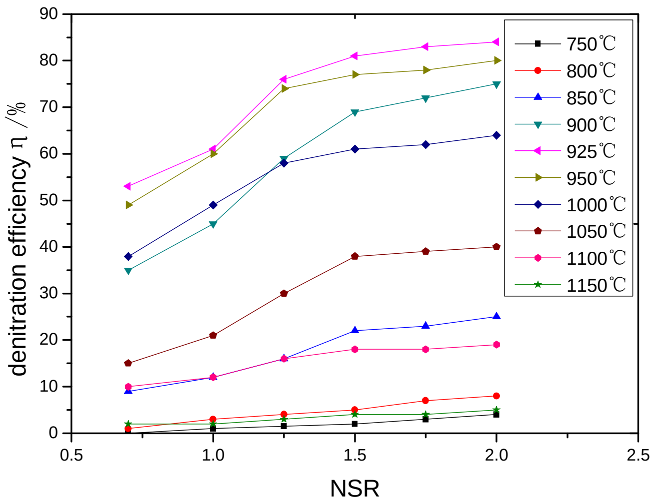


Fig.5 effect of NSR on denitration efficiency

Ammonia and nitrogen ratio (NSR) referred to the ratio of the actual and theoretical ammonia and nitrogen ratio. According to the chemical reaction equation, when NSR=1, it could completely remove NO. But for the influence of the actual process affected by factors such as the degree of mixing, NSR=1 couldn't achieve better denitration results, appropriate to increase the NSR value.

From Fig.5, the higher the NSR was, the higher denitration efficiency was. When NSR=1.5, the maximum $DeNO_X$ efficiency was 81%. But NSR was greater than 1.5, with the increase of NSR, increasing trend of removal efficiency was not obvious. when NSR=2, the maximum reduction efficiency was 84%. According to Fig. 3 and Fig. 4, the higher the NSR was, the higher ammonia leakage and N_2O concentration were, not conducive to NO reduction.

Due to increasing NSR and the decomposition of urea to generate NH_3, the excess NH_3 would lead to the increase of ammonia leakage concentration. At the same time, increasing NH_3 concentration would also accelerate the reaction, eventually to some extent resulting to N_2O concentration increasing, so NSR should not be too large. Considering denitration efficiency, ammonia leakage and N_2O concentration, the optimal NSR was 1.5.

Effect of O_2 concentration on SNCR

O_2 was an important condition for SNCR reaction, both involved in NO reduction reaction, but also the NH3 oxidation to NO. Therefore, it was essential to study the influence of O_2 concentration on the SNCR reaction.

As was shown in Fig.6, in the hypoxic conditions (O_2=0%), denitration efficiency was very low when temperature was below 1000℃. With temperature gradually increasing, efficiency increased rapidly, reaching a maximum value at

1100□. If temperature continued to increase, efficiency gradually decreased. In aerobic conditions, with the increase of temperature, efficiency at low temperature increased and it reached the maximum at 925□. DeNO$_X$ efficiency decreased if temperature continued to rise. When temperature reached 1100 □, efficiency was very low. In aerobic conditions, trends of denitration efficiency on different O_2 concentration were basically the same, reaching the maximum at about 925 □, but removal efficiency was slightly different under different circumstances.

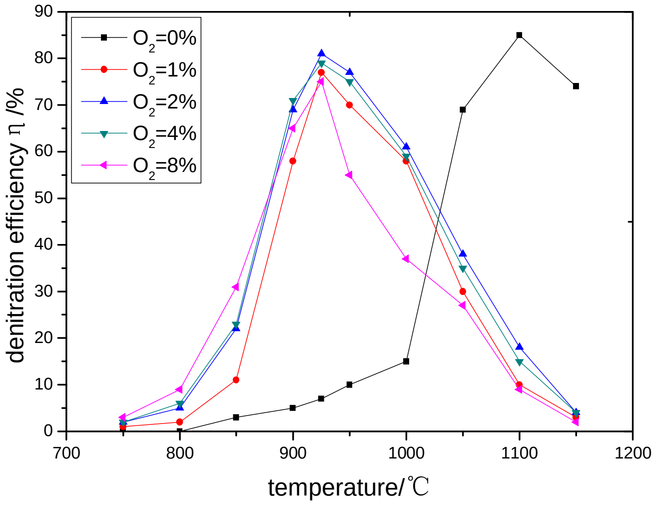


Fig.6 effect of O_2 concentration on denitration efficiency

On the Analysis of the reaction mechanism, OH, O and other active radicals were only formed by the reaction between H and H_2O in the absence of oxygen. However the reaction was extremely slow and greatly inhabits the reduction reaction at low temperature. Therefore, denitration efficiency was extremely low at low temperature. As temperature increased, the concentration of OH, O active radicals increased. It made that reduction reaction could be carried out smoothly and denitration efficiency increased. In aerobic conditions, the concentration of OH, O active radicals increased rapidly with temperature increasing, so that efficiency also increased rapidly. But temperature exceeded a certain limit, the oxidation reaction of ammonia became dominant reaction, nitrogen oxides concentration in the flue gas was even more than the original concentration. From Fig.6, the maximum denitration efficiency in anaerobic conditions was higher than the one in aerobic conditions. A possible reason was that temperature of generating OH, O and active radicals were improved by anaerobic conditions. At the same time, the oxidation of ammonia was greatly suppressed, so that the reduction reaction was more dominant at optimum temperature and there would be a higher DeNO$_X$ efficiency compared to aerobic conditions.

From Fig.6,when O_2 concentration arrived at 1%，2%，4%, denitration efficiency was high and had little difference. Considering optimum temperature window (875-1025□), the optimum O_2 concentration was between 1% and 4%.

Summary

The reaction temperature had a great influence of urea on SNCR removal of NO$_X$. The optimum denitration temperature of SNCR was about 925□, at the same time the maximum efficiency was 81%. The optimal temperature window of reduction was 875-1025□, within this interval denitration efficiency over 50%. Ammonia leakage decreased rapidly with increasing temperature. When temperature was higher than 950□, ammonia leakage concentration was below 10μL/L. With the increase of temperature, N_2O concentration also increased at first and then decreased, and at 925 □, N_2O concentration reached the maximum value of 53 μL/L.

The higher the NSR was, the higher denitration efficiency, ammonia leakage and N_2O concentration were. And the optimal NSR was 1.5.

In anaerobic conditions, denitration efficiency achieved the maximum value of 83% at 1100□. In aerobic conditions, the maximum efficiency was achieved at about 925□. The optimum O_2 concentration was between 1% and 4%.

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