Study on flue gas denitration method based on multistage high gravity technology

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Abstract—According to the mechanism of wet denitration, H_2O_2 solution and KOH solution are used as absorbents, and the two-stage countercurrent high-gravity rotating packed bed is used as the reaction equipment. The rotating speed, gas-liquid ratio, temperature, H_2O_2 solution concentration and KOH solution concentration are changed respectively to explore the impact of various factors on the denitration efficiency, and finally determine the best experimental conditions for flue gas denitration. The experimental results show that when the rotating speed reaches 900 rpm, the gas-liquid ratio is 12:1, and the temperature reaches 36 ℃, using 0.20 mol/l of H_2O_2 solution and 0.10 mol/l of KOH solution as absorbents, the denitration efficiency can reach the best, and the best denitration efficiency is 95.71%.

1. Introduction

The huge demand for electricity makes China's coal consumption still account for half of the total energy consumption [1]. Coal-fired unit power generation has always occupied a leading position in China, so it is inevitable that nitrogen oxides (NO_X) produced by coal combustion will cause important pollution to the atmosphere. It will generate acid rain and photochemical smog, which will cause great damage to forest vegetation, corrode buildings and equipment, cause human diseases, and cause damage to the ozone layer [2]~[3]. The Outline of the "Fourteenth Five-Year Plan" pointed out that the total amount of nitrogen oxide emissions in China during the "13th Five-Year Plan" period decreased by 19.7%, and set the target value of reducing the total amount of nitrogen oxide emissions during the "Fourteenth Five-Year Plan" period to be more than 10% [4], so it is urgent to study efficient denitration methods.

At present, the mainstream nitrogen oxide treatment methods are mainly divided into dry absorption method and wet absorption method. Non-selective catalytic reduction (SCR) technology, the most mature technology in dry absorption method, cannot be [5]~[6], and has become the mainstream choice of denitration technology in industry at home and abroad. However, there are shortcomings such as poor catalyst low-temperature activity, high recycling cost and liquid ammonia corrosion leakage [7]. Therefore, experts and scholars have turned their attention to the wet absorption method. Researchers [8]~[10] have studied the principle of gas-liquid mass transfer in RPB in detail. After using ozone to oxidize NO, nitric acid is used as the absorption liquid, and the denitration in RPB reaches 90%.

From the perspective of improving the high gravity rotating packed bed, our research group made a two-stage high gravity rotating packed bed as a denitration reactor to conduct systematic research on RPB denitration, and investigated the effects of rotating speed, gas-liquid ratio, temperature, H_2O_2 solution concentration and KOH solution concentration on NOx removal, which provided basic data for purifying industrial waste gas and recycling resources, as well as industrial application of denitration.

2. Introduction to the experimental part

2.1. Laboratory reagents and instruments

Experimental reagents: KOH and H_2O_2 are analytically pure reagents from China National Pharmaceutical Chemical Reagent Co., Ltd. The solutions are prepared with distilled water. Experimental instruments: the research group made a rotating packed bed reactor, the metering pump and circulating pump produced by Dalian Huanyou, the DN-200 gas vortex flowmeter, the DN-25 liquid vortex flowmeter, the induced draft fan produced by Zibo Zefa, and the Korno MOT500-NO_x nitrogen oxide detector.

2.2. Device and process

The experimental system flow is shown in Figure 1. The secondary high gravity rotating packed bed is composed of motor, shell, liquid inlet and outlet, gas inlet and outlet, packed rotor, liquid sprayer, etc. The metering pump is used to control the concentration of H_2O_2 solution and KOH solution, and the vortex flowmeter is used to control the flow of mixed absorption liquid and nitrogen oxide gas. The two-stage super-gravity rotating packed bed is used as the enhanced absorption equipment. The two meet in the packing of the rotating packed bed in a countercurrent manner. The centrifugal force generated by the rapid rotation makes the absorption liquid be divided into fine

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and dense droplets, which strengthens the mass transfer process and realizes the absorption treatment of nitrogen oxide, Detectors are installed at the air inlet and exhaust

emission outlet to calculate the denitration efficiency and complete the denitration experiment.
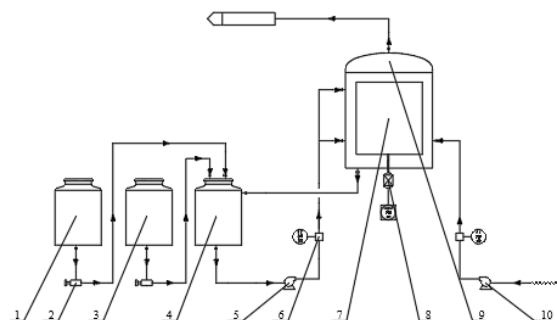

1-H_2O_2 storage tank 2-Metering pump 3-KOH storage tank 4-Reagent circulation tank 5- Circulating pump
6- Vortex Flowmeter 7- Hypergravity equipment 8- electric machinery 9- Rotating plate demister 10- Induced draft fane

Fig.1 Experimental flow chart

3. Experimental results and discussion

The main components of nitrogen oxide gas in flue gas are NO, NO_2 and N_2O_4, which react in the liquid phase. First, react with H_2O_2 solution to generate HNO_2 and HNO_3, and
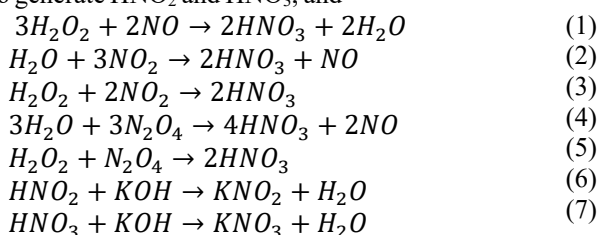
then react with KOH solution to generate products KNO_2 and KNO_3. See reaction equations (1)~(7) for the main chemical reactions.

$$3H_2O_2 + 2NO \rightarrow 2HNO_3 + 2H_2O \tag{1}$$
$$H_2O + 3NO_2 \rightarrow 2HNO_3 + NO \tag{2}$$
$$H_2O_2 + 2NO_2 \rightarrow 2HNO_3 \tag{3}$$
$$3H_2O + 3N_2O_4 \rightarrow 4HNO_3 + 2NO \tag{4}$$
$$H_2O_2 + N_2O_4 \rightarrow 2HNO_3 \tag{5}$$
$$HNO_2 + KOH \rightarrow KNO_2 + H_2O \tag{6}$$
$$HNO_3 + KOH \rightarrow KNO_3 + H_2O \tag{7}$$

3.1. Effect of rotating speed on denitration rate

First, preheat the system to 30 ℃, then adjust the circulating pump to make the liquid flow meter read 10m³/L, adjust the fan to make the gas flow meter read

100m³/L, select water, 0.05mol/L KOH solution, 0.1mol/L H_2O_2 solution and the mixed solution of the two solutions with the above concentration as reagents, finally adjust the motor frequency converter to change the speed from 150rpm to 1500rpm,and the results are shown in Figure 2.
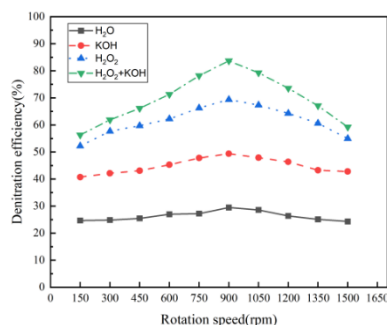

Fig.2 Effect of rotating speed on denitration efficiency

According to the analysis of Figure 2, the effect of rotating speed on the denitrification efficiency of the mixed solution of H_2O_2+KOH has the largest fluctuation, followed by 0.1mol/L H_2O_2 solution and 0.05mol/L KOH solution, and the fluctuation of aqueous solution is the smallest. However, in general, when the rotating speed is between 150rpm and 900rpm, the denitration efficiency increases with the increase of rotating speed. When the rotating speed is 900rpm, the denitration efficiency of each solution is the highest, of which the denitration

efficiency of the mixed solution of H_2O_2+KOH is 83.68%. When the rotating speed exceeds 900rpm, the denitration efficiency begins to decline with the increase of rotating speed.

According to the experimental results, at 150~900 rpm, the higher the rotating speed, the stronger the shear force of the filler, the finer the liquid droplets that the liquid phase is cut into, and then the liquid film and liquid wire are formed. Therefore, the surface renewal rate of the liquid phase is increased, the contact area of the gas phase

and the liquid phase is greatly increased, and the denitration efficiency is also improved. When the speed exceeds 900 rpm, the excessive centrifugal force causes the time of liquid phase on the packing to shorten, the contact time of gas and liquid is too short, some absorption liquid is thrown out before reaction, and the removal efficiency begins to decline, so the best speed is 900 rpm.

3.2. Influence of gas-liquid ratio on denitration rate

First, adjust the motor frequency converter, fix the speed at 900 rpm, use 0.1 mol/L H_2O_2+0.05 mol/L KOH mixed solution as absorbent, then adjust the circulating pump to keep the liquid flowmeter at 10 m³/L, adjust the fan, change the gas flowmeter reading from 20 m³/L to 300 m³/L, and finally increase the temperature from 10 ℃ to 90 ℃,and the results are shown in Figure 3.
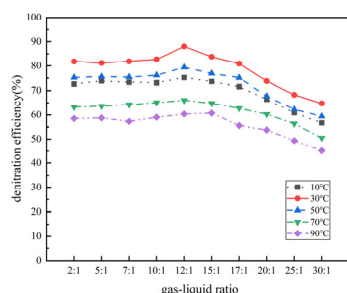

Fig.3 Effect of gas-liquid ratio on denitration efficiency

According to Figure 3, when the gas-liquid ratio is between 2:1 and 12:1, the denitration efficiency at various temperatures has a significant increase with the increase of the gas-liquid ratio. At 12:1, the highest denitration efficiency of the absorbent at 30 ℃ is 88.11%.When the gas-liquid ratio exceeds 12:1, the denitration efficiency begins to decline with the increase of the gas-liquid ratio.

In order to maintain sufficient wetness of the packing, the gas-liquid ratio is changed by changing the intake air volume.In a certain range, increasing the intake air volume has a certain positive effect on the gas-liquid mass transfer movement. But beyond this range, the excessive intake air volume causes some gases to be thrown out before they can fully contact with the liquid, resulting in the increase of the detected concentration of nitrogen oxides at the outlet and the reduction of denitration efficiency. At the same time, too large gas-liquid ratio will also increase the

power of the gas pump and increase the energy consumption. Therefore, the gas-liquid ratio should be selected according to the actual situation, taking into account both economy and efficiency. The best gas-liquid ratio is 12:1.

3.3. Effect of temperature on denitration rate

On the basis of the above experiments, adjust the fan to make the gas flow meter read 120m³/L, and choose two absorbents: one is to keep the H_2O_2 concentration unchanged to change the KOH concentration, the other is to keep the KOH concentration unchanged to change the H_2O_2 concentration, and gradually increase the reaction temperature from 30 ℃ to 50 ℃,and the results are shown in Figure 4.
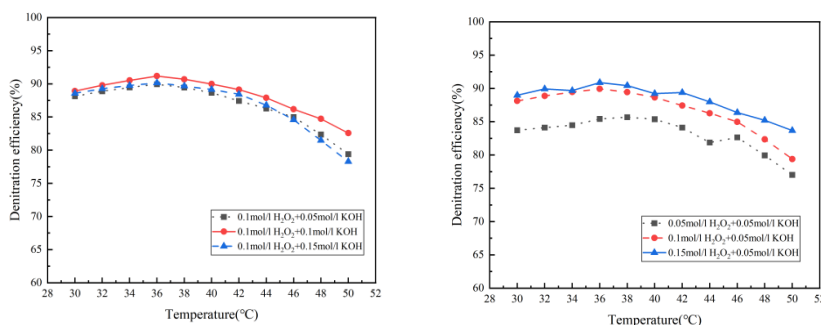

Fig.4 Effect of temperature on denitration efficiency

It can be seen from the analysis of Figure 4 that the concentration of KOH solution does not have a strong impact on the denitration efficiency. The overall trend of the denitration efficiency of the three curves with temperature is roughly the same, showing an upward trend in the range of 30 ℃ to 36 ℃, and gradually declining after exceeding 36 ℃. The highest denitration efficiency is 91.17% at 36 ℃. The effect of the concentration of

H_2O_2 solution on the denitration efficiency is stronger than that of KOH solution, The peak value of denitration efficiency of the three curves is between 36 ℃ and 38 ℃, and the highest denitration efficiency is 90.87% at 36 ℃. Temperature will affect the particle movement in the reaction process. The appropriate temperature can accelerate the collision of OH - and NO_X particles in the mixed solution, improve the reaction efficiency, and then

improve the denitration efficiency. However, overheating will affect the OH - activity and reduce the denitration efficiency. Based on the experimental results, the optimal temperature is 36 ℃.

3.4. Influence of H$_2$O$_2$ solution concentration on denitration rate

On the basis of the above experiments, the reaction temperature is adjusted to 36 ℃, the concentration of KOH solution is 0.05mol/l, 0.10mol/l and 0.20mol/l respectively, and the concentration of H$_2$O$_2$ solution gradually increases from 0.05mol/l to 0.40mol/l ,and the results are shown in Figure 5.
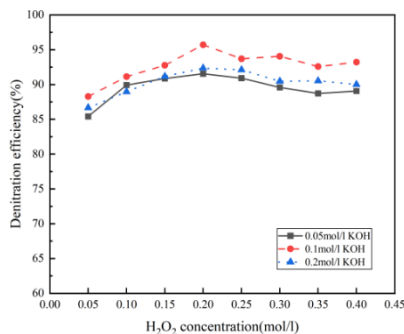

Fig.5 Effect of H$_2$O$_2$ concentration on denitration efficiency

According to Figure 5, the concentration of H$_2$O$_2$ solution is between 0.05mol/l and 0.20mol/l. With the increase of the concentration, the overall trend of denitration efficiency is gradually increasing, with the maximum of 95.71% of 0.20mol/l. After exceeding 0.20mol/l, the denitration efficiency tends to be stable. The increase of the concentration of H$_2$O$_2$ solution will strengthen the oxidation of NO, making more NO oxidized into NO$_2$ and N$_2$O$_3$ that are easily soluble in water, and thus indirectly improving the denitration efficiency of the mixed solution. However, with the further increase of the concentration of H$_2$O$_2$ solution, the main mode of denitration is the absorption reaction in progress, so the denitration efficiency begins to stabilize. At the same time, it can also be found that when the concentration of H$_2$O$_2$ solution is constant, the concentration of KOH solution is between 0.05mol/l and 0.20mol/l, The denitration efficiency increases with the concentration of KOH solution. Based on the results of experiment and analysis, considering the economy of actual production, 0.20mol/l is the best concentration of H$_2$O$_2$ solution.

3.5. Effect of KOH solution concentration on denitration rate

Based on the above experiments, the concentration of H$_2$O$_2$ solution is 0.10mol/l, 0.20mol/l and 0.25mol/l respectively, and the concentration of KOH solution gradually increases from 0.025mol/l to 0.2mol/l, The results are shown in Figure 6
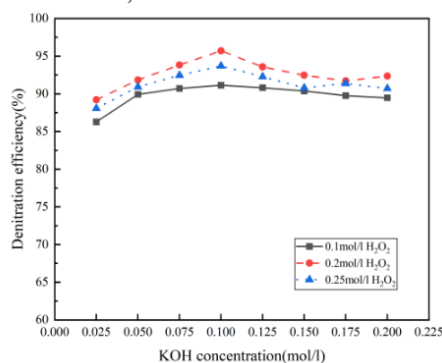

Fig.6 Effect of KOH concentration on denitration efficiency

According to Figure 6, when the concentration of KOH solution is between 0.025mol/l and 0.100mol/l, the denitration efficiency increases with the increase of KOH solution concentration, with the maximum of 95.71% of 0.100mol/l. When the concentration exceeds 0.100mol/l, the denitration efficiency decreases slightly with the increase of KOH solution concentration, and finally remains stable. The increase in the concentration of KOH solution increases the OH - ions in the solution, promotes the combination of OH - in KOH solution and H - in nitric acid, and accelerates the treatment of nitric acid and dilute

nitric acid generated by the reaction of H$_2$O$_2$ solution and nitric oxide. The reduction of the products helps to improve the chemical reaction rate. At the same time, the neutralization of nitrate and dilute nitric acid also increases the pH of the mixed solution, creating an adaptive environment for the reaction, However, after the treatment of the reactants has been completed for the KOH solution with high concentration, the excess OH - ions will further increase the pH and slightly reduce the rate of denitration reaction. Therefore, the concentration of KOH solution should be 0.1 mol/l.

4. Conclusion

Rotating speed, gas-liquid ratio, temperature, H_2O_2 and KOH solution concentration all have an impact on the denitration efficiency. After many experiments, it is found that when the rotating speed reaches 900rpm, gas-liquid ratio is 12:1, and temperature reaches 36 ℃ ,using 0.20mol/l H_2O_2 solution and 0.10mol/l KOH solution as absorbent, the best denitration efficiency is 95.71%,compared with the common single-stage high gravity machine, the denitration efficiency has been significantly improved.

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