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Research article

Optimization of wet denitration by dual oxidant ($H_2O_2/S_2O_8^{2-}$) advanced oxidation process

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ABSTRACT

A new type of wet denitration oxidant, which was composed of H_2O_2 and $Na_2S_2O_8$, had been studied for its removal performance of NO from flue gas in a bubble column reactor. The interactive effects of pH, temperature and molar ratio of $Na_2S_2O_8/H_2O_2$ on NO removal efficiency and NO absorption rate were investigated by a three-factor Box-Behnken design. The second order polynomial models were obtained to describe the relationship between the operation conditions and the responses. The results of analysis of variance such as the coefficient of determination, P values, adequate precision values and the lack of fit F-test implied the satisfactory adjustment of the quadratic models. The results of three dimensional plots showed that the interactive effect of temperature and pH had the most significant impact on these responses. The optimized parameters were successfully determined by BBD. Under the obtained optimum conditions of pH (a value of 11), temperature (50 °C) and $Na_2S_2O_8/H_2O_2$ molar ratio (a value of 0.005), NO removal efficiency of 78.68% and NO absorption rate of 7.58E-07 mol/m^2·s were observed respectively.

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1. Introduction

Sulfur dioxide and nitrogen oxides are mainly released from the coal-fired power plant and industrial boilers. These chemicals are poisonous and act as precursors for the fog-haze. Both SO_2 and NO_x are mainly responsible for regional air pollution and harm to the human health [1–4]. Due to much public pressure, the government of China has published ultra-low emissions' target for coal-fired thermal power units. These emissions' targets are that the emission concentrations of SO_2 and NO_x should be lower than 35 and 50 mg/m^3 respectively. Although the combined process consisting of wet flue gas desulfurization and selective catalytic reduction has been applied to control the emissions of these species in the coal-fired power plants of China, the process still uses an expensive catalyst to achieve the ultra-low emissions' target. Therefore, the disadvantages of high capital investment and large operational costs seriously restrict the progress of this combined technology [5–8]. Thus it is urgent to develop novel technologies having high efficiency, low cost and no associated secondary pollution to them to achieve the simultaneous removal of SO_2 and NO_x.

Due to its insolubility, NO which accounts for more than 90 % of NO_x from coal-fired flue gas is difficult to be removed by existing wet flue gas desulfurization method [9,10]. In the past, a number of chemical reagents, added to the liquid phase, have widely been studied to convert NO to NO_2, HNO_2, HNO_3 or N_2. This process is considered to be one of the most promising options to simultaneously remove SO_2 and NO_x by alkaline scrubbing. A number of chemical reagents used as additives in the liquid phase have been investigated for the removal of NO. Some of these include H_2O_2 [11], $H_2O_2/CO(NH_2)_2$ [12], NaClO [13], $NaClO_2$ [14], $NaClO_2$/NaOH [15,16], $KMnO_4$ [17], $Na_2S_2O_8$ [18], K_2FeO_4 [19], $KMnO_4$/NaOH [20], $KMnO_4/CO(NH_2)_2$ [10] and $NaClO_2/CO(NH_2)_2$ [21]. However, the high costs, secondary environmental pollutions or technical problems have hindered the industrial application of these chemicals. Hydrogen peroxide ($E^\theta(H_2O_2) = 1.77$ V) is widely favored as a clean oxidant by researchers, but its low oxidation ability restricts its application prospects. Sodium persulfate ($E^\theta(Na_2S_2O_8) = 2.01$ V) is an emerging oxidant, which is water soluble, environment friendly, and safe to handle. It has received extensive research attention in recent years. Adewuyi et al. had shown that $Na_2S_2O_8$ could be activated by temperature (23–90 °C) to generate active radicals of sulfate ($E^\theta(SO_4^{\bullet-}) = 2.60$ V) and hydroxyl ($E^\theta(HO\bullet) = 2.70$ V) with strong oxidation ability and also obtained fractional conversions of NO ranging from 10% to 92% within the temperature range of 23–90 °C [22]. Liu et al. had confirmed that $SO_4^{\bullet-}$ and HO• coexisted in an heat-activated persulfate system under 45, 55, and 75 °C, and that as the activation temperature increased, the yields of both increased [23]. It had been reported that a dual oxidant system composed of H_2O_2 and $Na_2S_2O_8$ may have several synergistic attributes due to the coexistence of the active $SO_4^{\bullet-}$ and HO• radicals [24]. In one of the previous studies, the results of contrast experiments had shown that the combination of $Na_2S_2O_8$ and H_2O_2 had a significant synergistic effect on NO removal [25,26]. The research also included

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Abbreviations

BBD	Box-Behnken design
RSM	response surface methodology
ANOVA	the analysis of variance
AP	adequate precision
3D	three dimensional
R^2	the coefficient of determination

traditional single factor experiments to study the effect of a single factor on NO removal, that was by varying a single factor while keeping all other factors constant [25,26]. But for multivariable systems, this traditional experimental method is not only time-consuming, but is also unable to obtain the true optimal parameters as it does not consider the interactions among various factors [27–29]. To solve this problem, RSM, which is a particular set of mathematical and statistical methods for analyzing the interactive effects of multiple variables on targeted responses with a minimum number of experiments, has been used for the first time in the current paper. BBD is a three-level spherical design with excellent predictability, particularly in cases when prediction of response at an extreme level is not the goal of the model. It requires minimum number of experiments while having the same number of factors as other RSM designs [30–33]. Therefore, in the present study, BBD has been chosen to investigate the interactive effects of multiple variables on the removal efficiency of NO and the absorption rate of NO.

2. Materials and methods

2.1. Materials

Standard gases included N_2 (99%) and NO span gas (99.9%). N_2 was the product of Xiang Yun Industry & Trade Co., Ltd. (Wuhan, China) and NO was the product of Oxygen Co., Ltd., of WISCO, China. Sodium persulfate (powder, ≥ 98%), 30 wt% H_2O_2, sodium hydroxide (pellets, ≥ 96%), and anhydrous calcium chloride (pellets, ≥ 96%) were the products of Sinopharm Chemical Reagent Co., Ltd. (Shanghai, China) and were of analytical grade.

2.2. Experimental procedure

The experimental setup used in this current work consisted of three sections, including a gas supply and mixing system, a bubble column reactor, and an analytical system, as shown in Fig. 1.

NO and N_2 were metered through mass flow meters from the corresponding cylinders and then were mixed in a surge flask, in which NO was diluted by N_2 to a desired concentration. This way, the mixed gas was formed. During all the experiments, the total flow of the mixed gas was kept constant at 800 mL/min by using a mass flow meter. The experimental temperature was regulated by means of a water bath connected to a digitally controlled thermostat. Until the specified temperature was reached, the mixed solution including sodium persulfate and hydrogen peroxide solutions was added in the desired concentration to the reactor. For pH-controlled experiments, appropriate quantities of NaOH solution were added. Then the mixed gas was bubbled through the reactor and data recording was initiated.

In each experiment, the gas exiting the reactor passed through the surge flask, the drying tube containing anhydrous calcium chloride which was used to remove the moisture content in the mixed gas, and finally through another surge flask in order to avoid the destruction of flue gas analyzer. The inlet concentrations of NO_x and O_2 were measured at the gas inlet. The outlet concentrations of NO_x and O_2 from the bubble column reactor were continuously monitored and recorded.

2.3. Analytical methods

The pH of reaction solution was measured by the CN113S pH meter (CNIC, Wuhan, China). The concentrations of NO_x and O_2 were analyzed by ecom-J2KN flue gas analyzer (German RBR Measurement Technology Co., Ltd., Germany).

2.4. Assessment indicators

2.4.1. NO removal efficiency

When the mixed gas was bubbled through the reactor, NO reacted with the dual oxidant ($H_2O_2/S_2O_8^{2-}$) and then was removed. The removal efficiency of NO was defined as:

$$\eta_{NO} = \frac{NO_{inlet} - NO_{outlet}}{NO_{inlet}} \quad (1)$$

where η_{NO} is the removal efficiency of NO (%), NO_{inlet} is the inlet concentration of NO (mg/m^3), while NO_{outlet} is the outlet concentration of NO (mg/m^3). The outlet concentration values of NO were obtained by taking averages after 30 min.

2.4.2. NO absorption rate

On the basis of the NO material balance, the NO absorption rate can be calculated by using Eq. (2) [34].

$$N_{NO} = 10^{-2} \cdot \eta_{NO} \cdot C_{NO,in} \cdot Q_G / 60 \cdot M_{NO} \cdot a_{NO} \cdot V_L \quad (2)$$

where N_{NO} is the NO absorption rate (mol/m^2·s), η_{NO} is the NO removal efficiency (%), $C_{NO,in}$ is the NO inlet concentration (mg/m^3), Q_G is the gas flow (L/min), M_{NO} is the NO molecular weight (g/mol) and V_L is the solution volume (mL).

$$a_{NO} = a_{CO_2} \quad (3)$$

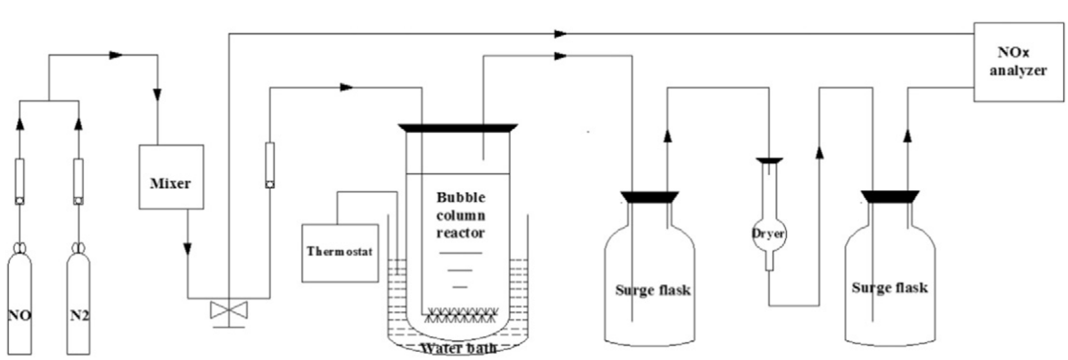

Fig. 1. Schematic of the experimental setup.

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