

original article

Kinetic modeling of nitric oxide removal by ultraviolet/H₂O₂ advanced oxidation process

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ABSTRACT

Aims: In present study, the mass transfer-reaction kinetic parameters of nitric oxide (NO) removal by ultraviolet (UV)/H₂O₂ process in a bubble column reactor in the presence of SO₂ are calculated.

Materials and Methods: The mass balance equation for NO through a layer thickness of δ , under the steady state condition is solved, and NO absorption rate is calculated. The value of rate constants and Ha numbers are obtained based on experimental data under different conditions.

Results: The calculations indicate that the values of Ha number are >3 . The values of rate constants (k_{obs}) are fitted to some empirical equations for different operating conditions. It is observed that the value of k_{obs} increases with an increase in H₂O₂ concentration and UV radiation intensity while it decreases with an increase in NO and SO₂ inlet concentrations. The values of rate constants are in order of 10⁻⁵, except for SO₂, which are in order of 10⁻⁷. The results reveal that there is a good agreement between calculated and experimental values where the maximum absolute error is 16.18% related to UV light intensities between 0 and 0.012 W/m³.

Conclusion: The obtained values of Ha numbers under different condition confirm that the absorption process of gas in the liquid phase is a fast reaction. The maximum error values resulted from a comparison between the calculated NO absorption rates and the experimental ones are acceptable.

Key words: Advanced oxidation process, kinetic model, mass transfer-reaction, ultraviolet/H₂O₂

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INTRODUCTION

SO₂ and NO_x as gaseous pollutants are released from different sources, especially from coal-fired power plants and cause serious environmental and health problems. These gases can form acid rain and photochemical smog, causing great harm to human health and ecosystems.^[1-10]

SO₂ and NO_x emissions can be controlled by combining of flue gas desulfurization and denitrification equipment.

However, the large and complex systems and high capital and operating costs limit the application of such combined systems. Recently, studying new and low-cost approaches for simultaneous removal of NO_x and SO₂ has become important issues in the field of gas purification.^[1,3-5,10]

Wet scrubbing is the most widely used approach in flue gas desulfurization (WFGD) process which ending in high SO₂

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removal efficiencies.^[11-13] A conventional WFGD process is based on using a chemical absorbent like lime or limestone slurry to converting SO₂ into waste solids which must be removed from absorbing slurry. The final product in calcium absorbent-based FGD is calcium sulfite (CaSO₃) which present many operational problems. For solving this problem, air is blown into the slurry which oxidizes CaSO₃ to CaSO₄.^[14]

However, this process fails to reach high NO_x removal efficiency because of very low solubility of nitric oxide (NO) which is a key component of NO_x and constitutes about 90–95% of the entire emission.^[1,8-10] This low solubility greatly increases the liquid phase resistance in relation to the mass transfer of NO.^[8,9] A solution to overcome this drawback could be application of complex agents such as Fe^{II}EDTA, Fe^{II} (CYS)₂, and Co^{III} (en)₃ and oxidants such as KMnO₄, Na₂S₂O₈, NaClO₂, H₂O₂, and Fenton and using sonochemical oxidation in order to increase the absorption rate of NO in solutions,^[1,7,8,13,15-17] while these technologies are still not developed because of high cost or existing technical problems.^[1,10]

Advanced oxidation processes (AOPs) are based on producing an oxidizing agent such as hydroxyl radicals to simultaneously oxidize and remove multiple pollutants. The ultraviolet (UV)/H₂O₂ process due to its strong oxidation ability, simplicity, and environmentally friendly character is widely used in wastewater treatment field.^[3,4,18-20] UV light is applied for photochemical decomposition of H₂O₂ (based on Eq. (1)) to produce hydroxyl free radicals. These radicals are very strong oxidizing agents. The redox potential of hydroxyl radicals is 2.80 eV while this value for H₂O₂ is 1.77 eV.^[5,6]



Recently, it is found that this process can be applied for simultaneous oxidizing of SO₂ and NO_x into sulfuric and nitric acid without the generation of any secondary pollution.^[10] Considering the aforementioned advantages of UV/H₂O₂ process, if applied effectively in improving the existing WFGDs, it may significantly reduce the costs related to the simultaneous removal of NO_x and SO₂.^[1,10]

The absorption of NO by UV/H₂O₂ process is essentially a reactive absorption process; therefore, the process performance is affected by chemical reaction.^[10] The study of reaction kinetics in the presence of mass transfer is an important issue. Knowing the mass transfer and kinetic parameters are essential for the numerical simulation of the process and the process development.

To the author's knowledge, there are a few studies on modeling of reaction kinetics of NO removal using UV/H₂O₂ process. Liu *et al.*,^[1] based on two-film theory, investigated the mass transfer–reaction kinetics of NO absorption from flue gas by UV/H₂O₂/NaOH process. Their findings revealed that the absorption process of NO from flue gas using

UV/H₂O₂/NaOH process is a pseudo-first-order fast reaction with respect to NO. The NO absorption rate described based on the cooperative effect of mass transfer and chemical reaction by a simple rate equation. The value of Ha numbers and rate constants are calculated in different experimental conditions.

Liu *et al.*^[6] studied the kinetic of simultaneous removal of NO and SO₂ from a simulated flue gas by the UV/H₂O₂ AOP without considering the mass transfer effect. Their results showed that the removal process of NO from SO₂-containing simulated flue gas using UV/H₂O₂ AOP is a pseudo-first-order reaction for NO. The effects of several parameters, including H₂O₂ initial concentration, UV lamp power, NO initial concentration, and reaction temperature on reaction rate constant were studied as well.

The focus of this study is on calculating the mass transfer-reaction kinetic parameters of NO absorption from SO₂-containing flue gas through UV/H₂O₂ AOP. The kinetic parameters are obtained and reported in terms of empirical reaction rate constants and Ha numbers. This study is based on experimental data reported by Liu *et al.*^[4] The kinetic parameters are obtained as a function of the concentration of the involved components and UV radiation intensity. Mass transfer-reaction kinetic parameters are important in process modeling, design or scale up procedures.

KINETIC MODELING APPROACH

In this section, the calculation procedure for obtaining the rate constant and Hatta number is presented. The major reaction pathway for the removal of NO from flue gas using UV/H₂O₂ includes the removal of NO by the oxidation of OH free radicals and the removal of NO by the oxidation of H₂O₂. The total reaction rate of NO absorption through UV/H₂O₂ process can be expressed as the sum of the oxidative removal rate of NO by OH free radical, and oxidative removal rate of NO by H₂O₂ by Eq. (2):

$$r_{\text{NO}} = -\frac{dC_{\text{NO}}}{dt} = r_{\text{OH,NO}} + r_{\text{H}_2\text{O}_2,\text{NO}} \quad (2)$$

where, r_{NO} is the total reaction rate of NO removal by using UV/H₂O₂, (mol/Ls); $r_{\text{OH,NO}}$ is the oxidative removal rate of NO by OH free radical, (mol/L s); and $r_{\text{H}_2\text{O}_2,\text{NO}}$ is the oxidative removal rate of NO by H₂O₂, (mol/L s).^[7]

The oxidative removal rate of NO by OH free radicals could be presented as Eq. (3):

$$r_{\text{OH,NO}} = k_{\text{OH}} C_{\text{OH}} C_{\text{NO}} \quad (3)$$

where, m and n are the reaction orders for H₂O₂ and NO, respectively. In addition, it is found that the oxidative removal rate of NO by H₂O₂ is a reaction with an overall order of two,

where the order of reaction with respect to each reactant is one. This reaction rate can be expressed as Eq. (4):

$$r_{H_2O_2,NO} = k_{H_2O_2} C_{H_2O_2} C_{NO} \quad (4)$$

The OH free radical has very low concentration because of its very short lifetime, so the concentration of OH free radical can be approximately assumed as a constant based on the steady-state approximation theory. Moreover in all experimental studies, the initial concentration of H₂O₂ (C_{SO_2}) is adjusted in the range of 10⁻¹–10¹ mol/L, which is much greater than that of the NO (10⁻⁴–10⁻⁵ mol/L). Therefore, the concentration of H₂O₂ can be approximately regarded as a constant, as well^[1,6,7] accordingly Eq. (4) is changed into the following Eq. (5):^[6]

$$r_{H_2O_2,NO} = k_{H_2O_2} C_{H_2O_2,0} C_{NO} \quad (5)$$

Thus, the total rate of NO absorption can be written as Eq. (6):

$$r_{NO} = -\frac{dC_{NO}}{dt} = k_{obs} C_{NO} \quad (6)$$

Based on the two-film theory, schematically shown in Figure 1, a mass balance for NO within a layer with the thickness of δ , under the steady state condition, leads to Eq. (7):

$$D_{NO,L} \frac{d^2 C_{NO}}{dx^2} = r_{NO} \quad (7)$$

where, $D_{NO,L}$ is the molecular diffusion coefficient of NO in the liquid phase (m²/s); x is the differential element thickness in the liquid film (m).

The Eq. (7) can be solved based on the following boundary conditions:^[21]

$$\text{At } x = 0, C_{NO} = C_{NO,i} \text{ and } x = \delta, C_{NO} = C_{NO,L}$$

It is assumed that the absorption of NO by UV/H₂O₂ is a fast reaction. Therefore, the concentration of NO in the bulk of liquid equals 0 ($C_{NO,L} = 0$).^[1,7]

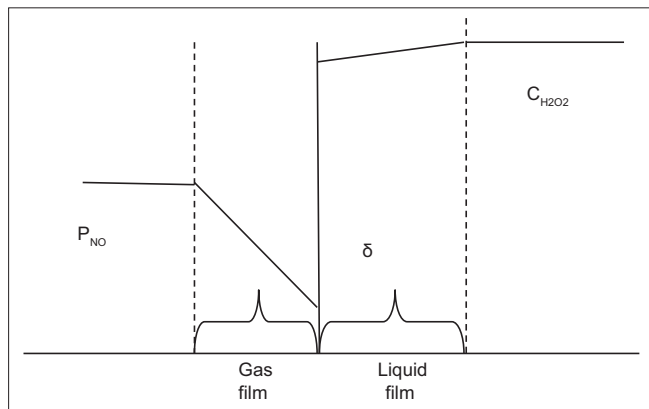


Figure 1: Schematic representation of two-film theory

Solving the Eq. (7) leads to NO concentration distribution in liquid as described by Eq. (8)

$$C_{NO} = \frac{C_{NO,i} \sinh[(\delta - x) \sqrt{\frac{k_{obs}}{D_{NO,L}}}]}{\sinh[\delta \sqrt{\frac{k_{obs}}{D_{NO,L}}}] \quad (8)$$

The rate of transfer or flux of NO into the liquid film, $N_{NO}(x = 0)$, equals the diffusion rate at the gas-liquid interface:^[7,21]

$$N_{NO}(x = 0) = -D_{NO,L} \left(\frac{dC_{NO}}{dx} \right)_{x=0} = \frac{C_{NO,i} \sqrt{k_{obs} D_{NO,L}}}{\tanh\left(\sqrt{\frac{k_{obs} D_{NO,L}}{k_{NO,L}}}\right)} \quad (9)$$

where, $k_{NO,L}(D_{NO,L}/\delta)$ and $C_{NO,i}$ are the liquid phase mass transfer coefficient (m/s) and the interface concentration, (mol/L), respectively. In the case of a fast reaction (i.e., very large value of k_{obs}), the denominator in Eq. (9) is almost one and this equation can be rewritten as Eq. (10):

$$N_{NO} = C_{NO,i} \sqrt{k_{obs} D_{NO,L}} \quad (10)$$

It should be noted that the argument of tanh function in the denominator of Eq. (11) is a definition of Hatta number for a first-order reaction.

$$Ha = \left(\frac{\sqrt{k_{obs} D_{NO,L}}}{k_{NO,L}} \right) \quad (11)$$

The two-film theory holds that under steady-state condition, the absorption rate of NO removal can be described as the following Eq. (12):

$$N_{NO} = k_{NO,G}(P_{NO,G} - P_{NO,i}) = Ek_{NO,L}(C_{NO,i} - C_{NO,L}) \quad (12)$$

where, N_{NO} is the absorption rate of NO, (mol/m²s), $k_{NO,G}$ is the gas phase mass transfer coefficient, (mol/s m²Pa); $P_{NO,G}$ is the NO partial pressure in bulk of gas, (Pa); $P_{NO,i}$ is the NO partial pressure in gas-liquid interface, (Pa); $C_{NO,L}$ is the NO concentration in the bulk of liquid, (mol/L); $k_{NO,L}$ is the liquid phase mass transfer coefficient (m/s), and E is the chemical reaction enhancement factor.

$C_{NO,i}$ can be correlated to $P_{NO,i}$ based on Henry's law (Eq. (13)):

$$P_{NO,i} = H_{NO,L} C_{NO,i} \quad (13)$$

where, $H_{NO,L}$ is Henry constant (Pa.m³/mol), which can be calculated by Eq. (14)

$$\log\left(\frac{1}{H_{NO}}\right) = \frac{-1463.32}{T} + 2.178 \quad (14)$$

The Eq. (14) is the Van Krevelen and Hofitzer empirical equation where, H_{NO} is the solubility of NO in the liquid phase (mol/L. atm).^[1]

Through manipulation of Eq. (12), $C_{NO,i}$ can be calculated by Eq. (15):

$$C_{NO,i} = \frac{1}{H_{NO,L}} (P_{NO,G} - \frac{N_{NO}}{k_{NO,G}}) \quad (15)$$

The NO diffusion coefficient in the liquid phase is calculated using Wilke and Chang empirical Eq. (16) as follows.^[1]

$$D_{NO} = 7.4 \times 10^{-12} \frac{(\phi_w M)^{0.5} T}{\mu_w V_A^{0.6}} \text{ m}^2 / \text{s} \quad (16)$$

where, j_w is the association factor, 2.6 for water; V_A is the molar volume of gas, 23.9 cm³/mol for NO; D_A is the diffusion coefficient of the solute A at very low concentrations in water (m²/s); and T is temperature in K.

The value of $D_{NO,L}$, $H_{NO,L}$, and the value of $k_{NO,G}$ are summarized in Table 1. The value of the gas phase mass transfer is the same as reported by Liu *et al.*^[7]

The values of k_{obs} under different experimental conditions are calculated through Eq. (10), where N_{NO} is calculated based on experimental data from Liu *et al.*^[4] by Eq. (17) and $C_{NO,i}$ can be calculated by Eq. (15). The experimental values are reported in Table 2.

$$N_{NO} = \frac{\eta C_{NO,in} Q_G}{1344 V_L a_{NO}} \quad (17)$$

where, $C_{NO,in}$ is the inlet concentrations of NO, (ppm_v), Q_G is the volumetric gas flow (L/min); V_L is the solution volume, (mL) and η is NO removal efficiency defined by Eq. (18):

$$\eta = \frac{C_{NO,in} - C_{NO,out}}{C_{NO,in}} \times 100 \quad (18)$$

where, $C_{NO,out}$ is the outlet concentration of NO, (ppm_v).

The calculation of interfacial area and liquid phase mass transfer coefficient

The gas-liquid interfacial area depends on gas hold-up value (ϵ_G) and almost all available correlations for the gas hold-up, and liquid mass transfer coefficient depends on the gas superficial velocity. Based on the gas flow rate (500 mL/min) and column diameter (8 cm) reported by the,^[1] the value of superficial gas velocity is calculated by Eq. (19):

$$U_G = \frac{Q_G}{A_c} \quad (19)$$

where, A_c is the column surface area, (m²).

Table 1: Henry constant, diffusion and gas phase mass transfer coefficient

| $H_{NO,L} \times 10^{-5}$ (m ³ .Pa/mol) | $D_{NO,L} \times 10^9$ (m ² /s) | $k_{NO,G} \times 10^6$ (mol/m ² .s.Pa) |
|-------------------------------------------------------|-----------------------------------------------|------------------------------------------------------|
| 0.55 | 2.52 | 1.86 |

Table 2: Experimental condition for NO absorption by UV/H₂O₂ process (H₂O₂ solution volume: 600 mL, gas flow rate: 500 mL/min, solution temperature: 298 K)

| Parameters | η_{NO} (%) |
|----------------------|-----------------|
| C_{NO} (ppm) | |
| 200 | 81.8 |
| 450 | 72 |
| 650 | 68.3 |
| 850 | 65.5 |
| 1100 | 58.5 |
| $C_{H_2O_2}$ (mol/L) | |
| 0.3 | 27 |
| 0.6 | 48.6 |
| 1 | 62.2 |
| 1.5 | 68.9 |
| 2 | 70.8 |
| 2.5 | 74 |
| C_{SO_2} (ppm) | |
| 1000 | 68.3 |
| 1500 | 57.4 |
| 1900 | 52.9 |
| 2500 | 50.4 |
| UV intensity (W/mL) | |
| 0 | 8.69 |
| 0.003 | 18.2 |
| 0.006 | 27.69 |
| 0.008 | 34.99 |
| 0.01 | 40 |
| 0.012 | 59.5 |
| 0.018 | 72 |
| 0.024 | 76.60 |
| 0.03 | 79.89 |

UV: Ultraviolet

The following correlations (Eqs.(20-22)) are developed by López-López *et al.*^[22] for gas hold-up and liquid phase mass transfer.

$$\epsilon_G = 4.86 U_G \quad (20)$$

$$d_{32} = 0.14 U_G + 0.0031 \quad (21)$$

where, d_{32} is the suture diameter of gas bubbles.

$$k_L a = 3.6 U_G \quad (22)$$

The gas-liquid interfacial area can be calculated by Eq. (23):

$$a = \frac{6 \epsilon_G}{d_{32}} \quad (23)$$

The values of calculated parameters are tabulated [Table 3].

RESULTS

The values of reaction rate constant are calculated based on Eq. (10) where CNO,i and NNO are obtained by Eq. (15) and Eq. (17), respectively. These values are fitted into the following empirical Eqs. (24) to (27) with NO concentrations, H₂O₂ concentrations, SO₂ concentrations and UV radiation intensities. The results are also illustrated in [Figure 2 a-e].

$$k_{obs,NO} = 2.21 \times 10^5 \exp(-8 \times 10^{-4} C_{NO}) s^{-1}$$

$$R^2 = 0.971, 200 \leq C_{NO} \leq 1100 \text{ ppm}_v \quad (24)$$

$$k_{obs,H_2O_2} = [0.623 \ln(C_{H_2O_2}) + 0.978] \times 10^5 s^{-1}$$

$$R^2 = 0.995, 0.3 \leq C_{H_2O_2} \leq 2.5 \text{ mol/L} \quad (25)$$

$$k_{obs,SO_2} = 2.45 \times 10^7 C_{SO_2}^{-0.76} s^{-1}$$

$$R^2 = 0.973, C_{SO_2} \geq 1000 \text{ ppm}_v \quad (26)$$

$$k_{obs,UV} = \{(7094UV^2 - 20.01UV + 0.045) \times 10^5 s^{-1},$$

$$R^2 = 0.993, 0 \leq UV \leq 0.012 \text{ W/mL},$$

$$(30.89UV + 0.916) \times 10^5 s^{-1},$$

$$R^2 = 0.995, 0.012 \leq UV \leq 0.03 \text{ W/mL} \quad (27)$$

Table 3: The values of gas superficial velocity, gas-liquid interfacial area and liquid phase mass transfer

| $U_G \times 10^3 \text{ (m/s)}$ | $a_{NO,L} \text{ (1/m)}$ | $k_{NO,L} \times 10^4 \text{ (m/s)}$ |
|---------------------------------|--------------------------|--------------------------------------|
| 1.7 | 15.1 | 4.1 |

As observed the k_{obs} increases with an increase in H₂O₂ concentrations and UV radiation intensities, while it decreases with an increase in NO concentrations and SO₂ concentrations. Since the NO absorption by UV/H₂O₂ process is a pseudo first order reaction, the NO absorption rate is proportional to the k_{obs} . Therefore the NO removal follows the same trend as k_{obs} with different parameters. This trend emphasizes the fact that the NO absorption rate increases with an increase in H₂O₂ concentrations and UV radiation intensities, while decreases with an increase in NO concentrations and SO₂ concentrations as indicated by Liu *et al.*^[4]

DISCUSSION

As can be seen from Eq. (24) to (27) and [Figure 2 b-e], the rate constant values increase by increasing the H₂O₂ concentration and UV light intensity per liquid volume. It is obvious that an increase in H₂O₂ concentration or UV intensity means more OH free radicals and consequently more NO absorption. However, an increase in NO concentration results in decrease in rate constant value. Increasing in NO content means more NO molecules compete for absorbing by specified amount of hydroxyl radicals which results in a decrease in NO absorption rate. Also, Eq. (26) and [Figure 2c] confirm that the values of rate constant decrease with an increase in SO₂ concentration. It reveals that at SO₂ concentration higher than 1000 ppm the rate of NO absorption is decreased when SO₂ concentration decreases which can be related to competitive behavior of this gas in consumption of hydroxyl radicals. It means that

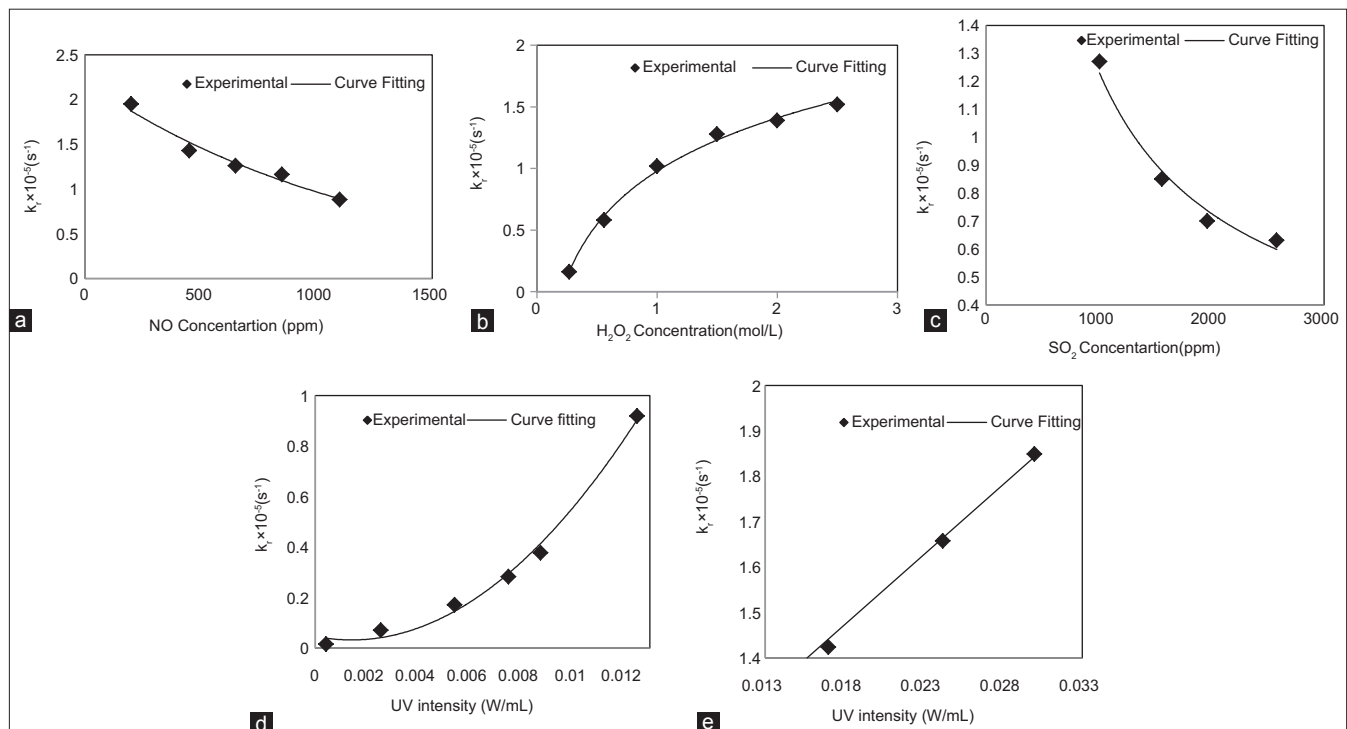


Figure 2: The effect of concentration of involved components in the reaction and radiation intensity on k_{obs} ; (a) Effect of nitric oxide concentration, (b) Effect of H₂O₂ concentration (c), Effect of SO₂ concentration and (d and e) Effect of Ultraviolet radiation intensity

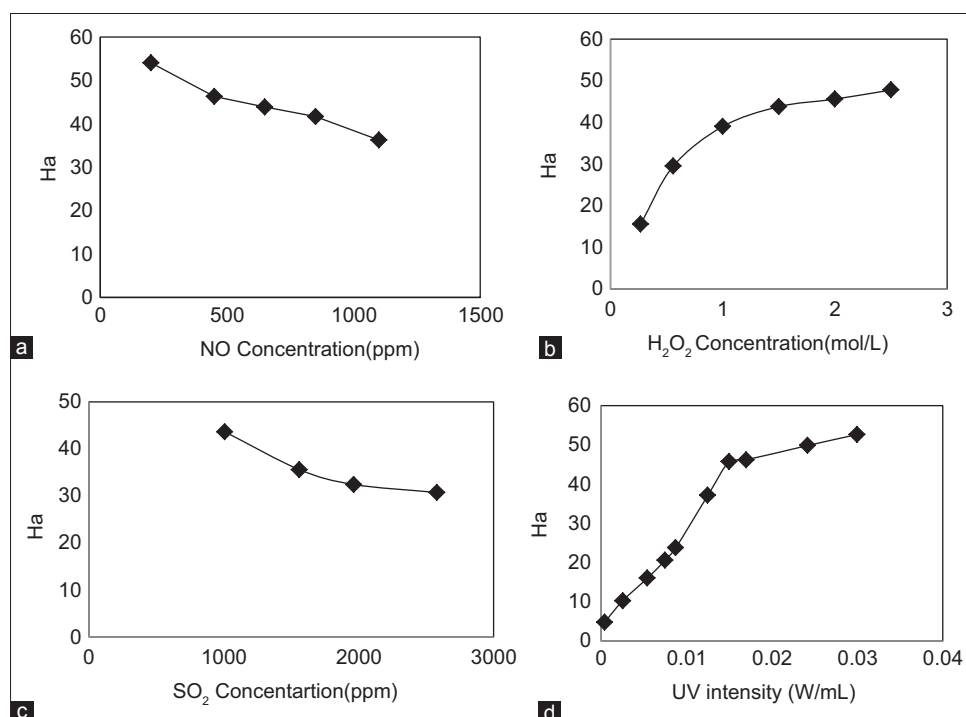


Figure 3: The values of Ha numbers at different (a) nitric oxide concentration, (b) H₂O₂ concentration, (c) SO₂ concentration and (d) Ultraviolet radiation intensity

in presence of SO₂ lower amount of hydroxyl radicals are available for NO oxidation.

The values of N_{NO} are calculated by Eqs. (10) and (24) to (27) and compared with the experimental values obtained by Eq. (17) [Table 4]. It is observed that there is a good agreement between calculated and experimental values where the maximum absolute error is 16.18%. This error relates to the values of N_{NO} calculated from different UV intensities between zero to 0.012 W/mL. The value of absolute error is obtained by Eq. (28):

$$Absolute\ error = \frac{\sum_i^n |N_{NO,cal} - N_{NO,exp}|}{n} \times 100 \quad (28)$$

where, *n* is the number of data points in each experiment.

Thus, the Eqs. (10) and (24) to (27) can be used to simulate the absorption process of NO by using UV/H₂O₂ advanced oxidation process.

Based on calculated *k_{obs}*s, the Has are calculated by Eq. (11) under different experimental conditions where, *k_{NO,L}* is taken from Table 3. The results are shown in Figure 3a-d, where all of the Ha numbers are greater than 3. This reveals that the removal process of NO through UV/H₂O₂ wet scrubbing is a fast reaction.

CONCLUSION

The mass transfer-reaction kinetic parameters of NO removal

Table 4: The values of errors relates to the values of N_{NO} calculated by Eqs. (9) and (24) to (27)

| Absolute error, NO | Absolute error, H ₂ O ₂ | Absolute error, SO ₂ | Absolute error, UV First equation | Absolute error, UV Second equation |
|--------------------|-----------------------------------------------|---------------------------------|-----------------------------------|------------------------------------|
| 2.18 | 1.38 | 2.29 | 16.18 | 0.41 |

UV: Ultraviolet

in UV/H₂O₂ AOP are obtained. The calculations reveal that under different experimental conditions, the values of Ha numbers are > 3, confirming that the removal of NO by absorption through the liquid film is a fast reaction. The values of reaction rate constants *k_{obs}* are calculated as well. These values are fitted by empirical equations, and the NO absorption rates are calculated through these equations under different conditions. The calculated values of N_{NO} are in good agreement with experimental ones. The maximum value of absolute error is 16.18% which is reasonable in this context.

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University of Isfahan, Isfahan, Iran.

Conflicts of interest

There are no conflicts of interest.

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