Nitrogen oxides absorption into aqueous nitric acid solutions containing hydrogen peroxide tested using a cables-bundle contactor

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A B S T R A C T

This work aims to study the removal of dilute nitrogen oxides from gaseous effluents by absorption into aqueous nitric acid solutions containing or not hydrogen peroxide.

NOx absorption tests (pNOx = 50 Pa with a varying composition) were performed in a cables-bundle contactor at ambient conditions. Tests runs were carried out with the recycling of the absorbent solution (0–8 mol/l for HNO_3 and 0–2 mol/l for H_2O_2) allowing to observe the temporal evolution of the absorption efficiency. The effects of the different operating conditions on the NO, NO_2 and NOx absorption performances (A) were investigated.

Results revealed in all the cases that A_{NOx} increases when increasing the proportion of more oxidized species in NOx. Without H_2O_2 A_{NOx} decreases when the HNO_3 concentration of the solution is increased and also with the recycling of the scrubbing liquid, leading to NOx desorption phenomena. When hydrogen peroxide is added to the nitric acid solution the NOx absorption performances are much higher and increase with the initial H_2O_2 concentration and the HNO_3 concentration.

1. Introduction

Nitrogen oxides emissions have a harmful effect on humans and environment. In fact, they contribute to the photochemical smog (Dvorak et al., 2010; Liang et al., 2011; Chen and Tan, 2012) and acid rain (Dvorak et al., 2010; Liang et al., 2011; Chen and Tan, 2012). Since more than 3 decades, NOx emissions by industrial sources have become the focus of air pollution control. Therefore, several methods have been developed in order to reduce NOx emissions including dry processes such as the famous selective catalytic reduction (SCR) (Jin et al., 2006; Dvorak et al., 2010; Liang et al., 2011; Sun et al., 2013; Xu et al., 2013). SCR using ammonia (or urea) as reagent is considered as a very effective technique but it presents disadvantages such as the high investment cost, the potential catalyst poisoning, the limited catalyst lifetime and the risk of ammonia slip (Koebel et al., 2000) which negatively influences environmental and public health as well as climate change (Behera et al., 2013). Selective non-catalytic reduction (SNCR) (Jin et al., 2006; Dvorak et al., 2010; Liang et al., 2011; Sun et al., 2013; Xu et al., 2013) requires more reagent and high temperatures to be operative. Its efficiency is also lower. Electro-scrubbing process via a redox medium like Ag(II)/Ag(I) or Ce(IV)/Ce(III) was found to lead to

high NOx removal performances (Pillai et al., 2009; Chung and Moon, 2013). However, it is still scarcely used in industrial applications because it requires high energy consumption (Chiba et al., 1994). Wet techniques use various reactants (Jin et al., 2006; Dvorak et al., 2010; Zhao et al., 2010; Khan and Andewuji, 2010; Sun et al., 2013) since water scrubbing tends to be very inefficient, being strongly limited by the relatively inert nature and low solubility of nitric oxide NO. Different solvents such as aqueous solutions of sodium hydroxide or calcium hydroxide (Patwardhan et al., 2002), hydrogen peroxide (De Paiva and Kachan, 1998; Myers and Overcamp, 2002; Liémans and Thomas, 2013; Li et al., 2014), sodium chlorite (Chu et al., 2001; Myers and Overcamp, 2002; Li et al., 2014) or potassium permanganate (Brogen et al., 1997; Chu et al., 2001; Myers and Overcamp, 2002; Li et al., 2014) can be added to the scrubbing solution in order to neutralize NOx or to convert by oxidation the insoluble NOx species to more soluble ones. Another example is given by Wang et al. (2016) who has demonstrated that the dual oxidant $H_2O_2 + S_2O_8^{2-}$ system can be used to remove efficiently NO, the optimal performances being reached at a pH equal to 11 and for the molar ratio of H_2O_2 to $Na_2S_2O_8$ of 0.3:0.1.

Actually, NOx absorption into acidic solutions containing hydrogen

Nomenclature

A_{NOx}	Absorption efficiency of NOx (%)		NOx oxidized species in total NOx) (%)
A_{NO}	Absorption efficiency of NO (%)	P	Potal pressure (Pa)
A_{NO_2}	Absorption efficiency of NO₂ (%)	P_{NOx}	Partial pressure of NOx (Pa)
[j]	Concentration of species j in the aqueous phase (kmol/m³)	P_j	Partial pressure of species j in the gas phase (Pa)
G	Gas flow rate (m³/s)	T	Temperature (K)
H_j	Henry's law constant (kmol/m³.Pa)	V_L	Volume of recycled liquid (m³)
L	Liquid flow rate (m³/s)	$y_{NOx,in}$	NOx molar fraction at the inlet of the absorption column
OR	Oxidation ratio of NOx in the gas phase (proportion of	$y_{NO,in}$	NO molar fraction at the inlet of the absorption column
		$y_{NOx,out}$	NOx molar fraction at the outlet of the absorption column

peroxide as oxidizing agent can be regarded as a suitable technology (Chu et al., 2001; Patwardhan et al., 2002) since it allows the elimination of gaseous pollutants in a clean way, without generating liquid wastes (De Paiva and Kachan, 1998; Thomas and Vanderschuren, 1998a; Liémans and Thomas, 2010, 2013). The oxidation product of NOx by H₂O₂ can be advantageously recovered and recycled.

Numerous studies (Thomas and Vanderschuren, 1996, 1997; De Paiva and Kachan, 2004; Liémans et al., 2011) have been conducted at relatively high values of NOx partial pressure, above 50 Pa, absorbing NOx into nitric acid solutions of different concentrations containing or not hydrogen peroxide. However, very few papers and reports (Thomas and Vanderschuren, 1998b) have been published on the application of this process at low partial pressures.

The present study investigates the NOx absorption into nitric acid solutions containing or not hydrogen peroxide at lower values of NOx partial pressures (below 50 Pa) and lower oxidation ratios (proportion of more oxidized species in total NOx ≤ 50%) which are the typical conditions of NOx emissions in an industrial gas from a Tunisian acid plant (TCG, 2016). This study was developed in order to design a new full-scale scrubbing system for NOx emission reduction issued from this plant.

Different absorption tests achieved using a laboratory absorption device called "cables-bundle contactor" were here analyzed to illustrate the effects on NOx absorption performances of the gaseous phase NOx oxidation ratio, the initial H₂O₂ and HNO₃ concentrations and the recycling of the scrubbing solution.

2. Reactions involved during NOx absorption

Absorption of NOx into aqueous solutions is a very complex mechanism owing to the numerous chemical reactions involved both in the gas and liquid phases.

The gas-phase reactions are given as follows:

$$2\,NO + O_2 \rightarrow 2\,NO_2. \tag{R1}$$

$$2\,NO_2 \leftrightarrow N_2O_4. \tag{R2}$$

$$NO + NO_2 \leftrightarrow N_2O_3. \tag{R3}$$

$$NO + NO_2 + H_2O \leftrightarrow 2\,HNO_2. \tag{R4}$$

The liquid-phase reactions are:

$$2NO_2 + H_2O \rightarrow HNO_3 + HNO_2 \tag{R5}$$

$$N_2O_3 + H_2O \rightarrow 2\,HNO_2. \tag{R6}$$

$$N_2O_4 + H_2O \rightarrow HNO_3 + HNO_2 \tag{R7}$$

$$3HNO_2 \rightarrow HNO_3 + H_2O + 2\,NO \tag{R8}$$

The liquid-phase reactions are considered as irreversible and kinetically controlled rions (Hupen and Keing, 2005; Dalaouti and Seferlis, 2005). Via reactions (R. 5) (R. 6) and (R. 7), nitrogen dioxide NO₂, dinitrogen trioxide N₃ and dinitrogen tetraoxide N₂O₄ compounds react rapidly with water (hydrolyses reactions) and produce nitrous and nitric acids. Nitrous acid HNO₂ is unstable and can decompose with the

Table 1

NOx Henry's law constants at T = 293 K $\left(law: H_j = \frac{[j]}{P_j}\right)$.

Nitrogen oxide species	NO	NO₂	N₂O₃	N₂O₄
H_j kmol/m³.Pa)	$1.80\ 10^{-8}$	$1.2\ 10^{-7}$	5.9^{10-6}	1.38^{10-5}

release of NO (R. 8) while nitric acid is stable in the liquid phase (Hupen and Keing, 2005; Dalaouti and Seferlis, 2005).

T Henry's constants for the different NOx species (NO, NO₂, N₂O₃ and N₂O₄) are presented in Table 1 (Patwardhan and Joshi, 2003) at 25 °C. At this temperature the Henry's constants for NO₂, N₂O₄ and N₂O₃ are largely higher than the one for NO, underlining a very low solubility of this last component.

Thus, conversion of both NO and HNO₂ into HNO₃ will result in a significant increase in the absorption process performances.
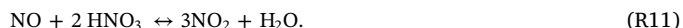
It has been found that NO reacts with nitric acid via the following reaction (Carta and Pigford, 1983; Weisweiler et al., 1991):
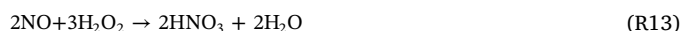
$$2\,NO + HNO_3 + H_2O \leftrightarrow 3HNO_2. \tag{R9}$$

Subsequently

$$2HNO_2 \rightarrow NO + NO_2 + H_2O. \tag{R10}$$

The sum of two reactions (R.9) and (R.10) can be written as follows:

$$NO + 2\,HNO_3 \leftrightarrow 3NO_2 + H_2O. \tag{R11}$$

A it is reported that the presence of H₂O₂ in the liquid phase improves NOx absorption thanks to the oxidation of HNO₂ (Thomas and Vanderschuren, 1998a; Lee and Lind, 1986) and NO (Baveja et al., 1979) into HNO₃ according to reactions (R2) and (R.13), respectively.

$$HNO_2 + H_2O_2 \rightarrow HNO_3 + H_2O. \tag{R12}$$

$$2NO + 3H_2O_2 \rightarrow 2HNO_3 + 2H_2O \tag{R13}$$

Reactions (R.12) and (R.13) are known to be fast and irreversible. Also, it was shown that the presence of nitric acid promotes the oxidation reaction of HNO₂ by H₂O₂ (R.12) thanks to an auto-catalytic effect of the ion H⁺ (Liémans et al., 2011).

T complete NOx absorption mechanism into water and into nitric acid solutions containing or not hydrogen peroxide can be schematized by Fig. 1.

3. Materials and methods

3.1. Experimental set up

The experimental setup is shown in Fig. 2 (a) and (b). The absorption device is a homemade ring-shaped column made of a vertical glass tube in the axis of which stands a polypropylene rod which supports 6 twisted polypropylene cables, constituting the packing (Fig. 2 (a)). The dimensions of the contactor are given in Table 2. The gas-liquid contact takes place around the six cables and the device is well suited for such laboratory studies in order to compare absorption performances linked to liquid phase kinetics. The carrier gas is preliminarily humidified in a
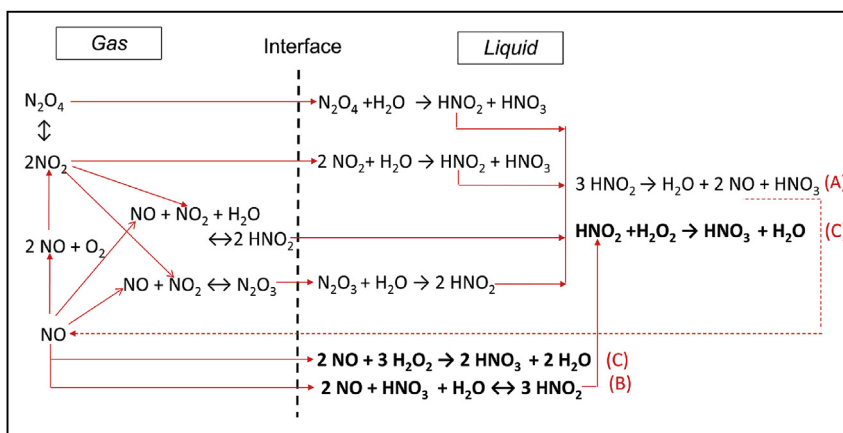
Fig. 1. Absorption mechanisms of NOx into water (A), into nitric acid solutions (B) and into nitric acid solutions containing hydrogen peroxide(C) (adapted from Thomas and Vanderschuren, 1996).
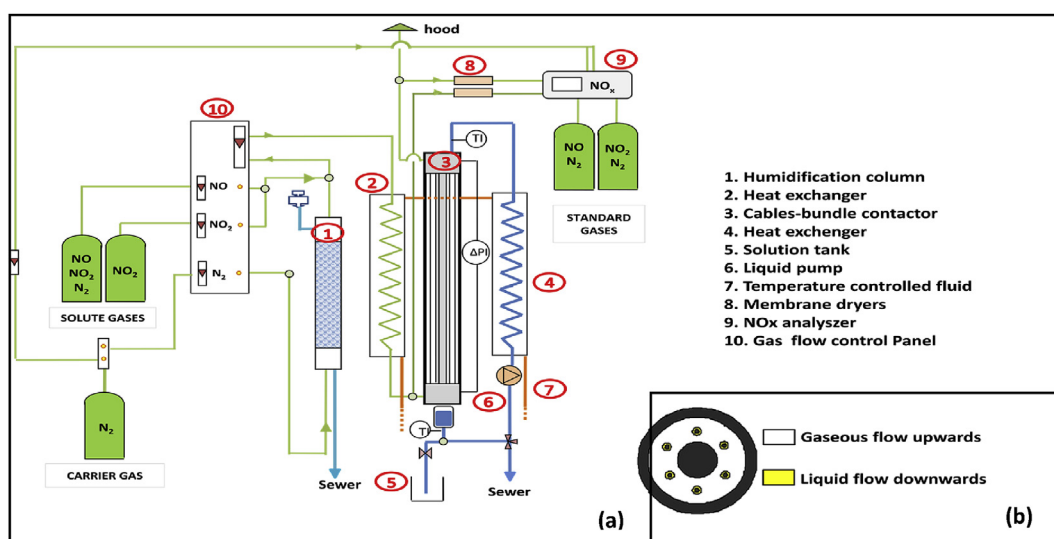

Fig. 2. (a) Experimental apparatus, **(b)** Horizontal cross-section of the contactor.

Table 2
Contactor dimensions.

Contactor	Useful height	Inside diameter	Diameter of the central rod	Cable diameter
Dimensions (m)	0.54	0.045	0.02	$1.7 \ 10^{-3}$

Table 3
Operating conditions of the absorption tests.

P_{NOX}(Pa)	Around 50
OR (%)	5 - 30–50
$[H_2O_2]$ (M)	0–0.2–1 – 2
$[HNO_3]$ (M)	0–2 – 4–6 -8
G (m³/s)	$2.77 \ 10^{-4}$
L (m³/s)	$3.63 \ 10^{-6}$
V_L	$3 \ 10^{-4}$ m³
T (K)	293
P (kPa)	101.325

Fig. 3. NOx absorption efficiencies into water versus recycling time for three OR values.

saturator (a small packed column fed with distilled water), in which NOx are added to obtain the desired concentration. The gas enters axially at the bottom of the gas-liquid contactor flowing upwards, while the prepared scrubbing solutio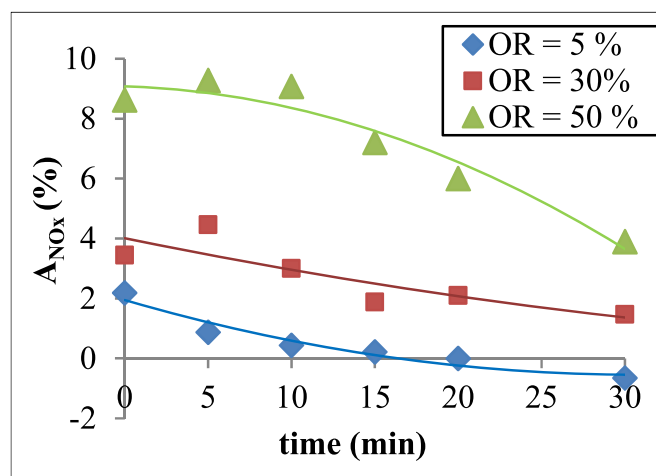n is fed to the top distributing chamber thanks to a peristaltic pump, flowing downwards. The liquid is distributed around each cable through individual holes drilled in the bottom of the chamber. A steady flow having the shape of cylindrical films is set up around the yarns (Fig. 2 (b)). The liquid and gas feed
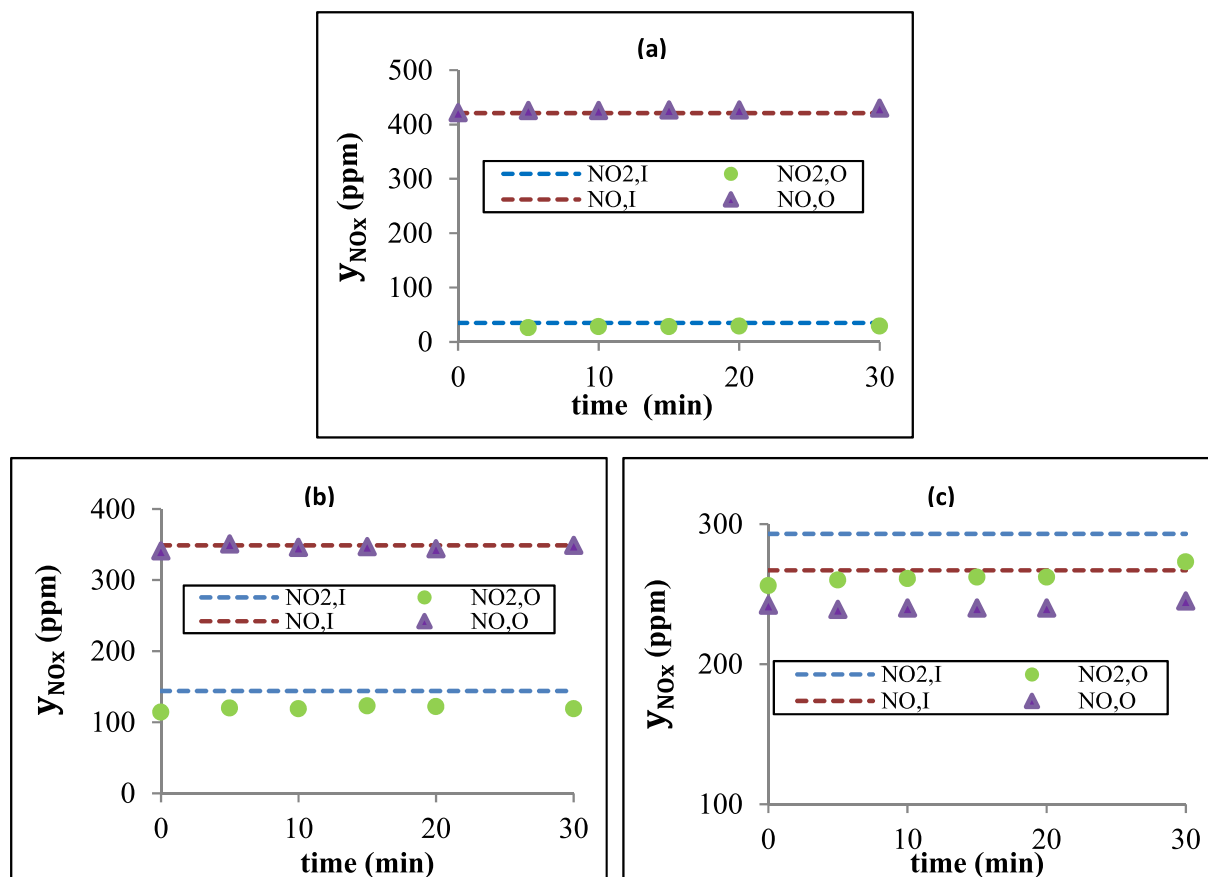
Fig. 4. Evolution of NO and NO$_2$ input (NO$_{2I}$, NO$_I$) and output (NO$_{2O}$, NO$_O$) concentrations versus recycling time for absorption into water: **(a)** for OR = 5%, **(b)** OR = 30% and **(c)** OR = 50%.
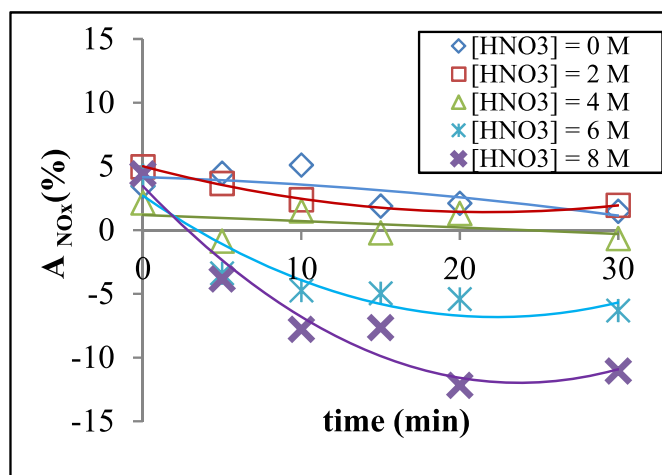

Fig. 5. NOx absorption efficiencies into nitric acid solutions over time, for OR = 30%.

temperatures are controlled by means of a thermostatic bath. Operation conditions are given in Table 3. The gas flow rate was maintained at 2.77 10^{-4} m^3/s corresponding to a superficial velocity of 0.22 m/s and the liquid flow rate was fixed at 3.63 10^{-6} m^3/s.

Absorption tests were performed for various oxidation ratios OR (5, 30 and 50%) with 50 Pa as NOx partial pressure, gaseous conditions obtained by judiciously varying and mixing the NOx partial flow rates coming from the cylinders and controlled by volumetric flowmeters.

Semi-continuous tests with recirculation of 3 10^{-4} m^3 of the scrubbing solution were conducted, allowing to follow the temporal

evolution of the absorption performances in parallel with the progressive loading of the solution with nitrogen oxides species (absorbed and hydrolyzed).

The relatively small height of the contactor and the limited number of cables (6) leads to a specific surface area of 30–35 m^{-1} (for the present liquid flow rate, but varying slightly with the physico-chemical properties of the scrubbing solutions) which explains quite low NOx absorption efficiencies illustrated here after, combined to very low NOx partial pressures investigated.

3.2. Analytical methods

Scrubbing solution were prepared by weighting HNO$_3$ and/or H$_2$O$_2$ mixed with water. The concentration of HNO$_3$ in absorbent solution was checked using the classical titration with a sodium hydroxide solution. H$_2$O$_2$ analysis was also performed by an iodometric method which involves the addition of potassium iodure KI and titration of the formed I$_2$ with a fresh sodium thiosulfate solution using of starch as an indicator.

As far as the gas phase analysis is concerned, sampling of gas simultaneously at the input and the output of the absorption column is performed continuously through membrane dryers followed by a NO−NO$_2$ analyzer (Model Rosemount XSTREAM X2GP continuous analyzer, IR type for NO and UV for NO$_2$), allowing the calculation of the oxidation ratio (OR.) characterizing the inlet gas composition and the absorption efficiency of NOx (A$_{NOx}$.):

$$OR = {(y_{NOx,in} - y_{NO,in})}/{y_{NOx,in}}. \tag{1}$$

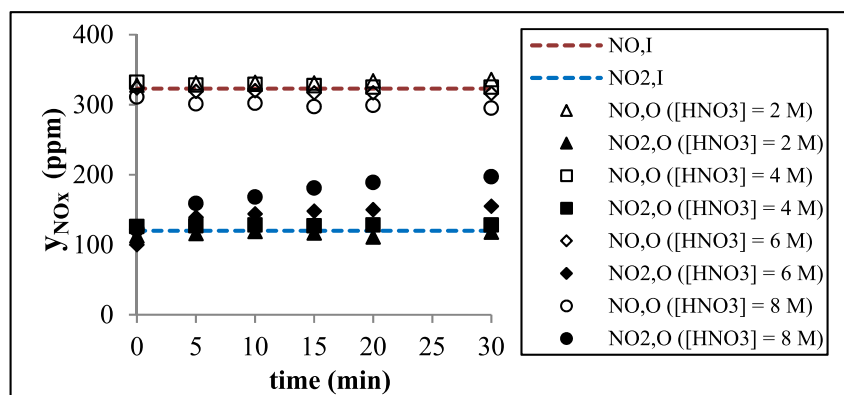$$A_{NOx} = {(y_{NOx,in} - y_{NOx,out})}/{y_{NOx,in}}. \tag{2}$$

Fig. 6. Evolution of NO and NO₂ output concentrations versus recycling time for OR = 30% for NOx absorption into nitric acid solutions.
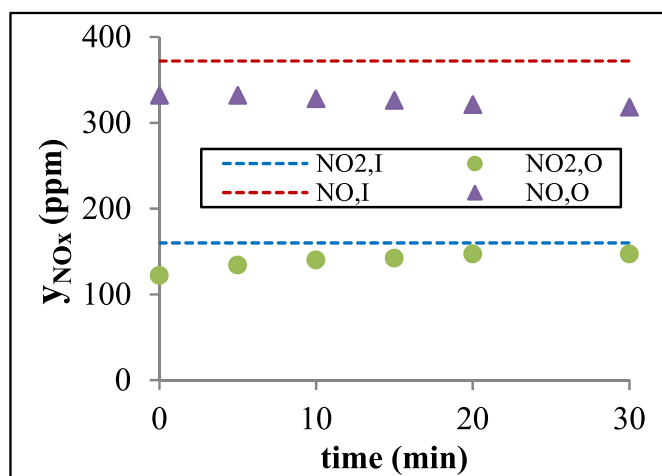

Fig. 7. Evolution of NO and NO₂ inlet and outlet concentrations over time for $[HNO_3] = 8M + [H_2O_2] = 2 M$.
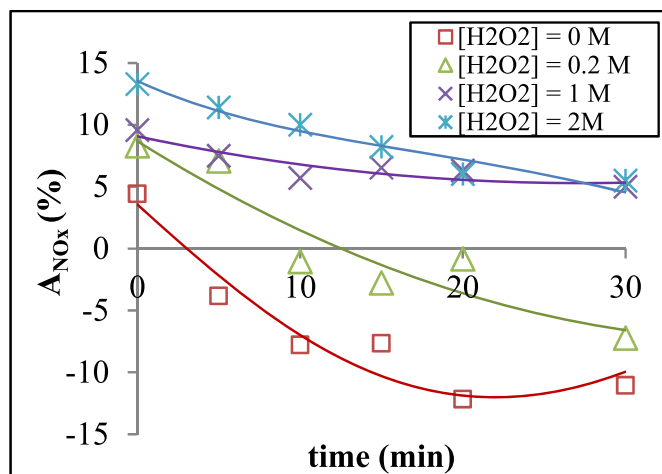

Fig. 8. Temporal evolution of NOx absorption efficiencies into nitric acid solutions (8M) containing an increasing H_2O_2 concentration.

Precisions of 1 ppm on NO and NO₂ are mentioned for our analysis ranges. This measurement accuracy leads to negligible error bars, which will be therefore not illustrated ithe following graphs.

4. Chemical reagents

The chemical products used in the experiments were:

- For the scrubbing solutions:
  - Nitric acid HNO₃ 65% weight (Chem-Lab NV)
  - Hydrogen peroxide H₂O₂ 30/35% weight (Chem-Lab NV)
  - Distilled water
- For the gas cylinders:
  - Nitrogen N₂, 99.9% pure gas under pressure (Westfallen product)
  - Nitrogen monoxide NO, 9500 ppm NO + 500 ppm NO₂ in N₂ (Westfallen product)
  - Nitrogen dioxide NO₂, about 1% in N₂ (Westfallen product)
- For the liquid phase analyses:

all reagents, sodium hydroxide NaOH (> 97%), sodium thiosulfate Na₂S₂O₃ (> 98%) and potassium iodide KI (> 99.9%) are analytical grade purchased from Chem-Lab NV.

5. Results and discussion

Prior to studying the absorption of NOx into nitric acid solutions containing or not hydrogen peroxide, we have studied the absorption of NOx into water.

5.1. NOx absorption into water for a varying oxidation ratio

Absorption tests were performed into water with the recycling of the scrubbing solution. The operating conditions applied in all experiments are indicated in Table 3.

Fig. 3 presents the evolution of absorption efficiencies (A_NOx) versus the recycling time for the different oxidation ratios.

From this figure, it can be seen that the NOx absorption efficiencies increase as the OR increases. These results can be explained by the fact that at low OR values (5%) the gaseous phase is mainly composed of NO which is a quite insoluble compound into water (De Paiva and Kachan, 1998, 2004; Hupen and Keing, 2005; Dalaouti and Seferlis, 2005; Suchak and Joshi, 1994) in accordance with Henry's constants values indicated in Table 1. Moreover, NO is quite non reactive. In contrast, A_NOx increases for OR equal to 30 and 50% because in this case, the inlet gas phase contains more soluble species such as NO₂, N₂O₃ and N₂O₄, which are more reactive with water, thus involving better absorption performances in the liquid phase.

Also, from Fig. 3, it can be seen that for the three values of OR tested, the NOx absorption efficiencies decrease gradually with the recycling of the scrubbing solution. This result can be attributed to the accumulation, into the scrubbing solution, of the soluble and hydrolyzed species which will no longer be absorbed once reaching a given level.

Evolutions of NO and NO₂ input (I) and output (O) concentrations versus recycling time are illustrated in Fig. 4 (a), (b) and (c) for the absorption into water.

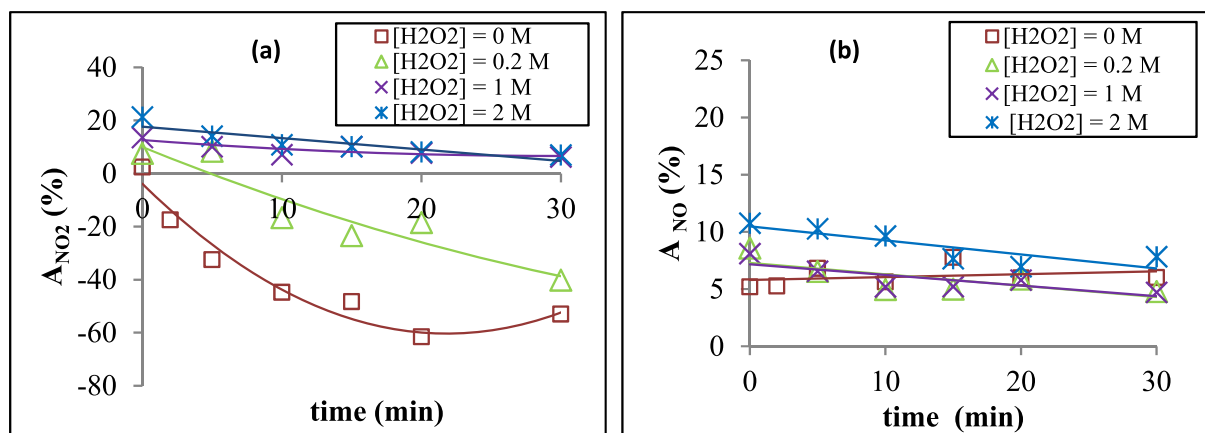From Fig. 4 (a) for OR equal to 5%, it can be seen that the NO and

Fig. 9. Temporal evolution of NO₂ **(a)** and NO **(b)** absorption efficiencies into nitric acid solutions (8M) containing an increasing H_2O_2 concentration.

NO_2 concentrations at the outlet are almost equal to those at the inlet. For OR = 30%, only NO_2 is weakly absorbed by water while NO is completely unabsorbed (Fig. 4 (b)). However, for OR = 50%, with equimolar NO and NO_2 inlet concentrations, it is observed that the NO content at the outlet is lower than the one at the inlet (Fig. 4 (c)). In view of the results obtained, we can conclude that water is quite ineffective in the removal of NOx mostly at low OR values.

5.2. NOx absorption into nitric acid solutions without H_2O_2 for OR = 30%

In the following part we will focus on an oxidation ratio equal to 30% (this was previously measured in the tail gas issued from a Tunisian nitric acid manufacturing plant (located in Gabes) using an online analyzer (Model-EL3010, IR type)) which is quite typical for NOx in waste gases produced by nitric acid plants.

In order to see what will be the effect of the oxidation reaction of NO by HNO_3 on the NOx absorption process, nitric acid solutions (2, 4, 6 and 8M) were used rather than water. The results obtained for different concentrations of HNO_3 are depicted in Fig. 5.

At the beginning of the test, the different nitric acid solutions have led to quite similar results than those obtained with water. This is due to the fact that the reaction of nitric acid with NO is very ineffective and hence NO is not efficiently absorbed. The recycling of nitric acid solution in the column has also a negative effect on NOx absorption efficiencies, leading particularly to a desorption phenomenon for $[HNO_3]$ = 8 M (already appearing for 6M), already observed in Thomas and Vanderschuren (1996). These results are due to the fact that the reaction of HNO_3 with NO gives NO_2 through reaction (R.11). From Fig. 6, we can observe that the NO_2 output concentration increases with recycling time particularly for the HNO_3 concentration equal to 8M. Similar results were obtained by Suchak et al. (1991) and Ramanand and Phaneswara Rao, 1996.

5.3. NOx absorption in a 8 M nitric acid solution containing hydrogen peroxide (OR = 30%)

Hydrogen peroxide was added to a 8 M nitric acid solution in order to enhance the absorption of NOx species. This nitric acid concentration was selected because it is produced industrially as intermediate of commercial nitric acid. Three H_2O_2 initial concentrations (0.2, 1 and 2 M) were tested and compared to the case without H_2O_2.

Fig. 7 presents the evolution of NO and NO_2 outlet concentrations when H_2O_2 is equal to 2 M whereas Fig. 8 shows the evolution of NOx absorption efficiencies versus recycling time, for three initial H_2O_2 concentrations.

As clearly illustrated by Fig. 8 and whatever the recycling time is, $\mathbf{A_{NOx}}$. increases sharply when hydrogen peroxide is gradually added to

the nitric acid solution because H_2O_2 can oxidize HNO_2 to HNO_3 (R.9) and can therefore avoid its decomposition. In fact, HNO_3 reacts with NO to produce HNO_2 which in turn is oxidized by H_2O_2 to form HNO_3 as already demonstrated by Suchak and Joshi (1994), De Paiva and Kachan (1998), Thomas and Vanderschuren (2000) and Liémans and Thomas (2010). Since the oxidation reactions are very fast and irreversible, the liquid phase mass transfer resistance decreases (due to an enhancement factor relative to the reactive absorption) which induces a higher NOx absorption efficiency. Furthermore, the oxidation reaction of HNO_2 is enhanced by the auto-catalytic effect of H^+ (Liémans et al., 2011). The increase of A_{NOx} with $[H_2O_2]$ is attributed mainly to the higher absorption of NO_2 in the presence of H_2O_2 such as illustrated by Fig. 9(a). The effect of H_2O_2 on the absorption of NO is less pronounced as demonstrated by Fig. 9(b). From these figures, it is clear that $\mathbf{A_{NOx}}$ decreases with the recycling of the scrubbing solution at all the initial H_2O_2 concentrations, leading to a desorption phenomenon for 0.2 M in H_2O_2. All these results are attributed to the gradual consumption of H_2O_2 during the recycling time, which is critical in the case of low initial H_2O_2 concentrations.

Ican be concluded from this part that an appreciable advantage of the use of H_2O_2 for NOx absorption is the possibility to carry out the gas treatment while producing rather concentrated nitric acid solutions (here 8 M).

6. Conclusions

The main purpose of this study was to investigate the absorption of NOx in a gaseous effluent under a partial pressure equal to 50 Pa, tested in a cables-bundle contactor, using nitric acid (0–8 M) scrubbing solutions containing or not hydrogen peroxide (0–2 M).

The effects on NOx absorption efficiencies of the oxidation ratio, the initial concentration of H_2O_2 and HNO_3 into the scrubbing solution as well as the recycling of the scrubbing solution were studied under ambient conditions (293 K and 1 atm). The main results of our study can be summarized as follows:

- the oxidation ratio has a significant positive influence on NOx absorption efficiencies which increase when OR is varied from 5 to 50%;
- nitric acid solutions without H_2O_2 as absorbent have a negative effect on the NOx absorption efficiency. This effect increases with a HNO_3 initial concentration increasing;
- the addition of the hydrogen peroxide to the nitric acid solution improves the NOx absorption efficiency because H_2O_2 is able to oxidize HNO_2 to HNO_3 and thereby enhance the NOx mass transfer, especially with higher HNO_3 concentrations as H^+ ions are known to catalyze the liquid oxidation reaction;

- in all cases, the NOx absorption efficiencies decrease with the recycling of the scrubbing solution.

As perspective of this work, we suggest to study the continuous addition of H_2O_2 to the scrubbing solution, whose the concentration must be kept constant and sufficient in order to continuously ensure the oxidation of HNO_2 to HNO_3. All these elements will be essential to help to design a new full-scale scrubbing system for the absorption of NOx released from a Tunisian nitric acid plant. Actually a packed column operating with the NOx-HNO_3-H_2O_2 chemical system could be used to comply with an environmental legislation once properly designed.

Acknowledgment

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