

Optimization of the operation conditions for NiO dissolution with different leachants

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ABSTRACT

In this study, the leaching of NiO from NiO/ α -Al₂O₃ catalyst in acidic (HCl, HNO₃ and H₂SO₄) and ammoniacal ((NH₄)₂CO₃ and CH₃COONH₄) media was investigated. The effects of leachant concentration, liquid/solid ratio, stirring speed and temperature were studied. It was found that 100% of nickel was dissolved after 30 min of reaction with HCl at 2M, 80°C and liquid/solid ratio of 50 mL/g, while HNO₃ and H₂SO₄ dissolved 77.15 and 46.12%, respectively, under the same operation conditions. Mixing two strong acids led to a synergetic effect on nickel leaching at the beginning of the reactions followed by a rapid stabilization in dissolution. Ammoniacal leaching was less efficient than acidic one registering 41.43% with ammonium carbonate and 29.16% with ammonium acetate after 180 min. However, the addition of chloride to ammonium carbonate led to totally dissolve NiO.

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1. Introduction

Soils are the major sink for heavy metals released into the environment. Heavy metals are non-renewable resources and do not undergo microbial or chemical degradation. Their total concentration in soils increase due to anthropogenic activities and persists for a long time after their introduction which may pose risks and hazards to humans and ecosystems through direct ingestion or contact with contaminated soil, the food chain and drinking of contaminated ground water.

Some of these activities include mining and smelting of metals, burning of fossil fuels, production of batteries and industrial wastes disposal. The increasing demand for metals in the world has required intensive studies for the extraction of metals from low-grade ores and/or secondary resources. Nickel products are known for superior resistance to both heat and corrosion as well as low thermal expansion properties. These properties make it useful in industries that require stability as well as the ability to resist corrosion over a wide range of temperatures. Nickel in the form of oxide (NiO) generally supported on alumina or silica is used in chemical processing as catalyst in hydrogenation,

hydrodesulphurization, steam reforming of methane and oxidation of carbon monoxide [1-5].

Once the catalyst is deactivated the product becomes an important source of environmental contamination that needs to be treated. During the last decades, Ni has become a serious concern as its concentration has reached up to 26,000 mg/kg in polluted soils and 0.2 mg/L in polluted surface waters, which is 20 and 30 times higher than found in unpolluted areas [6]. Extraction techniques for the removal and recovery of nickel are of increasing interest because of the growing importance given to environmental protection problems.

Methods for processing nickel from secondary resources include high temperature pressure acid leaching [7,8] and atmospheric acid leaching [9,10].

High pressure leaching is considered as the most efficient treatment but the process requires drastic conditions (high temperature and pressure), higher capital cost and has problems with material of construction at commercial level [11]. Leaching at atmospheric pressure has been recently receiving more attention despite its less efficiency, longer reaction time and higher acid consumption. In this framework, conventionally, sulfuric acid is used as leaching reagent for the recovery of nickel from catalysts [12-15] because of its lower corrosion property and lower cost. In fact, Ivascanu and Roman [16] recovered 99% of Ni from a spent catalyst

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in the form of nickel sulfate by leaching with sulfuric acid under the following conditions (sulfuric acid concentration of 80%, 50 min of reaction and 70°C). Al-Mansi and Abdel Monem [17] recovered 99% of nickel from Egyptian spent catalysts under the following experimental parameters (50% sulfuric acid concentration, a solid-to-liquid ratio of 1:12 g/g and a contact time over 5 h). Abdel-Aal and Rashad [14] recovered 94% of nickel after leaching spent nickel oxide catalysts with sulfuric acid under the conditions (-200 + 270 mesh spent particle size, 85°C, 150 min, sulfuric acid concentration of 50% and solid/liquid ratio of 1:20 g/mL).

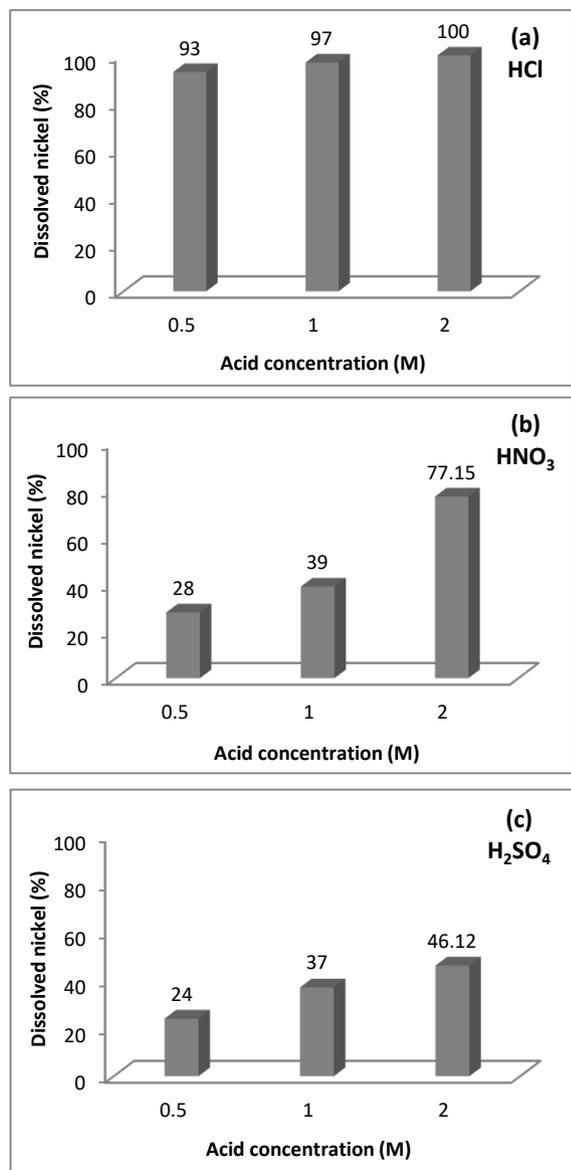


Fig. 1 - Effect of acid concentration on nickel dissolution with the three acids after 30 min of reaction.

Thus, most existing hydrometallurgical processes for the leaching of nickel employ sulfuric acid. Alternative technologies that utilize other leaching agents such as hydrochloric acid, nitric acid, alkaline reagents and various organic acids offer potential for

improved nickel processing technologies and are currently being examined and developed. In fact, Astuti et al. [18] reported leaching experiments on nickel extraction from low grade Indonesian saprolitic ores by using several lixiviants (sulfuric, nitric, chlorhydric, citric, lactic and oxalic acids). They found that citric and sulfuric acids were the most effective leachants while oxalic acid was the weaker one. Chaudhary et al. [19] reported hydrochloric acid leaching process for the recovery of nickel as nickel oxide from a spent catalyst containing 17.7% Ni. They found that maximum of nickel extraction of 73% could be achieved by carrying out the leaching process with 28.8% HCl at 80°C. Xian et al. [20] extracted 95.8% of Mo and 91.3% of Ni from a roasted Mo-Ni ore by using the following optimum conditions (hydrochloric acid at 0.219M, 65°C, a liquid-to-solid ratio of 3 mL/g and 30 min) followed by sulphation roasting (51.9% sulfuric acid, roasting temperature 240°C for 1h) and leaching with HCl at 95°C for 2h.

Kinetic data for pure minerals are important as the data may then be used to evaluate leaching of more complex systems containing several mineral constituents such as in ores or spent catalysts. The work described in this paper is an attempt to optimize a leaching process to recover nickel from NiO/ α -Al₂O₃ fresh catalyst by testing 5 reagents under the same conditions: three inorganic acids (HCl, HNO₃ and H₂SO₄) and two alkaline solutions ((NH₄)₂CO₃ and CH₃COONH₄). To the best of our knowledge, no comparative studies were found dealing with the leaching of nickel oxide with the selected solutions. The aim was to address the factors governing the leaching process and to determine the most efficient and/or an alternative leachant for this system.

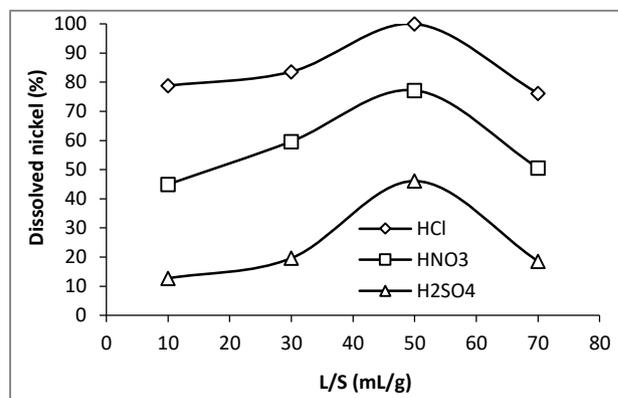


Fig. 2 - Effect of liquid/solid ratio on nickel dissolution with the three acids after 30 min of reaction.

2. Materials and methods

The catalyst used in this study was a 10% NiO/ α -Al₂O₃ (n/n) prepared using the incipient wetness method. The catalyst was calcined in an oven at 500°C for 2 h and the nickel composition was checked by atomic absorption. H₂SO₄ (96-98%, Biochem), HNO₃ (60%, Cheminova), HCl (37%, Carlo Erba), (NH₄)₂CO₃ (99.5%, Biochem) and CH₃COONH₄ (>97%, Fluka) were used as leaching reagents. Leaching experiments were conducted in a spherical glass batch reactor of 100 mL magnetically stirred and heated by a temperature-controlled water bath and equipped with a return flow cooler (glass condenser) to minimize water losses due to

evaporation. A typical experiment was conducted as follows: 0.1 g of the sample was placed into the glass reactor with a desired volume of the selected leachant. After the leaching process, the reaction mixture was filtered and the Ni²⁺ species was analyzed by titration with ethylenediamine tetraacetic acid disodium salt (EDTA), using murexide as an indicator. The data presented are an average of three test replicates with an error of 5%. The percentage of dissolution was calculated from the following equation:

$$\frac{\text{Mol number of nickel in the solution}}{\text{Mol number of nickel in the catalyst}} \times 100 \quad (1)$$

The process conditions studied included the nature of the leachants, their concentration, liquid/solid ratio, stirring speed, temperature, mixture of acids, the addition of anions and time.

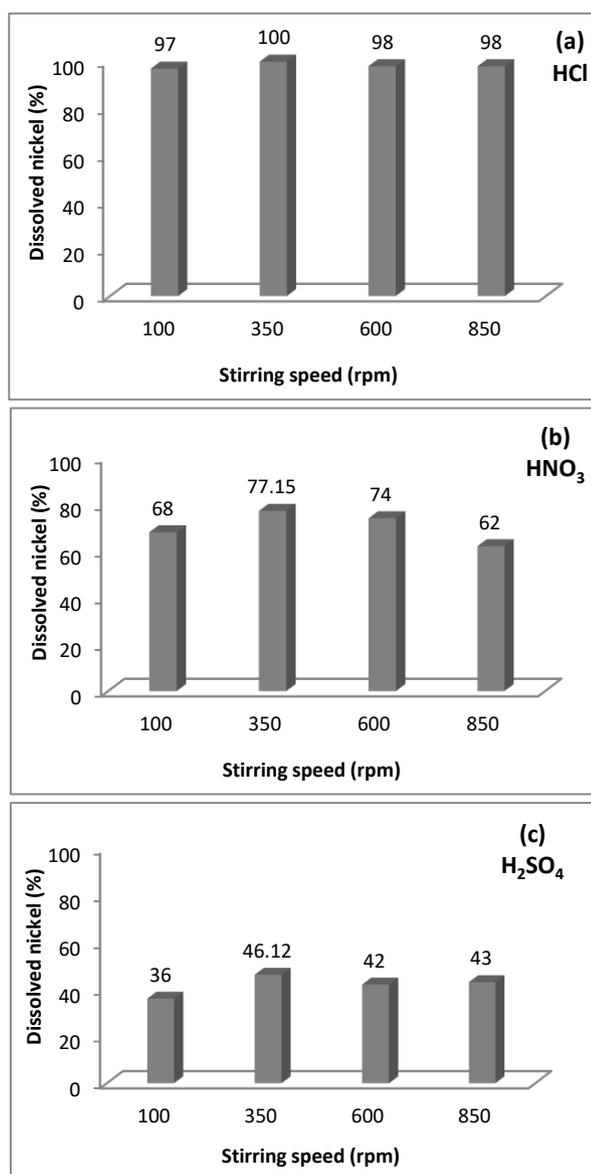
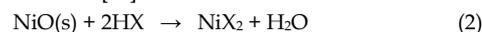


Fig. 3 - Effect of stirring speed on nickel dissolution with the three acids after 30min of reaction.

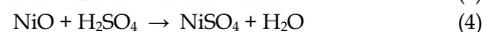
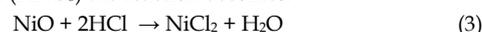
3. Results and discussion

3.1. Effect of acid concentration

Nickel oxide may be dissolved in inorganic acids according to the general reaction [21]:



In the case of hydrochloric acid (HCl), sulfuric acid (H₂SO₄), or nitric acid (HNO₃) the reaction becomes:



To determine whether nickel oxide dissolved easily in acid solution or not, a preliminary test was conducted with HCl solution at 2M, ambient temperature, L/S = 50 mL/g, and without agitation. No dissolution of nickel oxide was observed after two days. This result indicated that nickel oxide belongs to hard dissolving compounds. High temperature (80°C) and an agitation of 350 rpm were applied to decrease the dissolution time. The effect of acid concentration was investigated by testing three values (0.5, 1 and 2M) at 80°C, L/S = 50mL/g and 350 rpm with the three acids.

The results (Fig. 1(a-c)) show that the fraction of the dissolved nickel increased with increasing acid concentration. It reached 93, 28 and 24% at 0.5 M and 100, 77.15 and 46.12% at 2M with HCl, HNO₃ and H₂SO₄, respectively, after 30min. The concentration of 2M was used to investigate the effect of the other parameters.

The dissolution of Alumina was checked with the three acids at different concentrations. Alpha alumina was found to be completely inert toward acids because of its great stability [17]. This was observed in our previous studies [22-24].

3.2. Effect of liquid/solid ratio

Different L/S ratios were tested (10, 30, 50 and 70mL/g) at 2M, 80°C, 350rpm at a duration of 30min of reaction. The results (Fig. 2) show that a maximum of dissolution efficiency was obtained with L/S value of 50mL/g. At lower or higher values than 50mL/g the dissolution of NiO was less efficient. The value of 50mL/g was used to investigate the rest of the parameters.

3.3. Effect of stirring speed

The effect of stirring speed on the dissolution rate of nickel was investigated using different agitation speeds (100-350-600 and 850rpm) at 80°C, L/S=50mL/g and at acid concentration of 2M (Fig. 3a-c). It can be seen that agitation speed had almost no remarkable effect on NiO dissolution since the values of NiO leaching varied very slightly with the three acids indicating that the diffusion did not control the reaction in these cases. A slight improvement in nickel dissolution was however observed when 350rpm was used.

3.4. Effect of temperature

The effect of temperature on the rate of nickel dissolution was investigated by testing different values: 50, 60, 70 and 80°C at acid concentration of 2M, stirring speed of 350 rpm, and L/S = 50 mL/g. The results (Fig. 4(a-c)) show that the dissolution of NiO with the three acids was highly temperature-dependent since the time

needed to reach high dissolution efficiency was decreased with increasing temperature. In the case of HCl, the dissolution efficiency increased from 58.6% at 50°C to 100% at 80°C after 30 min of reaction. With HNO₃ and H₂SO₄, 8.27% and 7.92% of NiO were dissolved, respectively, at 50°C after 30 min of reaction and increased up to 77.15% and 46.12%, respectively, after the same period of time at 80°C. Even after prolonging reaction time, nitric and sulfuric acids did not reach the dissolution efficiency of HCl since 77.3% with HNO₃ and 72.8% with H₂SO₄ were registered as maximum at 80°C after 180 min of reaction.

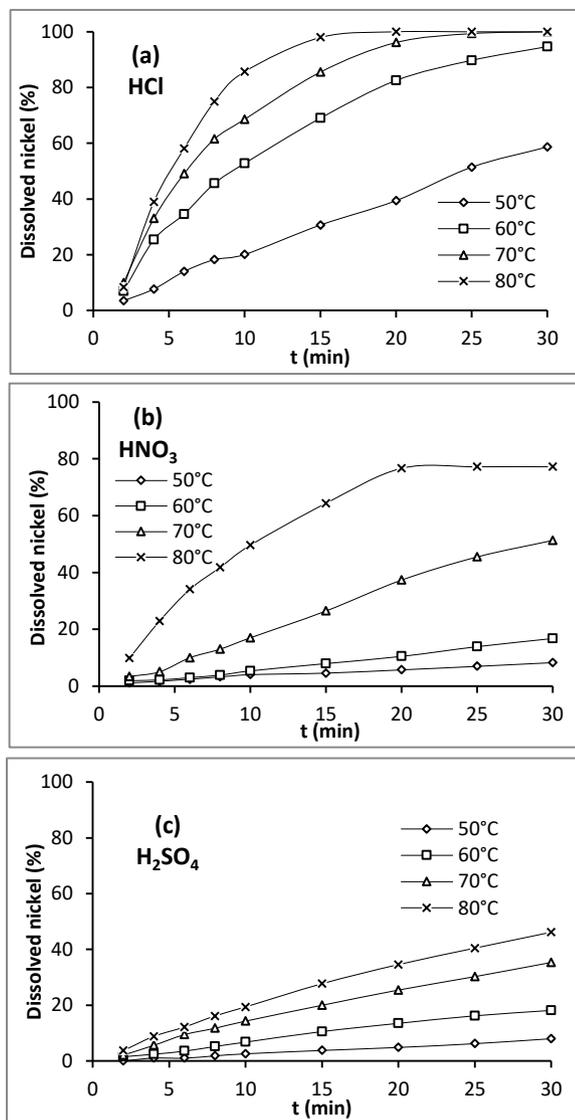


Fig. 4 - Effect of temperature on nickel dissolution with the three acids.

With HCl and HNO₃ a stabilization in the dissolution values occurred after 20 min at 80°C. For HCl, this can be simply explained by the fact that at 20 min the system reached already total dissolution. In the case of HNO₃, the dissolution attained 75.6% after 15 min and slightly increased to 77.15% after 30 min and the values remained so up to 180 min without reaching total dissolution. A

clear difference is observed between the results obtained with the three leachants. This is probably related to the nature of the anion of each acid.

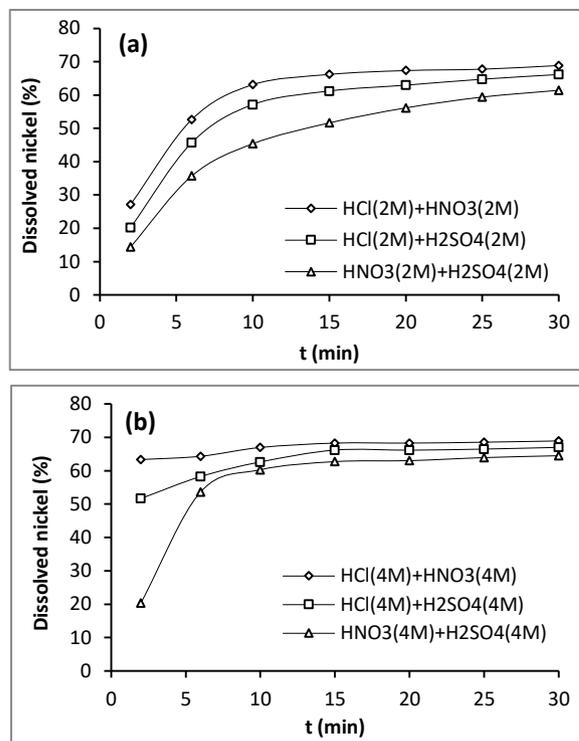


Fig. 5 - Effect of the mixture of acids at 2M (a) and 4M (b) on nickel dissolution.

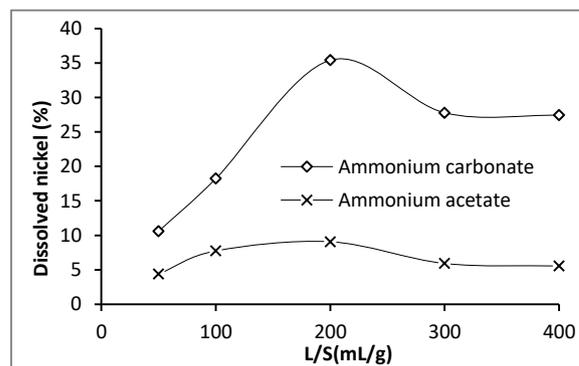


Fig. 6 - Effect of L/S ratio with ammonium carbonate and acetate after 60 min.

In the literature, there is an increasing interest on the effect of anions on the dissolution of minerals [25]. In fact, it was reported that the aggressivity of chlorides in metal oxides dissolution was due to the formation of chloro-complexes as adsorbed species with high value of adsorption constant that leads to enhance the metal oxide dissolution [21]. In the case of nitric acid, nitrates and protons played an inverse role leading to a competing process between oxide growth and metal dissolution. It was reported that increasing the

concentration of nitric acid may form a passive film that protects metal surface from protons attack while in diluted nitric acid nitrate ion may play the role of corrosion promoter [26]. Our experiments were carried out under concentrated conditions and it seems from the results that the building of passive film was more rapid than the rate of its dissolution. Sulfuric acid was the weakest leachant regardless of the conditions used. In this case, also sulfate anion seems to be the cause of the weak efficiency of H_2SO_4 . It was reported that sulfate ions because they are doubly charged have a strong adsorptive affinity for solid surface hampering consequently its dissolution [27].

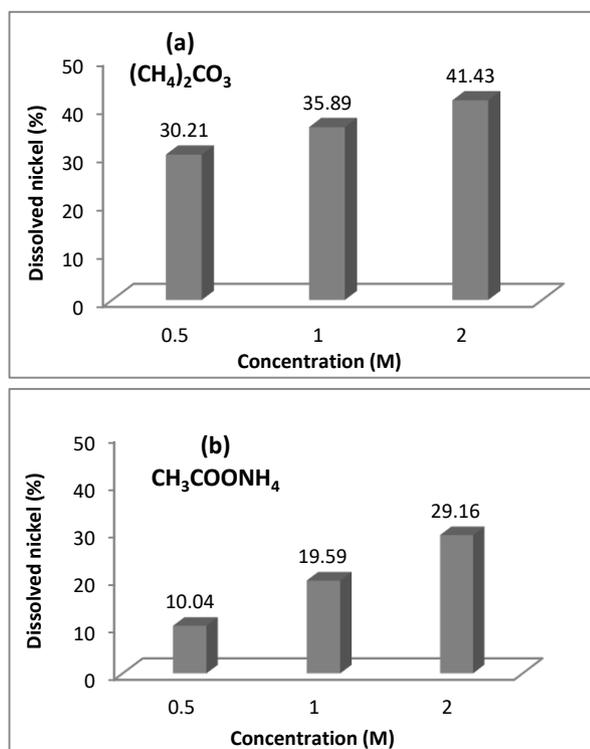


Fig. 7 - Effect of ammonium carbonate (a) and ammonium acetate (b) concentration on NiO leaching after 180 min of reaction.

3.5. Effect of the mixture of acids

The results have shown that HCl was the unique acid which could dissolve 100% of NiO. HNO_3 and H_2SO_4 were less efficient. It is an interesting task to investigate whether the efficiencies of the latest acids may be improved when they are mixed. For this purpose, three mixtures ($\text{HNO}_3 + \text{H}_2\text{SO}_4$), ($\text{HNO}_3 + \text{HCl}$) and ($\text{H}_2\text{SO}_4 + \text{HCl}$) were prepared for which two concentrations of acids were used (2 M and 4 M) thereby the final concentration of each acid in the mixtures was 1 M and 2 M, respectively. The experiments were carried out at 80°C , 350 rpm, and $L/S = 50\text{mL/g}$. The results (Fig. 5(a,b)) show a remarkable improvement in dissolution rate as the reactions started. In fact, when HCl, HNO_3 and H_2SO_4 were used alone at 2 M, they dissolved 8.3, 4.8 and 3% of NiO, respectively, after 2 min of reaction. It increased up to 27.2, 20.2 and 14.4% for the mixtures ($\text{HNO}_3 + \text{HCl}$), ($\text{H}_2\text{SO}_4 + \text{HCl}$) and ($\text{HNO}_3 + \text{H}_2\text{SO}_4$), respectively, after the same period of time. After about 10 min, the

reactions stabilized. The dissolution efficiencies of the three mixtures were again improved when the concentration of each acid used in the mixtures was increased to 4 M giving rise to 63.36, 51.65 and 20.4% of nickel dissolved by the same mixtures cited before, respectively, after 2 min. In this case, the dissolution stabilized after 5 min. Thus, there was a synergetic effect in both cases since the dissolution efficiencies of the mixtures were higher than the sum of acid efficiencies. However, it seems that this synergetic effect was observable only few minutes after the beginning of the reactions since the dissolution stabilized rapidly thereafter at around 61–68% for both cases which is lower than that obtained with HCl (100%) and HNO_3 (77.13%) alone. This stabilization may indicate the interruption of the dissolution reactions.

This interruption may be explained by the presence of high amount of ions that may be adsorbed on the solid surface preventing further dissolution. On the other hand, it can be observed that the highest dissolution efficiency was registered with the mixture ($\text{HNO}_3 + \text{HCl}$), while the lowest one was registered with ($\text{HNO}_3 + \text{H}_2\text{SO}_4$) with both concentrations. Thus, the mixtures containing H_2SO_4 were less efficient. This is another indication that a competitive adsorption of sulfate occurred leading to decrease protons, chlorides and nitrates attacks.

3.6. Effect of the operation conditions on NiO leaching in ammoniacal solutions

Ammoniacal leaching is a promising method for the recovery of valuable metals (such as nickel, copper and zinc) from low-grade oxide resources [28], lean complex ores [29], ocean nodules [30] and various secondary resources [31]. Although it is usually considered that the metal leaching in ammoniacal system is less effective comparing with acidic leaching, ammonia leaching has already been applied to metal extraction from low-grade ores and electronic waste and is gradually expanding [32,33]. The advantages of ammonia leaching lie in its leaching selectivity towards Cu, Zn, Ni, Co etc., because of the formation of stable metal ammine complexes [34,35], its mild alkaline characteristics, ease of use, and potential ability to be recycled [34]. Furthermore, aqueous ammonia as a leaching agent is fairly non-corrosive and leaching reactions occur in relatively mild conditions [36]. Moreover, undesirable metals such as Fe, Mg, Ca and Mn precipitate as hydroxides because of their poor complexation ability with ammonia. In this study, two ammonium products were tested for nickel dissolution: ammonium carbonate (Scharlau, 100%) and ammonium acetate (Prolabo, 98%). Both products form stable and soluble complexes with nickel.

Nickel leaching from the catalyst was tested under the same operation conditions applied for mineral acids namely ($L/S=50\text{mL/g}$, 350rpm, 80°C and 2M). The pH_0 of ammonium carbonate was 7.9 while that of ammonium acetate was 8.46. The results show that the dissolution was very weak in the case of ammonium carbonate (2%) and absent (0%) with ammonium acetate after 120min. The ratio L/S was then increased to (70, 100, 150, 200 and 400mL/g) and the dissolution efficiency was measured after 60min. The other parameters were maintained constant. The results show an improvement in the dissolution efficiency of NiO up to 200mL/g registering 35.4% and 9.1% with ammonium carbonate and ammonium acetate respectively after which it decreased in both cases at higher L/S values (Fig. 6).

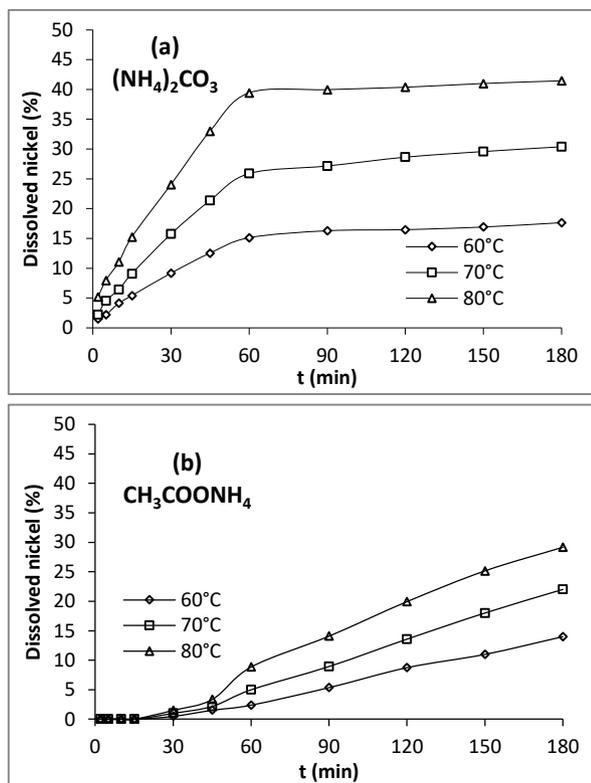


Fig. 8 - Effect of temperature on NiO leaching with ammoniacal solutions.

The effect of the concentration (0.5, 1 and 2 M) of both solutions was carried out with $L/S = 200\text{mL/g}$ at 80°C , 350rpm and the reaction time was prolonged to 180 min. The results show that ammonium carbonate dissolved more rapidly NiO than ammonium acetate but its efficiency remained weak compared to mineral acids (Fig. 7(a,b)).

The effect of temperature was tested in the range $60\text{--}80^\circ\text{C}$. The results show that the dissolution was improved by increasing temperature in both cases (Fig. 8(a, b)). In fact, it registered 17.65, 30.39 and 41.43%, with ammonium carbonate and 14, 22 and 29.16% with ammonium acetate at 60, 70 and 80°C , respectively, after 180 min. In the case of ammonium acetate, an ignition period of 15 min was observed in which no dissolution occurred followed by a second period from 15 to 45 min in which the dissolution slightly increased after which the effect of temperature was clearly observed as shown in Fig. 8(b). With ammonium carbonate, two stages of reaction are clearly observed; an initial stage up to 60 min where the dissolution of NiO proceeds rapidly and a second stage from 60 min up to 180 min where the dissolution strongly decreased. The presence of these stages may indicate a change in the mechanism of dissolution.

In the first part of this study, the results have shown that the dissolution of nickel with the three mineral acids was different under the same operation conditions and this was related to the nature of the anion that seems to play an important role on the dissolution process. To highlight the effect of the three anions, NaCl (99.5 %, Biochem), NaNO_3 (99 %, Fluka) and Na_2SO_4 (99 %, Sigma-Aldrich) were added at the concentration of 0.5 M to ammonium

carbonate and acetate under the same conditions (2 M of ammoniacal solution, 350 rpm, 80°C , $L/S = 200\text{ mL/g}$).

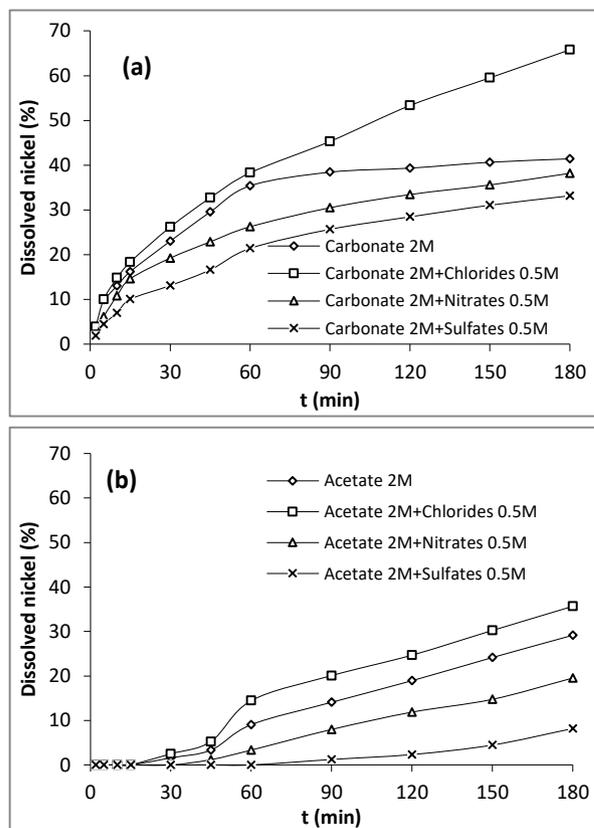


Fig. 9 - Effect of the addition of anions to ammonium carbonate (a) and ammonium acetate (b) on the dissolution of NiO.

The results (Fig. 9(a, b)) show that only the addition of chloride enhanced the dissolution process of NiO with both ammoniacal solutions. It registered 65.78% with ammonium carbonate and 35.67% with ammonium acetate after 180 min, while alone both solutions registered 41.43 and 29.16%, respectively, after the same period of time. In the presence of nitrate and sulfate, the dissolution was lower than that attained by the ammoniacal solutions alone indicating an inhibiting effect of both anions under the conditions used in this study.

Since the results showed an enhancement in nickel dissolution with chloride, other experiments were carried out in which the concentration of NaCl was increased up to 2 M. The solutions were prepared such as the mixtures contained 0.5, 1 and 2 M of NaCl and 2 M of each ammoniacal solution. The experiments were carried out at $L/S = 200\text{ mL/g}$, 350 rpm and 80°C . A remarkable improvement in NiO dissolution was observed with ammonium carbonate (Fig. 10(a)) where 65.78, 80.38 and 100% of NiO were dissolved after 180 min when NaCl at 0.5, 1 and 2 M, respectively, was used. The addition of chloride to ammonium acetate under the same conditions slightly improved the dissolution efficiency on the contrary of ammonium carbonate (Fig. 10(b)). It attained 35.67, 42.87 and 51.76% after 180 min of reaction. Ammonium carbonate seems to have strong complexing ability with respect to nickel under the conditions used in this study on the contrary of ammonium acetate.

It is likely that carbonate anion plays a role in this result as with anions in inorganic acids.

slopes of the straight lines the apparent rate constants, k , were evaluated.

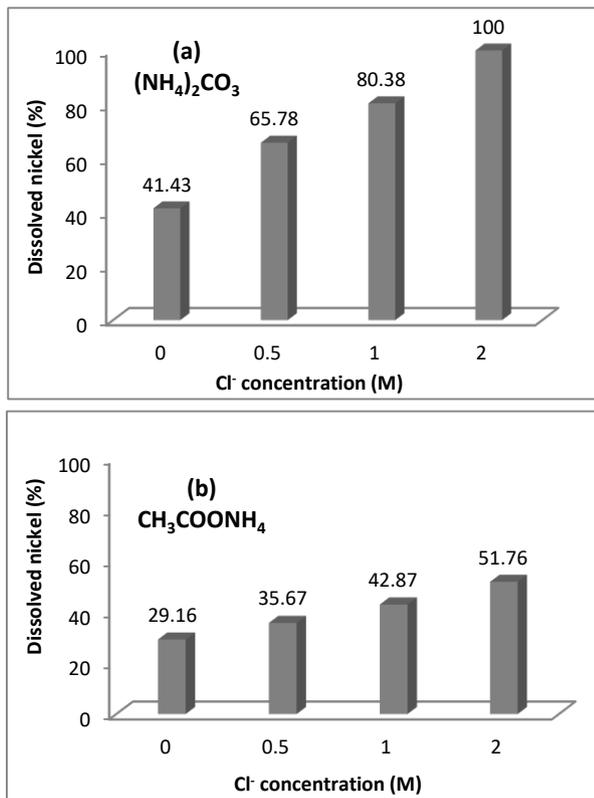


Fig. 10 - Effect of chlorides concentration on NiO leaching by ammonium carbonate (a) and acetate (b) after 180 min.

3.7. Kinetics study

Determining the kinetic aspect of any chemical reaction is an essential part in designing a chemical process and this is true for leaching systems as well as for the nickel leaching from NiO/ α -Al₂O₃ catalyst. The shrinking core model is the best simple representation for the majority of reacting fluid-solid systems [37]. This model was used for the kinetics investigation in this study.

In this model, the overall leaching process may be controlled by intrinsic chemical reaction or by external mass transfer. The following expressions can be used to describe the dissolution kinetics of the process:

For liquid film diffusion control:

$$x = kt \tag{6}$$

For film diffusion control through product layer:

$$1 - 3(1 - x)^{2/3} + 2(1 - x) = kt \tag{7}$$

For surface chemical reaction control:

$$1 - (1 - x)^{1/3} = kt \tag{8}$$

Where x is the fractional conversion of nickel at time t and k is the apparent rate constant (min⁻¹). The overall rate of dissolution is controlled by the slowest of these sequential steps.

Based on the experimental data of figures (4 (a-c)) and (8(a,b)) plots corresponding to equation (8) show good linear correlations for the dissolution of nickel with all leachants (Fig. 11(a-e)). From the

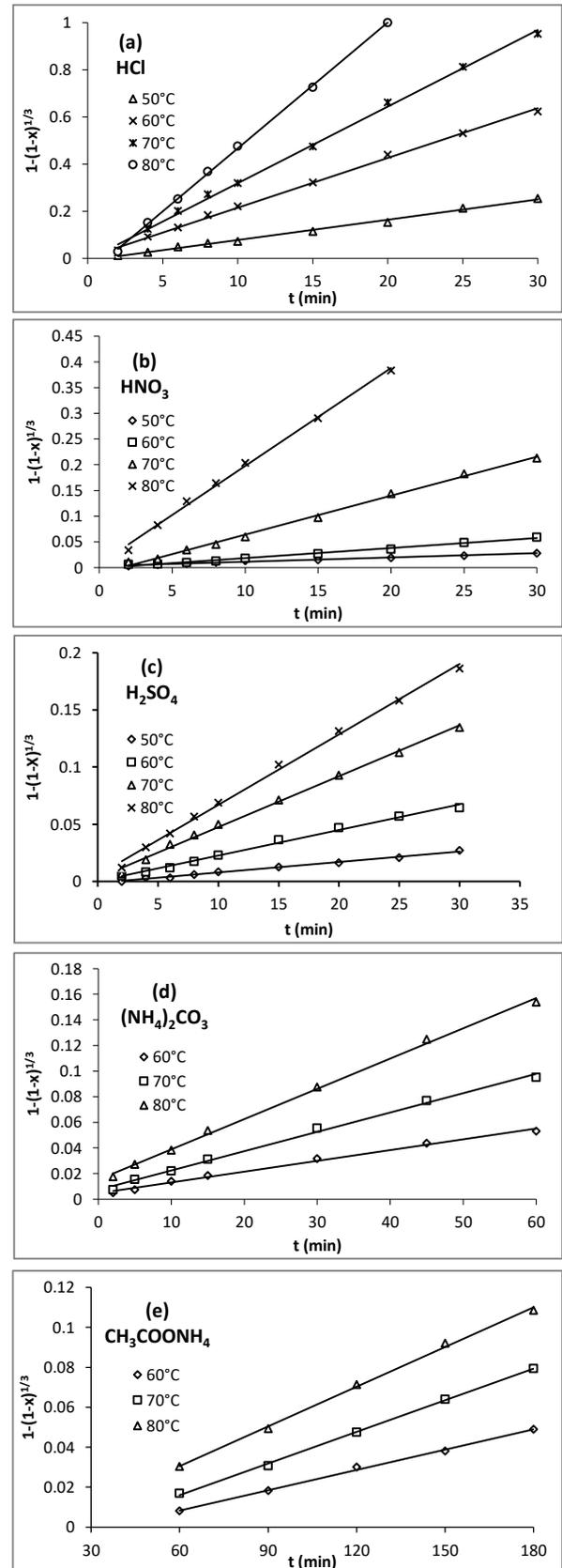


Fig. 11 - Fit of Shrinking core model to the experimental data.

Arrhenius equation $k = A e^{-E_a/RT}$ was plotted as $\ln k$ vs. $1/T$ for each temperature and the activation energies were calculated from the slopes $-E_a/R$. The following values of E_a were determined: $E_a(\text{HCl}) = 56.23$, $E_a(\text{HNO}_3) = 102.68$, $E_a(\text{H}_2\text{SO}_4) = 61.73$, $E_a(\text{carbonate}) = 53.47$ and $E_a(\text{acetate}) = 41.27$ kJ/mol.

A chemically controlled reaction is generally a temperature dependent process and has an activation energy higher than 40 kJ/mol. The activation energies were all higher than 40 kJ/mol but the values varied from 41 to 103 kJ/mol indicating different degrees in temperature dependence. In fact, the highest activation energy was that obtained with nitric acid (102.68 kJ/mol). The effect of temperature in this case was stronger than with the other leachants. As a comparison, increasing temperature from 50 to 80°C increased the rate constant by 23.75 times with HNO_3 (from 0.0008 min^{-1} at 50°C to 0.019 min^{-1} at 80°C) while it increased by 7 times with H_2SO_4 , 6.2 times with HCl, 3 times with ammonium carbonate and 2.33 times with ammonium acetate. It is not possible to give a rigorous explanation for the results obtained in this study because of the complexity of the heterogeneous systems where all parameters are dependent from each others, but what was observed is that HCl was the most effective leachant in NiO dissolution.

4. Conclusion

In this study, it was found that the conditions: 50 mL/g, 80°C, 2 M and 350 rpm were appropriate to dissolve 100, 77.15 and 46.12% of NiO with HCl, HNO_3 and H_2SO_4 , respectively, after 30 min. Acid leaching gave different results depending on the nature of the leachant indicating that the anion played an important role on accelerating or inhibiting the dissolution process.

Mixing the acids led to a synergetic effect registered after the first few minutes but the reactions stopped rapidly probably because of the presence of high amounts of ionic species that may sterically protect the solid surface against protons attack.

The use of ammoniacal leaching gave less good results than mineral acid leaching since a maximum of 41.43 and 29.16% of nickel were dissolved by ammonium carbonate and ammonium acetate, respectively, after 180 min at 80°C and 200 mL/g. Among the five leachants tested, HCl was the most efficient in nickel dissolution. However, HCl is a non-selective lixiviant and may cause the formation of sludge in the case of treatment of spent catalysts because of the dissolution of impurities that require post treatments. An alternative leachant could be a combination of ammonium carbonate and chloride ions to reach the same efficiency as that of HCl while being selective.

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