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Dissolution Kinetics of Metallic Silver using Ammonium Nitrate and Hydrogen Peroxide Solution as an Environmentally Friendly Cyanide Substitute

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Abstract

This article describes the kinetics of utilizing ammonium nitrate to dissolve pure metallic silver in hydrogen peroxide solution (H2O2). Using pure metallic silver allows for precise leaching kinetics research by removing interference from impurities and facilitating accurate interpretation of rate-controlling mechanisms. The impact of temperature, rotation speed, H2O2 concentration, and ammonium nitrate concentration were all examined. The results show a favourable relationship between the rate of silver (Ag) dissolution and the rotation speed. Additionally, a low concentration of ammonium nitrate (between 0.003 and 0.20 M) has advantageous effects on Ag dissolution. The dissolution rate was significantly impacted by H2O2 concentrations between 0.08 and 0.15 M, because this range of H_2O_2 concentration required to provide sufficient oxidative potential for significant silver solubility. However, this effect is less pronounced in the 0.20–0.50 M range. 20 - 50 °C range of temperatures are advantageous since H2O2 is stable in this range. It was calculated that the activation energy was 25.66 kJ/mol.

1. Introduction

Silver (Ag) is an essential metal that is used in jewelry, dental amalgam, electrical and electronic equipment, photographic materials, catalysts, and brazing alloys [1]. The cyanidation process is the most widely used extraction method for gold and silver extraction at the moment [2].

Cyanide is highly toxic and can harm both human health and the environment, despite its many benefits [3]. In order to identify substitutes that are less detrimental to the environment and public health, considerable investigations have been conducted [4]. Many hydrometallurgical methods have recently been developed to recover silver without the use of cyanide. Many studies have concentrated on the thiourea [5, 6], thiocyanate [7-9], halides [10–12], and thiosulfate techniques [13–16].

Thiourea (NH_2CSNH_2) is a less hazardous chemical for both humans and the environment, which is the reason it was selected to be researched as one of the cyanide substitutes. When using thiourea leaching, acidic conditions are usually employed. When using acidic thiourea as a leaching reagent, a number of problems occur such as high thiourea consumption when compared to cyanide, challenging thiourea lixiviant regeneration and substantial equipment corrosion in acidic solutions compared to steel passivation in alkaline cyanide solutions [17]. These problems

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have prevented thiourea from being used in industry.

Thiocyanate (SCN^-) is also used as an alternative for cyanide to extract silver and gold. Thiocyanate produced when sulfur species react with cyanide. Thiocyanate solutions are less toxic and more stable than potassium and sodium cyanide. It is also renewable and accessible as a result of technological processes.

Moreover, it has been found that the halides are efficient agents for leaching gold and silver in water-based solutions. Typically, the halogen species functions as both a ligand and an oxidant in halide processes. The pH, solution potential, presence of reductants, and halide concentration all have a substantial impact on the stability of the halide complexes during the leaching process with halide ions [18]. For the halide systems to reach the high oxidation potentials needed for leaching, an oxidant such as bromine (Br_2) or iodine (I_2) must be used, but both are toxic [19].

Thiosulfate $(S_2O_3)^{2-}$ is another alternative to cyanide that is less dangerous. Up to 12 g of thiosulfate can be taken orally every day without risk [19]. Thiosulfate leaching has gained favor over cyanide leaching because it is a non-toxic reagent and leaches more quickly. But the main problems with the thiosulfate leaching system are that it uses a lot of reagents and does not have a good method for recovering gold [20, 21].

Despite considerable efforts to develop environmentally benign alternatives to traditional cyanide-based leaching reagents for silver extraction, these substitutes still present several limitations. Common drawbacks include high operational costs, limited potential for reagent recycling, and challenges in maintaining control over key process variables. These issues underscore the need for continued research into effective cyanide alternatives.

Although Starovoytov [22] investigated the dissolution of silver (Ag) in ammonia solution, Aydogan et al. [23,24] studied the dissolution of Ag in ammonium salts, noting that ammonia is volatile and can easily escape from open leaching vessels, potentially causing environmental harm. Aydogan et al. [23,24] concluded that using only ammonia solution as a leaching agent for silver extraction is not advisable. Instead, they recommended the use of ammonium salts such as ammonium carbonate, diammonium tartrate, or ammonium nitrate.

Since the leaching kinetics of ammonium carbonate and diammonium tartrate have already been studied, the authors chose to investigate ammonium nitrate in this study to evaluate the feasibility of using an ammonium nitrate and hydrogen peroxide solution as an alternative leaching system for metallic silver.

Ammonium nitrate, commonly used as a chemical fertilizer in agriculture, known for its high solubility and availability [25,26], making it an attractive candidate for hydrometallurgical applications. When combined with hydrogen peroxide, the system can provide the oxidative potential required to dissolve metallic silver while minimizing environmental impact. This approach avoids the generation of harmful byproducts, aligning well with the principles of green chemistry. The work specifically aims to investigates the leaching kinetics of this eco-friendly system under various experimental conditions, including changes in temperature, reagent concentration, and reaction time. By analyzing these variables, the study aims to determine the reaction order, activation energy, and potential rate-controlling steps.

2. Materials and Methods

A high-purity metallic silver disc (99.99%) was used in all leaching experiments. The disc had a surface area of 23.85 cm² and a thickness of 3 mm. Ammonium nitrate (NH_4NO_3), used as the leaching reagent, was obtained from Merck, as was the hydrogen peroxide (H_2O_2) solution employed as the oxidizing agent.

A silver disc was attached to a Teflon-coated mechanical shaft, which was immersed in a 1000-milliliter beaker containing the leaching solution and placed in a temperature-controlled water bath for the experiments (Figure 1). The effects of 0.08–0.50 M H2O2, rotating speed between 314 rad/min and 942 rad/min, 0.03–0.20 M ammonium nitrate, and 20–50 °C temperature ranges were investigated. In each test, half a liter of solution was used to extract the metallic silver over a period of 24 min. The dissolved silver was analyzed with an atomic absorption spectrophotometer (GBC SensAA model). Samples of the leaching solution were taken frequently. High-quality reagents and distilled water were used for each experiment.

3. Results and Discussion

3.1. Leaching Mechanism of Ag with ammonium nitrate

To comprehend the mechanism of formation of ammonia complex with Ag, it is imperative to examine the impact of pH on Ag dissolution. The experiment was run with the following parameters:

30 °C, 0.15 M ammonium nitrate concentration, 314 rad/min rotation speed, and 0.15 M hydrogen peroxide concentration. The pH value under these circumstances was found to be 4.84. NaOH was used to raise the pH above 4.84. As shown in Figure 2, the solubility of silver increases in an alkaline medium with increasing pH. This experiment indicates that ammonia and silver combine to form a stable complex under conditions of an alkaline pH. The stability of the ammonia-silver complex is demonstrated by equation (1), which also shows a relatively high formation constant ($K_f = 1.1\mathrm{x}10^7$) [27].

$$\mathrm{Ag}^+ + 2NH_3 \rightarrow [\mathrm{Ag(NH_3)_2}]^+ \qquad Kf = 1.1\mathrm{x}107 \tag{1}$$

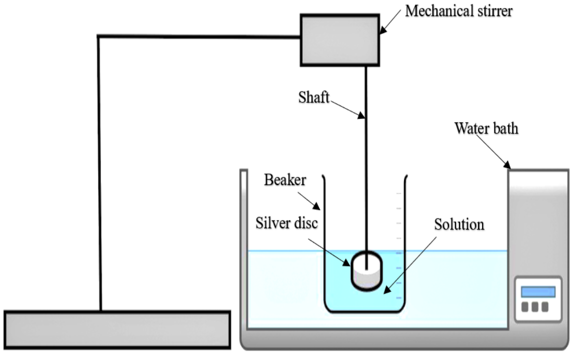


Figure 1. Experimental set-up.

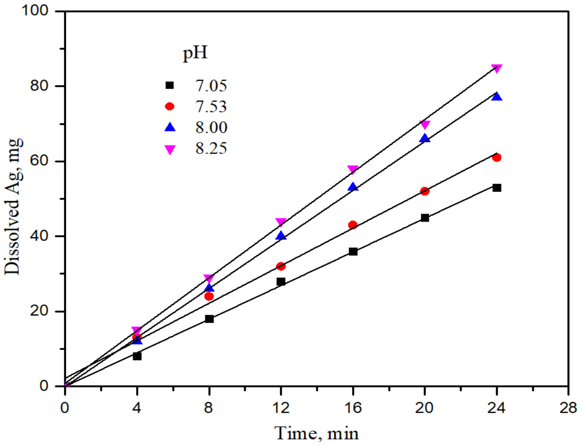


Figure 2. Impact of pH on the leaching of Ag rotating disc in alkaline medium (conditions: rotation speed of 314 rad/min, 0.15 M ammonium nitrate concentration, 30 °C, 23.85 cm2 disc surface area and 0.15 M H2O2).

In this situation, the following equations can be used to explain the mechanism of Ag dissolution in alkaline medium. Equation 2 demonstrates that H_2O_2 has a strong oxidizing agent value of 1.007 V in an alkaline medium [27].

$$H_2O_2 + 2e^- \rightarrow 2OH^- \quad E° = +1.007\ \mathrm{V} \tag{2}$$

The following formulas can be used to understand the mechanism of silver-ammonia complex formation [27]:

$$2\mathrm{Ag}^{\circ} \rightarrow 2\mathrm{Ag}^+ + 2e^- \quad E° = -0.8\ \mathrm{V}\ \text{at } 25\ °C \tag{3}$$

$$2\mathrm{Ag}^{\circ} + H_2O_2 \rightarrow 2\mathrm{Ag}^+ + 2OH^- \quad E° = +0.207\ \mathrm{V}\ \text{at } 25\ °C \tag{4}$$

Equation 5 and 6, which expresses the final equations describing the dissolution of Ag with NH_4NO_3 in an alkaline medium, can be derived from Equations 3 and 4.

$$2\mathrm{Ag}^+ + 2NH_4\mathrm{NO_3} \rightarrow 2[\mathrm{Ag(NH_3)_2}]^+ + 2\mathrm{NO_3^-} + 2\mathrm{H}^+ \tag{5}$$

$$2\mathrm{Ag}^{\circ} + H_2O_2 + 2NH_4\mathrm{NO_3} \rightarrow 2[\mathrm{Ag(NH_3)_2}]^+ + 2\mathrm{NO_3^-} + 2H_2O \tag{6}$$

Figure 3 also demonstrates that silver is soluble in acidic media. The reaction orders with respect to solution pH were determined to be 0.009 in acidic medium and 0.17 in alkaline medium.

As shown in Figure 4, the reaction order obtained for the acidic medium is lower than that for the alkaline medium, indicating that silver dissolution is minimally affected by changes in pH under acidic conditions.

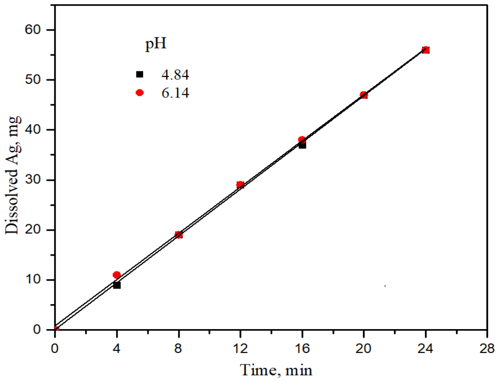


Figure 3. Impact of pH on the leaching of Ag rotating disc in acidic medium (conditions: rotation speed of 314 rad/min, 0.15 M ammonium nitrate concentration, 30 °C, 23.85 cm^2 disc surface area and 0.15 M H_2O_2).

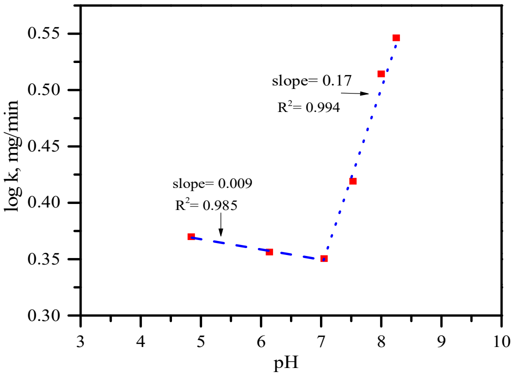


Figure 4. The reaction orders in relation to pH (conditions: rotation speed of 314 rad/min, 0.15 M ammonium nitrate concentration, 30 °C, 23.85 cm^2 disc surface area and 0.15 M H_2O_2).

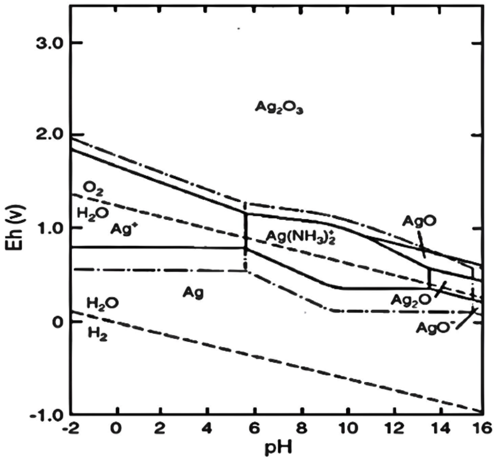


Figure 5. The Eh-pH diagram of silver-ammonia-water (Ag-NH_3-H_2O) system [23]

The Pourbaix diagram of the NH_3–H_2O–Ag system (Figure 5) suggests that an acidic environment inhibits the formation of silver-ammonia complexes. In this case, the following formulas can be used to explain the mechanism of silver dissolution.

Equation 7 shows that H_2O_2 has a 1.763 V redox potential value in acidic condition [24].

$$H_2O_2 + 2H^+ + 2e^- \rightarrow 2H_2O \quad (7)$$
E= +1.763 V

The following formulas can be used to explain the mechanism of silver nitrate formation [24]:

$$2Ag \rightarrow 2Ag^+ + 2e^- \quad (8)$$
E°= -0.8 V

$$2Ag + H_2O_2 + 2H^+ \rightarrow 2Ag^+ + 2H_2O \quad (9)$$
E= +0.903 V

Equation 10 and 11, can be utilized to express the final equations that characterizes the leaching of Ag with NH_4NO_3 in an acidic medium, based on Equations 8 and 9.

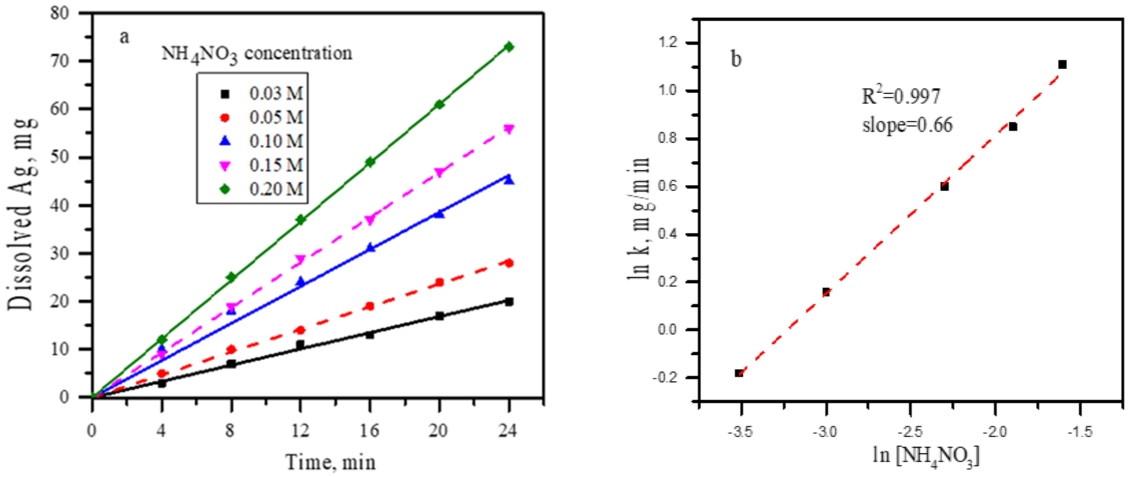
$$2Ag^+ + 2NH_4NO_3 \rightarrow 2AgNO_3 + 2NH_4^+ \quad (10)$$

$$2Ag^\circ + H_2O_2 + 2H^+ + 2NH_4NO_3 \rightarrow 2AgNO_3 + 2NH_4^+ + 2H_2O \quad (11)$$

3.2. Effect of Ammonium Nitrate Concentration

At a 314 rad/min rotation speed, a temperature of 30 °C, and a concentration of 0.15 M H_2O_2, the effect of ammonium nitrate concentration on Ag dissolution is shown in Figure 6(a). The dissolution of Ag was found to be significantly influenced by the concentration of ammonium nitrate, which ranges from 0.003 to 0.200 M. Increasing concentration of ammonium nitrate causes a proportion of free ammonia to rise, which facilitates the formation of ammonia- silver complexes and explains why the dissolution of Ag is increasing.

A logarithmic linearization method was used to determine the reaction order in order to quantify the impact of ammonium nitrate. The natural logarithm of the dissolution rate (ln(rate)) was plotted against the natural logarithm of the ammonium nitrate concentration (ln [NH_4NO_3]). The resulting linear plot, shown in Figure 6(b), yielded a slope of 0.66, indicating a reaction order less than unity. This result suggests that while the dissolution rate of silver increases with ammonium nitrate concentration, the relationship is not directly proportional.



(a) (b)

Figure 6. (a) Influence of ammonium nitrate concentration on the Ag leaching (conditions: rotation speed of 314 rad/min, 30 °C, 23.85 cm^2 disc surface area and 0.15 M H_2O_2); (b) The reaction order in relation to ammonium nitrate concentration.

3.3. Effect of H_2O_2 Concentration

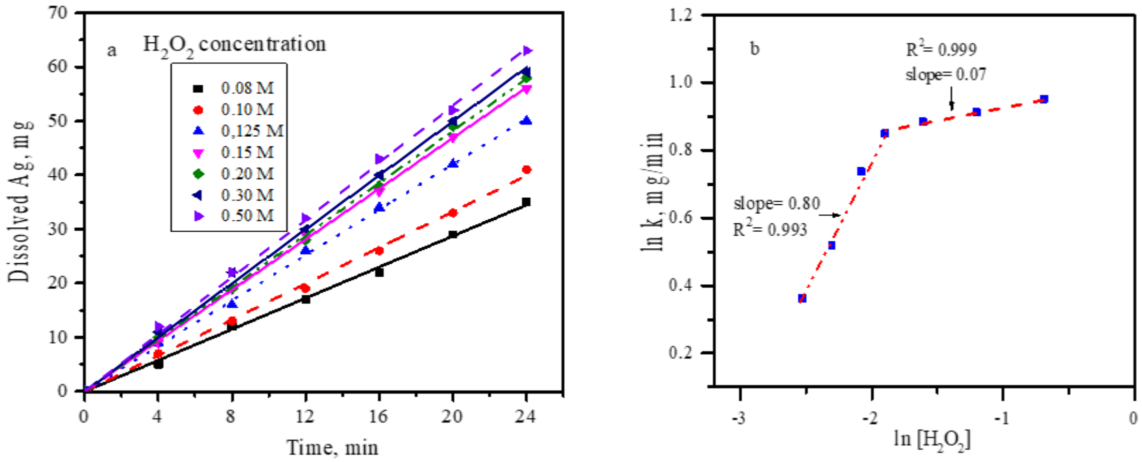
Using a 0.15 M ammonium nitrate concentration, 314 rad/min rotation speed, and 30 °C temperature, Figure 7(a) shows the Ag dissolution versus H_2O_2 concentration in the 0.080–0.50 M range. The results show that silver dissolves more readily when H_2O_2 concentration is raised to a range of 0.08–0.15 M. However, at a concentration of 0.20–0.50 M, there is no appreciable changes on the Ag dissolution. The chemical reaction order of H_2O_2 concentration in silver dissolution was determined to be 0.80 and for the concentrations of 0.08–0.15 M and 0.20–0.50 M, respectively, based on the linearized plots in Figure 7(b).

The concentrations of H_2O_2 exhibit two slopes, as illustrated in Figure 7(b). This behavior can be attributed to the role of H_2O_2 as an oxidizing agent, which facilitates the reaction between silver and ammonium nitrate. The concentration range of H_2O_2 required to provide sufficient oxidative potential for significant silver solubility is between 0.08 and 0.15 M. Beyond this range, particularly at concentrations above 0.20 M, the slope of the curve approaches zero (0.07), indicating that further increases in H_2O_2 concentration have a negligible effect on silver dissolution. Aydogan et al. [27] noted that at high H_2O_2 concentrations, an oxygen layer formed on the silver disc.

3.4. Impact of Temperature

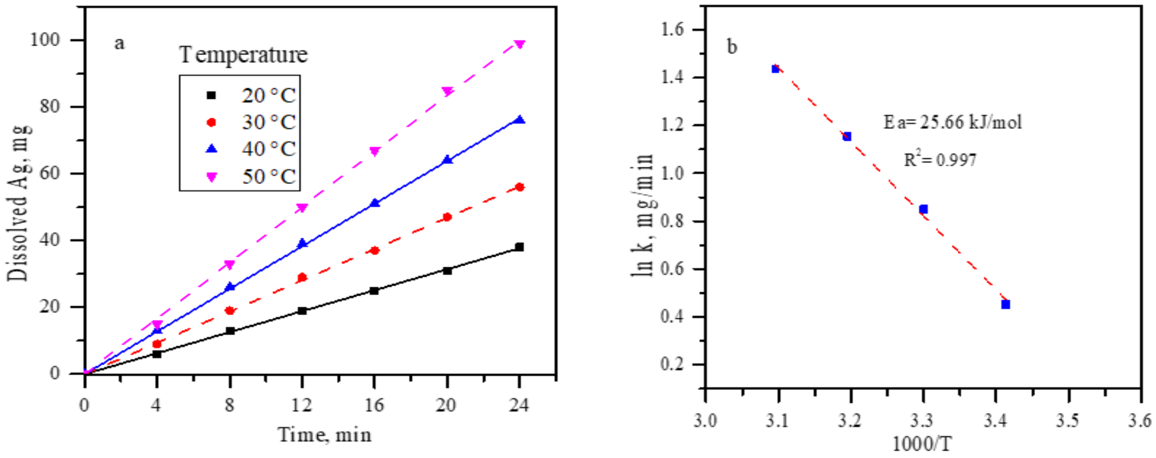
The impact of temperature between 20–50 °C on the leaching rate of Ag at a rotation speed of 314 rad/min, an ammonium nitrate concentration of 0.15 M, and a H_2O_2 concentration of 0.15 M presented in Figure 8(a). It demonstrates how rising temperatures cause silver to dissolve more readily. Since hydrogen peroxide normally breaks down at temperatures above 60 °C, the rate at which silver dissolves has not been investigated at higher temperatures [28, 29].

After calculation based on the Arrhenius plot, the activation energy of the process was found to be 25.66 kJ/mol as illustrated in Figure 8(b). This value falls within the typical range for mixed kinetic control (12–39 kJ/mol) [30, 31] suggesting that the leaching process is controlled by a combination of surface chemical reaction and diffusion mechanisms. Similar observations have been reported in the literature. For instance, Yang and Honaker [32] reported an activation energy of 27 kJ/mol, which indicated a mixed control mechanism. Likewise, Xu et al. [33] obtained a value of 22.056 kJ/mol in their study on willemite leaching, also supporting the interpretation of mixed kinetic behavior.



(a) (b)

Figure 7. (a) Influence of H_2O_2 concentration on the Ag leaching (conditions: 314 rad/min rotation speed, 30 °C, ammonium nitrate concentration of 0.15 M, 23.85 cm^2 disc surface area); (b) reaction order regarding to H_2O_2 concentration

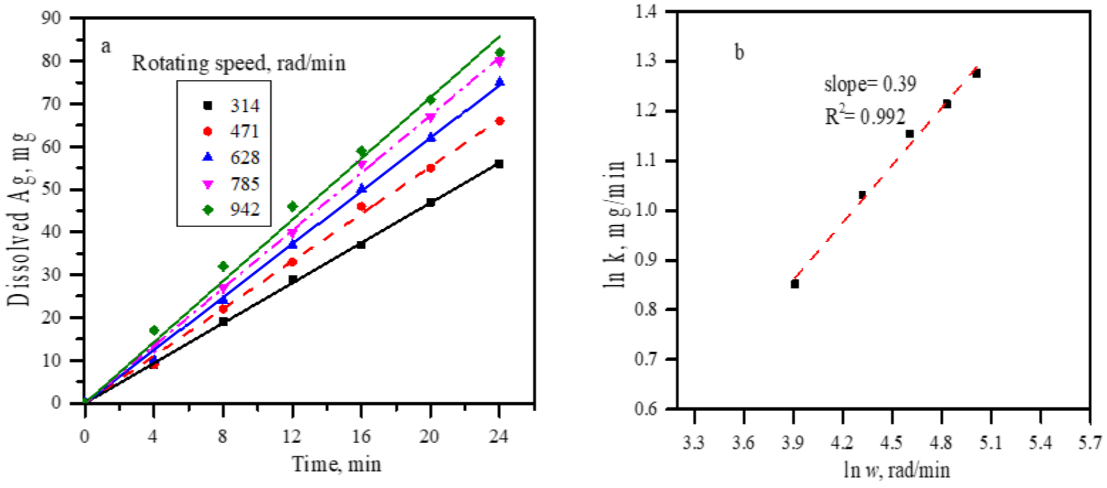


(a) (b)

Figure 8. (a) Impact of temperature on the leaching of Ag (conditions: ammonium nitrate concentration of 0.15 M, rotation speed of 314 rad/min, 0.15 M H_2O_2, 23.85 cm^2 disc surface area); (b) reaction order in relation to temperature

3.5. Impact of Rotating Speed

The impact of rotation speed on the dissolution of silver at an ammonium nitrate of 0.15 M, a temperature of 30 °C, and a H_2O_2 concentration of 0.15 M presented in Figure 9. The results proved that the rotation speed has a good impact on the dissolution rate of Ag, which has an order of 0.388, as can be seen in Figure 9(b). Higher rotation speeds enhance the leaching rate by improving mass transfer at the solid–liquid interface. Additionally, the increase in the rotation speeds ensures a more homogeneous distribution of oxidants and complexing agents, which leads to more consistent dissolution rates. These results are in agreement with previous studies, such as Aydogan et al. [27], who reported that the silver leaching rate was significantly influenced by the rotation speed. Based on the supposition that there is laminar flow on the surface of the rotating disc [34], the Levich equation is validated by the result from Figure 10.



(a) **(b)**

Figure 9. (a) Impact of rotation speed on the Ag leaching (conditions: 0.15 M H_2O_2, ammonium nitrate concentration of 0.15 M, 30 °C, disc surface area of 23.85 cm^2); (b) reaction order according to rotation speed.

3.6. Impact of Rotating disc surface area

Figure 11 (a) shows the impact of the disc surface area on the Ag leaching rate at an ammonium nitrate concentration of 0.15 M, a temperature of 30 °C, a rotation speed of 314 rad/min, and a H_2O_2 concentration of 0.15 M. Increases in surface area have been found to accelerate the rate at which silver dissolves. As shown in Figure 11(b), the slope of approximately 1. In this case, Levich and Tobias assume that the dissolution rate of Ag is directly proportional to the surface of disc [34].

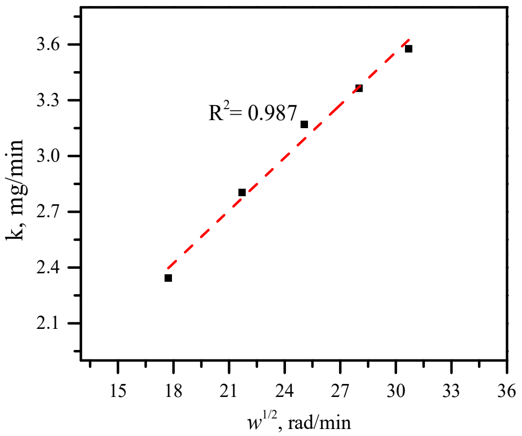
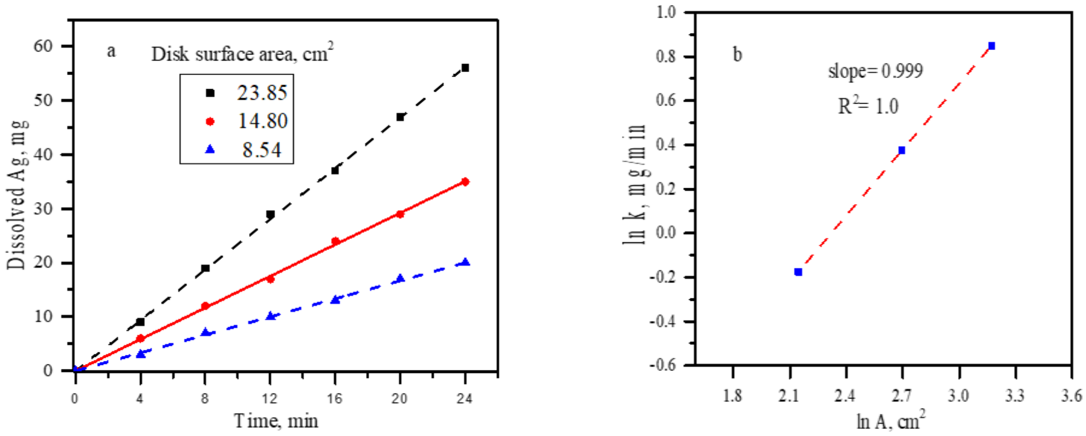


Figure 10. The relationship between the leaching rates and $\omega^{1/2}$ at 0.15 M H_2O_2, ammonium nitrate concentration of 0.15 M, 30 °C, 23.85 cm^2disc surface area).



(a) **(b)**

Figure 11. (a) Impact of disc surface area on the Ag leaching (conditions: 0.15 M H_2O_2, ammonium nitrate concentration of 0.15 M, 30 °C, rotation speed of 314 rad/min); (b) reaction order according to disc surface area

3.7. Derivation of the Overall Kinetic Model

In 1963, Levich and Tobias presented an equation for determining the rate of dissolution of the rotating disc [34]. The amount of dissolved metal is established by this equation as a function of multiple parameters—such as rotation speed, surface area, concentrations of hydrogen peroxide and ammonium nitrate, time, and temperature—as expressed in Equation (12). This comprehensive approach makes it more robust and informative compared to many previous models.

$$X = f(\omega, A, C_{\text{hydrogen peroxide}}, C_{\text{ammonium nitrate}}, t, \exp(-E/RT)) \qquad (12)$$

Where are:

A - surface area (cm^2)

E - activation energy (kJ/mol),

k - rate constant

X - mass of silver dissolved (mg)

ω - the rotational speed of the disc (rad/min)

$C_{\text{ammonium nitrate}}$ - the concentration of ammonium nitrate(M)

and $C_{\text{hydrogen peroxide}}$ - the hydrogen peroxide concentration (M)

R - the universal gas constant

By changing the parameters, equation (13) was produced.

$$X = k\,\omega^{e} A^{f} C_{\text{hydrogen peroxide}}^{\;g}\, C_{\text{ammonium nitrate}}^{\;h}\, e^{-E/RT}\, t \qquad (13)$$

where e, f, g, and h are constants to be found in this work.

The following empirical model can be used to express the degree of the Ag dissolution (X) of this process:

$$X = 4.419\times10^{3}\, \omega^{0.5} A^{0.999} C_{\text{hydrogen peroxide}}^{\;0.80}\, C_{\text{ammonium nitrate}}^{\;0.66} e^{-25660/RT}\, t \qquad (14)$$

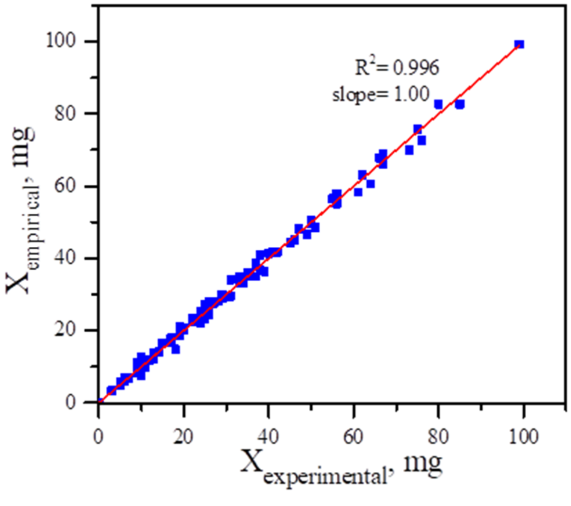


Figure 12. Empirical vs. experimental dissolved Ag

Ammonium nitrate has an order (0.66) less than that of hydrogen peroxide (0.80). These orders indicate that the most effective parameter in the system was the hydrogen peroxide.

As demonstrated by the linear correlation in Figure 12 with a slope of 1 and a high coefficient of determination ($R^2 = 0.996$), this model exhibits strong agreement with the experimental results, confirming the predictive accuracy of the model.

4. Conclusions

This work examined the leaching kinetics of pure Ag using ammonium nitrate in H2O2 solution instead of cyanidation process due to the environmental problems of cyanide. The dissolution kinetics of Ag were studied by investigating the influences of temperature, ammonium nitrate concentration, H_2O_2 concentration, and rotating speed on the dissolution rate. The findings show that a small concentration of ammonium nitrate (0.15 M) was sufficient to dissolve silver alternatively to cyanide. This suggests that ammonium nitrate may serve as a viable, potentially safer alternative to cyanide in Ag extraction processes. It was found that the H_2O_2 concentrations between 0.08-0.15 M are enough to provide adequate oxidative potential to achieve a high degree of Ag solubility. Increasing the H_2O_2 concentration beyond 0.20 M did not result in further improvement in dissolution efficiency, indicating a saturation point in oxidative activity.

Kinetic analysis yielded an apparent activation energy of 25.66 kJ/mol

The study recommends further application of ammonium nitrate leaching to silver-bearing ores and comparative analysis with traditional cyanidation methods to evaluate industrial feasibility. Such investigations will help establish the practicality and environmental benefits of adopting ammonium nitrate-based leaching systems for large-scale silver extraction.

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Conflict of interest

The authors declare that they have no conflict of interest.

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سینتیک انحلال نقره فلزی با استفاده از محلول نیترات آمونیوم و پراکسید هیدروژن به عنوان جایگزین سیانید سازگار با محیط زیست

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کلمات کلیدی

نیترات آمونیوم
پراکسید هیدروژن
انحلال
نقره
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چکیده

این مقاله سینتیک استفاده از نیترات آمونیوم برای حل کردن نقره فلزی خالص در محلول پراکسید هیدروژن (H_2O_2) را شرح می‌دهد. استفاده از نقره فلزی خالص با حذف تداخل ناشی از ناخالصی‌ها و تسهیل تفسیر دقیق مکانیسم‌های کنترل سرعت، امکان تحقیق دقیق سینتیک لیچینگ را فراهم می‌کند. تأثیر دما، سرعت چرخش، غلظت H_2O_2 و غلظت نیترات آمونیوم همگی مورد بررسی قرار گرفتند. نتایج، رابطه مطلوبی را بین سرعت انحلال نقره (Ag) و سرعت چرخش نشان می‌دهد. علاوه بر این، غلظت کم نیترات آمونیوم (بین ۰.۰۰۳ تا ۰.۲۰ مولار) اثرات مفیدی بر انحلال نقره دارد. سرعت انحلال به طور قابل توجهی تحت تأثیر غلظت‌های H_2O_2 بین ۰.۰۸ تا ۰.۱۵ مولار قرار گرفت، زیرا این محدوده از غلظت H_2O_2 برای فراهم کردن پتانسیل اکسیداتیو کافی برای حلالیت قابل توجه نقره مورد نیاز است. با این حال، این اثر در محدوده ۰.۲۰ تا ۰.۵۰ مولار کمتر مشهود است. محدوده دمایی ۲۰ تا ۵۰ درجه سانتیگراد مفید است زیرا H_2O_2 در این محدوده پایدار است. محاسبه شد که انرژی فعال‌سازی ۲۵.۶۶ کیلوژول بر مول بود.

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