

Investigation of thermodynamic and kinetic regularities of asbestos waste leaching processes

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ABSTRACT

The asbestos wastes were exploited for extraction of magnesium by agitation leaching process in atmospheric conditions, using hydrochloric acid as the leaching agent. The study was focused on investigating the kinetics of magnesium leaching. The effects of some variables, including stirring speed (150–300 r/min), leaching temperature (25–90 °C), leaching duration (10–60 min), the concentration of hydrochloric acid (150–300 g/L) and the solid - liquid ratio (1:2–1:5), on the leaching rate were carefully evaluated. It was found that increasing the stirring speed, leaching temperature, and concentration of hydrochloric acid can significantly increase the rate at which magnesium is leached. A comprehensive kinetic scrutinization was performed by fitting the experimental data with exponential kinetic equation, Yander diffusion equation, Drozdov-Rotinyan equation, and Prout-Tompkins equation. Amongst all these tested kinetic equations, the diffusion-controlled process showed a higher goodness-of-fit with experimental data. Thermodynamic aspects were also appraised carefully. The apparent activation energy for the process was calculated to be 19.8 kJ/mol, indicating a relatively low-level energy needed to initiate the process efficiently. Overall, the current study provided insight into how changing certain parameters can affect the overall performance when attempting to extract magnesium from the asbestos wastes by leaching process.

1. Introduction

Asbestos and associated materials have been used for centuries in many different industries. Asbestos is made up of tiny, microscopic fibers which are resistant to heat, fire, and chemical damage, as well as being highly durable. Hence, it has been widely utilized in various fields due to its high strength and insulation properties [1,2]. In the past century alone, asbestos has been employed in numerous industrial sectors, including construction materials (e.g., roofing shingles), automotive parts (e.g., brakes), and manufacturing processes (e.g., paper mill machinery) [3]. Due to its resistance to fire, it is commonly used as insulation material in buildings as well as a soundproofing substance for walls or ceilings. The latter usage particularly put workers at a great risk if not handled properly, given how easily airborne particles can be inhaled by those present onsite during installation or maintenance activities. In addition, other asbestos common applications are in

production of brake linings for automobiles and gaskets inside boilers, where temperatures are often very high. Such high temperatures create an environment which is susceptible for fiber release, resulting in further exposure risks [4,5].

As it is known, asbestos poses a significant health risk when disturbed or handled improperly. Given that prolonged exposure can lead to serious illnesses, such as mesothelioma cancer, there have been growing calls over recent years from governments around the world urging manufacturers to abandon their reliance upon this hazardous substance altogether. Indeed, some countries have taken some steps towards completely phasing out any further mining operations related to this toxic material. Apart from this, some countries have also started to reprocess the previously accumulated asbestos wastes. Utilizing such wastes as metallurgical raw materials can not only improve the environmental situation in the targeted region, but also contribute significantly to the country's raw material base [6,7].

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It is widely recognized that asbestos production waste is present on all continents and poses a significant hazard if left untreated. The lack of scientific and technological advancements in research data contributes to the accumulation of billions of tons of this waste across continents inhabited by humans. The issue of asbestos waste extends to North [8], South America [9], Europe [10,11], Asia [12], Africa [13], and Australia [14], underscoring its global nature and the absence of effective waste recycling technologies. The failure to recycle asbestos waste results in an estimated 255,000 annual deaths [15]. Moreover, the waste contains valuable metals such as magnesium, iron, nickel, cobalt, and gold, amounting to substantial resource loss. These circumstances highlight the need for comprehensive research data to facilitate the implementation of industrial technologies for addressing asbestos waste.

The extraction and processing of asbestos ores are still going on in several countries, including Russia, China, Brazil, Kazakhstan, and Canada. In particular, Kazakhstan is one of the leading producers of products based on asbestos. For more than 55 years, JSC "Kostanay Minerals" has collected over 300 million tons of waste from asbestos production in Kostanay Region of northern Kazakhstan. The current technology and the plant used for managing the asbestos wastes only yields 5–8% as marketable product, leaving the remaining 90% thrown into a waste warehouse [16,17]. In this regard, there is a need to process these wastes as raw materials for other purposes, like the magnesium production.

China and Israel are the leading producers of magnesium in the world. In China, a magnetothermic method is used that involves exploiting dolomite as raw material, containing 9–11% magnesium, while in Israel deposits of dead Sea salts with no more than 11–13% magnesium are utilized. The most attractive raw material for production of this metal is accumulated asbestos waste containing up to 25% magnesium and, as such waste is crushed on the surface, it is ready for processing [16]. However, in order to effectively utilize this resource, it is necessary to conduct an extensive study on the kinetics of leaching the gathered waste materials. Such research will provide valuable insights into how best to optimize the process of extracting metals from asbestos waste materials. It will also enable us to assess potential risks associated with such operations and develop appropriate safety measures. Moreover, understanding the kinetics of leaching can lead to exploiting more efficient methods that reduce costs while maximizing metal recovery rates. Thus, conducting a study on the kinetics of leaching asbestos waste is essential if we are to successfully extract magnesium from this material in an environmentally responsible manner [17–20].

Before commencing this research, we conducted a comprehensive literature review on the hydrometallurgical processing methods for asbestos waste. We discovered a significant knowledge gap in the existing global literature regarding the kinetic and thermodynamic aspects of hydrometallurgical processing of asbestos waste. While some articles focused on the processing of asbestos waste discussed the leaching of mercury and other elements (excluding magnesium) from asbestos cement sheets using acetic acid [21], this topic remains largely unexplored.

Our research holds practical and scientific significance. From a practical perspective, our findings could potentially facilitate the production of magnesium-containing salts, such as bischofite and carnallite, which are highly sought after by the titanium and magnesium plant in Kazakhstan. Furthermore, our work contributes to scientific knowledge by determining the mechanism of the leaching reaction involved in this process. We have identified key scientific aspects that are crucial in comprehending the reaction mechanism involved in the hydrochloric leaching process. These aspects allow researchers to gain insights into the sequential steps that occur during the dissolution of a magnesium from a solid substance, as well as the interactions between the hydrochloric acid and the asbestos wastes. Understanding these processes provides important information about the fundamental principles underlying leaching.

As it is known, the analysis of kinetics aids in identifying the factors

that govern the leaching process. By examining the rates of the reactions, researchers can determine whether the reaction is limited by the transfer of the leaching agent to the surface of the solid material (controlled by diffusion) or by the reaction kinetics at the interface between the solid and liquid phases (controlled by kinetics). This knowledge is critical for effectively controlling the leaching speed and optimizing the different stages involved. Additionally, the study of kinetics offers the opportunity to develop mathematical models and simulation methods based on the kinetic data. These models and simulations can be utilized to predict how the leaching process will behave under various conditions. By employing such predictive tools, researchers can enhance the efficiency of the leaching process and scale it up for industrial applications.

2. Experimental

2.1. Raw material characterization

Samples of asbestos tailings were collected from an asbestos plant located in Zhetikara, North Kazakhstan. The samples were already in a fine powder form and did not require any additional preparation before being subjected to leaching process. Analysis by X-ray diffraction showed the presence of the following mineralogical species in the tailings: MgO (39.0–42.0 %); SiO₂ (37.0–41.0 %); CaO (1.1–1.6 %); Fe₂O₃ (1.9–5.4 %); FeO (1.0–2.7%); Al₂O₃ (0.8–1.4 %); NiO (0.1–0.25 %); Cr₂O₃ (0.1–0.25 %). The mineral base of the ore was serpentine, chemically expressed as 3MgO·2SiO₂·2H₂O (Table 1).

Scanning electron microscopy (SEM) was applied to study the morphology of asbestos tailing samples. The SEM images showed that the asbestos tailings comprised a very fine material and formed aggregates (Fig. 1). The scanning electron microscopy image of initial asbestos tailing shows a sample composed of small, lightweight particles that are tightly packed together. The particles range in size from 0.5 to 10 µm and have a variety of shapes, including long thin fibers, short rounded fibers, and some irregularly shaped pieces. The particles include ridges or wavy lines along the length of some of the longer fibers as well as tiny pores on their surfaces. Many of these individual particles appear to be coated with material that is either lighter or darker than the underlying particle itself.

The humidity of the initial sample was 8.2 %. The poured bulk density was 1.65 g/cm³ and the tap bulk density was 1.97 g/cm³. After drying to a constant mass, representative tailings samples were analyzed using both physical and chemical methods. Before the leaching experiments, we dried the asbestos tailings at a temperature of 95–100 °C.

2.2. Leaching experiments

The study of the leaching process was conducted through a specific method. A 50-g sample of asbestos waste was placed in a sealed glass reactor with an agitator and a sampler for further experimentation. The temperature was kept constant using a thermostat with a accuracy within ±1 °C. Before starting the leaching process, the dry waste material was heated alongside its container until reaching the desired temperature, at which an acid solution that had also been heated

Table 1
Phase composition of the initial asbestos tailings.

Component	Formula	Weight, %
Serpentine	3MgO·2SiO ₂ ·2H ₂ O	68
Brucite	Mg(OH) ₂	6
Magnesium–nickel oxides	MgNiO ₂	2
Forsterite	Mg ₂ SiO ₄	8
Magnesium oxide	MgO	4
Silimanite	Al ₂ O ₃ ·SiO ₂	3
Hematite	Fe ₂ O ₃	5
Vustite	FeO	2

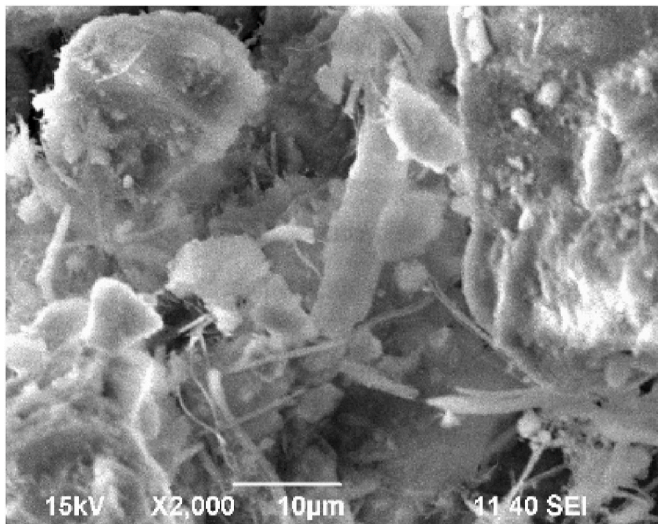


Fig. 1. Scanning electron microscopy image of initial asbestos tailing.

separately in another flask could be added. As soon as acid solutions entered the reactor, mixing started while simultaneously beginning timekeeping for the experiment. Sampling was performed after 10, 20, 30, 40, 50, and 60 min from the process's commencement for analysis purposes. Each leaching experiment was replicated three times and the obtained results were averaged. The laboratory glassware was kept overnight in a 2% (v/v) hydrochloric acid solution and then was rinsed thoroughly with deionized distilled water thoroughly. Highly pure deionized water (Milli-Q Millipore, $18.2 \text{ M}\Omega \text{ cm}^{-1}$ resistivity) was used for all dilutions throughout this work.

At selected time intervals, a known amount (5.0 mL) of pulps was withdrawn by sampling port and filtered quickly. 5.0 mL of filtrate was then diluted to 250 mL for magnesium analysis using an atomic absorption spectrometer (TAS-990).

The leaching efficiency of magnesium (LE), represented as the percentage of magnesium extracted from the initial sample during leaching, relative to the total mass of magnesium in the sample, was defined by the following mathematical expression:

$$LE (\%) = \frac{C_A \cdot V}{C_0 \cdot M} \cdot 100 \quad (1)$$

where:

C_A - is the concentration of magnesium in the samples (leach liquor), (g L^{-1});

V - is the leach liquor volume, (L);

C_0 - is the magnesium content in the asbestos wastes, (%);

and M - is the mass of the magnesium oxide, (g).

The initial asbestos wastes were analyzed by XRD (Japan Rigaku Model TTRIII + 40 kV/250 mA with $\text{Cu K}\alpha$ radiation), SEM, and back-scattered electron micrographs (BEM) (Japan Jeol JSM-6360LV).

2.3. Thermodynamic regularities of leaching process

For the thermodynamic justification of the dissolution processes of magnesium silicates, thermodynamic calculations were performed using the OUTOKUMPU OU thermodynamic calculation program (Table 2). The behavior of the main components of asbestos waste is considered in such a calculation.

The standard free energy of a reaction is an important measure of the thermodynamic feasibility of that reaction. In the case of leaching magnesium from serpentinite minerals with hydrochloric acid, it can be used to predict whether or not this process will take place and its magnitude is a measure for the thermodynamic favorability of the process. The standard free energy values for this specific reaction are

Table 2

Standard free energy (ΔG^0 , kJ) of leaching reactions of magnesium-containing components in HCl (calculated for 1 mol MgO).

Reaction	ΔG^0 , at the temperature, °C		
	30	60	90
$3\text{MgO} \cdot 2\text{SiO}_2 \cdot 2\text{H}_2\text{O} + 6\text{HCl} \rightarrow 3\text{MgCl}_2 + 2\text{SiO}_2 + 5\text{H}_2\text{O}$	-5.06	-3.11	-2.23
$\text{Mg}(\text{OH})_2 + 2\text{HCl} \rightarrow \text{MgCl}_2 + 2\text{H}_2\text{O}$	-9.98	-8.08	-7.17
$\text{MgNiO}_2 + 4\text{HCl} \rightarrow \text{MgCl}_2 + \text{NiCl}_2 + 2\text{H}_2\text{O}$	-4.65	-4.23	-4.02
$\text{Mg}_2\text{SiO}_4 + 4\text{HCl} \rightarrow 2\text{MgCl}_2 + \text{SiO}_2 + 2\text{H}_2\text{O}$	-5.89	-4.4	-3.65
$\text{MgO} + 2\text{HCl} \rightarrow \text{MgCl}_2 + \text{H}_2\text{O}$	-16.34	-14.09	-12.99

dependent on the concentrations and activities of all species involved, including HCl and Mg^{2+} ions. When these parameters fall within certain ranges, then there is enough potential energy available for Mg^{2+} transition from the solid form in serpentinite minerals into solution as part of a chemical reaction with HCl. This means that if sufficient amounts of both reactants are present at suitable concentrations and activity levels, then they will interact to form soluble products such as magnesium chloride (MgCl_2). If these conditions are not met, however, the reaction may not occur or may happen at very low rates due to insufficient potential energy being available.

Eh-pH diagram of the Mg-O-Cl system was also considered (Fig. 2), at 298 K. At a reduction potential below -1.0 , the formation of MgH_2 is favored and at potentials from $+1.2$ to -1.1 mV, magnesium ion (Mg^{+2}) is formed in solution (Fig. 2). At pH values between 0 and 8, both MgH_2 and Mg^{+2} are present in equilibrium with each other. Above pH 8.2, only the magnesium hydroxide species exist as the dominant form of magnesium in solution, while at lower pH values near 7 or less than 6 only free magnesium ions are present. The formation of perchlorate ion (ClO_4^-) occurs over a wide range of pH. As the system's acidity increases (pH decreases), HCl become more prevalent.

3. Kinetic regularities of asbestos waste leaching processes

As it is known, leaching processes are a form of chemical reaction which involve the transfer of components from one phase to another. This process can be studied through mass action kinetics, where simplifying assumptions such as pseudo-steady-state are used for equilibrium evaluations and determining rate determining step. The rate determining step is assumed to be the slowest stage in terms of speed and it controls the overall leaching kinetics. The controlling step can be identified exploiting the kinetic equations for modeling the process and finding an equation which governs the leaching. The controlling step can either be a diffusion process inside or outside the particles or be a surface

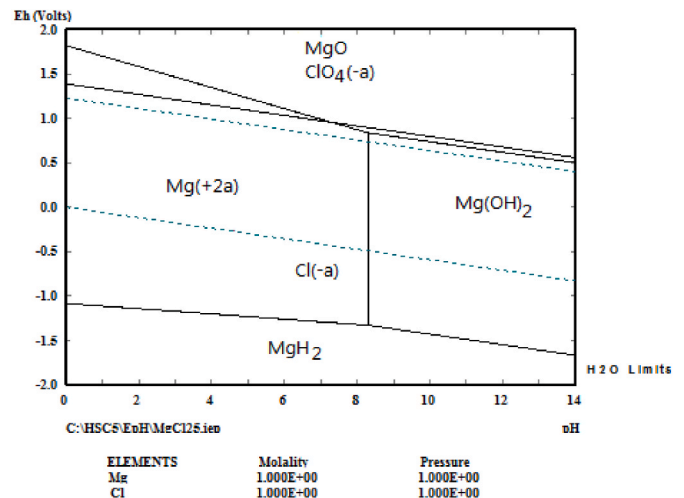


Fig. 2. Eh-pH diagram of the Mg-O-Cl system at 298 K.

reaction at the particle's surface [15].

Diffusion occurs when molecules move randomly between two phases due to concentration differences. Diffusion can occur either as film diffusion (diffusion in liquid films) or as particle diffusion (diffusion into pores). Film diffusion involves molecules (or ions) travelling across molecular boundaries, while particle diffusion includes molecules (or ions) passing through solid particles via their pore structures. Surface reactions occur on solids surfaces when reactants come into contact with each other, resulting in changes that lead to product formation. These reactions vary depending on factors such as temperature and pressure, therefore they need to be monitored carefully during experiments so that accurate results can be obtained for analysis purposes.

In order for leaching processes to occur effectively and efficiently, an understanding of these three types of processes (film diffusion, particle diffusion, and surface reactions) is needed so that proper parameters can be adjusted accordingly during experiments. Such information also helps researchers determine whether any further optimization may be necessary for better yield rates or faster completion timescales. Furthermore, knowledge about these processes allows scientists to predict how different conditions may affect leaching behavior. In fact, such knowledge leads to making rational decisions regarding optimal operating conditions for specific applications involving leaching procedures, such as metal extraction from ore materials [22,23].

The study of the kinetics of leaching processes is an important part of many industrial and scientific processes. Understanding the kinetic parameters that govern the rate at which the materials are removed during leaching processes is essential for effective design and optimization of these operations.

By studying the rate at which different substances are removed from their source material during leaching, scientists can gain insight into the mechanisms governing these processes. Such information are essential for designing efficient and cost-effective leaching operations that produce high-quality products with minimal environmental impact. By analyzing how different materials behave during leaching experiments, researchers can gain insights into the properties of minerals [24,25].

As new technologies are developed for extracting valuable resources from environment, while minimizing potential harms to ecosystems, having an accurate understanding of the kinetic parameters governing these processes is essential. The study of the kinetics of leaching processes provides invaluable data which allows optimization of existing procedures as well as developing new ones that meet both economic and environmental goals efficiently while protecting public health and safety [26].

3.1. Procedures exploited for interpreting kinetic data

3.1.1. The Yander diffusion equation

The Yander diffusion equation is a mathematical model used to describe the leaching process of ore. It is based on Fick's second law of diffusion, which states that the rate at which a substance diffuses through a medium is proportional to the concentration gradient between two points in space. The Yander diffusion equation takes this reality into account and provides an mathematical expression which can be given as:

$$1 - (1 - \alpha)^{1/3} = k\tau^{0.5} + b \quad (2)$$

where:

k - reaction rate constant, (is a proportionality constant for a given reaction), (min^{-1});

α - is the leaching efficiency, (%);

b - is a rate constant that describes the rate at which solutes are transferred from one medium to another.

The Yander diffusion equation has been successfully used to model many different types of leaching processes including those involving copper, zinc and gold ores [27,28]. In particular, it can be used to predict

not only particle transport but also other important parameters such as total dissolved solids concentrations along with pH values during leaching operations. Furthermore, its numerical implementation allows researchers to simulate various scenarios related to ore processing under different conditions like temperature and pressure changes. This, in turn, helps us identify optimal operating parameters for efficient extraction processes with minimal environmental impacts.

3.1.2. The exponential kinetic equation

The kinetic equation $1 - (1 - \alpha)^{1/3} = k\tau + b$ is an important tool for data processing in leaching processes. This equation provides a mathematical model for the rate of release of a substance from a solid matrix, and can be used to calculate the rate constant (k). The equation is derived from the assumption that the reaction follows first-order kinetics, which means that the amount released at any given time is proportional to both the concentration of material remaining in solution and its rate constant [22,29].

When applied to leaching process, this kinetic equation helps us understand how quickly certain substances are being extracted from their source material. By measuring concentrations over time, one can determine how long it will take for all or most of a substance to be removed by leaching. We can also use this information to assess potential environmental risks associated with releases into water bodies or soils due to leaching activities.

The kinetic equation also assists researchers in identifying optimal conditions for extracting desired materials from their sources using various extraction techniques such as solvent extraction or ion exchange methods.

Overall, the kinetic equation has significant scientific relevance when it comes to interpreting results obtained through data processing of leaching processes, experiments, and simulations. It provides valuable insight into reaction rates as well as optimal conditions necessary for effective extractions, while simultaneously helping us minimize environmental impacts caused by these activities [30].

3.1.3. The Drozdov-Rotinyan equation

The Drozdov-Rotinyan equation is a mathematical expression that describes the leaching process of solids in liquids. The equation was developed by Soviet scientists, Alexander Drozdov and Vladimir Rotinyan, in the early 1970s, and it remains an important tool for understanding how solutes are released from materials during dissolution [31]. In its most basic form, the equation states that the rate at which a solid dissolve into a liquid is proportional to both the concentration of dissolved material in solution (C) and the surface area of exposed material (A). This relationship can be expressed mathematically as:

$$\text{Rate} = k(C)(A) \quad (3)$$

where:

k - reaction rate constant (is a proportionality constant for a given reaction), (min^{-1});

c - is the concentration of dissolved material in solution, (g L^{-1});

A - is the surface area of exposed material, (m^2/g).

The Drozdov-Rotinyan equation is a useful tool for predicting the rate of dissolution and quantifying the amount of material released from solids over time. It can be used to understand how quickly various chemicals are released into aqueous solutions. It can also be used to evaluate processes such as leaching in soils and groundwater contamination. The equation has been extended to more complex forms that takes into account additional factors, such as particle size distribution and surface area variations due to chemical reactions that occur during leaching. Recent formula is often expressed in the form of equation (3):

$$\frac{1}{\tau} \ln \frac{1}{1 - \alpha_1} - \beta \frac{\alpha_1}{\tau} = M \quad (4)$$

where:

τ - is the duration of interaction, (min);
 α_1 - is the leaching efficiency, (%);
 β and M – are the coefficients of braking and total speed, respectively.

Panshin et al. [32] reported that the method describes well if the dissolution is inhibited by a layer enveloping the particles. These modifications provide further insight into the dynamics of solute release from materials under different conditions. In addition, various theoretical models have been developed based on this equation which allow researchers to better understand how changes in environmental parameters (such as temperature or pH) affect the rate at which substances are dissolved from solid materials. If the leaching process is described by the Drozdov-Rotinyan equation, it means that there is a proportionality between two important variables: concentration (C) and surface area (A). This indicates that an increase in either one will result in an increase in dissolution rate; likewise, if either variable decreases then so does the rate of dissolution. The constant k provides information about how much each variable affects dissolution. Higher k values indicate greater sensitivity while its lower values suggest less influence on overall rate of material's release from solids into liquids. Knowing these relationships allows scientists and engineers to predict how much material will be released under different circumstances and develop strategies for controlling leaching processes accordingly.

3.1.4. The Prout-Tompkins equation

The Prout-Tompkins equation is an important tool used to model the leaching process. This equation was developed by American chemist Henry Prout and British physicist Thomas Tompkins, in 1939, and ever since it has been used as the basis for many models of physical and chemical processes involving solute transfer [33]. It also provides a mathematical framework for understanding and predicting the behavior of leaching systems, which can be used to develop efficient and cost-effective processes. The equation takes into account factors such as temperature, pressure, acid concentration, particle size distribution, surface area of solids and solubility constants of compounds in solution. By using this equation to analyze data obtained from a leaching process, it is possible to accurately predict the rate at which components are released into solution. Such information can then be used to determine the optimal parameters for running a given leaching system in order to maximize efficiency or optimize production costs.

At its most basic level, the Prout-Tompkins equation states that the rate at which a given amount of material (e.g., a dissolved salt) moves from one medium to another is proportional to both the difference in concentration between them and an exponential decay factor related to mass transfer resistance. The equation can be written as:

$$\ln[a / (1 - \alpha)] = k\ln\tau + b \quad (5)$$

where:

k - reaction rate constant (is a proportionality constant for a given reaction), (min^{-1});
 α_1 - is the leaching efficiency, (%);
 τ - is the time, (min);
 b - is a rate constant that describes the rate at which solutes are transferred from one medium to another.

3.1.5. The activation energy

The activation energy of a leaching process is an important measure of the overall efficiency and effectiveness of this type of chemical extraction. It represents the amount of energy required to initiate or catalyze a reaction, thus providing insight into how quickly and effectively certain compounds can be extracted from their source material. This value is expressed in joules per mole (J/mol) and provides information about the thermodynamic stability associated with each step in the reaction pathway [34].

On a fundamental basis, it helps scientists better understand the

energies involved in leaching processes and identify what factors may make them more or less efficient. For example, if two different types of materials are being used for leaching operations but one has an activation energy that is much higher than the other's, then it might suggest that reactions involving this material will require more energy input to achieve complete extraction. It also allows researchers to compare different systems side-by-side when analyzing which ones offer superior performance characteristics.

From an application standpoint, knowledge about activation energies can help us optimize various aspects in industrial operations as well as enable designers to create new products or improve existing ones by choosing materials with lower activation energies that allow faster extractions without sacrificing quality or yield. The activation energy can be calculated by employing the linearised form of the Arrhenius equation (equation (6)).

$$\ln k = \ln A_0 - \frac{E_a}{RT} \quad (6)$$

where:

A - is the frequency factor;
 E_a - is the apparent activation energy, (kJ/mol);
 R - is gas equilibrium constant, $R = 8.314$ (J/mol);
 T - is the absolute temperature, (K).

Overall, knowing what kind of impact changes in reactant composition have on activation energies plays a key role when designing successful leaching processes since it enables engineers to determine what conditions must be met before extracting target molecules out from their sources with greatest degree success possible under given constraints faced during implementation stage [35].

4. Results and discussion

4.1. The effects of leaching temperature and time

Temperature is a critical factor in the leaching process of ore raw materials. It has an influence on both the rate and extent of leaching, which can affect the quality of extracted products. High temperatures increase reaction rates due to increased molecular motion, while low temperatures decrease reaction rates by reducing molecular motion. This can significantly alter the time required to achieve optimal extraction levels. Additionally, temperature affects how much of each mineral is extracted from ore raw materials, so that higher temperatures usually tend to extract more minerals than lower temperatures do. The temperature range which is most suitable for extracting desired minerals depends upon specific characteristics such as particle size and chemical composition of the material being processed.

The value of temperature effect evaluations in leaching processes lies in its ability to optimize extraction efficiency and product quality at acceptable costs. Beside temperature, by controlling other variables such as reactant concentrations, solid-liquid ratio, and stirring speed, it is possible to maximize yield with minimal losses due to side reactions or incomplete extractions from ore raw materials.

In this study, the leaching of asbestos waste was investigated to determine the effect of temperature and time on magnesium extraction rate. Four temperatures, i.e., 25 °C, 50 °C, 75 °C, and 90 °C, were used with an initial hydrochloric acid concentration of 250 g/L, a stirring speed of 300 rpm, and a solid-liquid ratio of 1:3. The results are presented in Fig. 3 which shows the extraction rate curves obtained from this experiment. It is seen that as the leaching temperature increases linearly so does the extraction rate.

An increase in duration can result in a more complete reaction between acid and magnesium, as can be seen by an elevation from 9 to 36% extraction (Fig. 3.). Nevertheless, at extended durations beyond 60 min, pulp filtration rate will be decreased which can have negative implications for process costs.

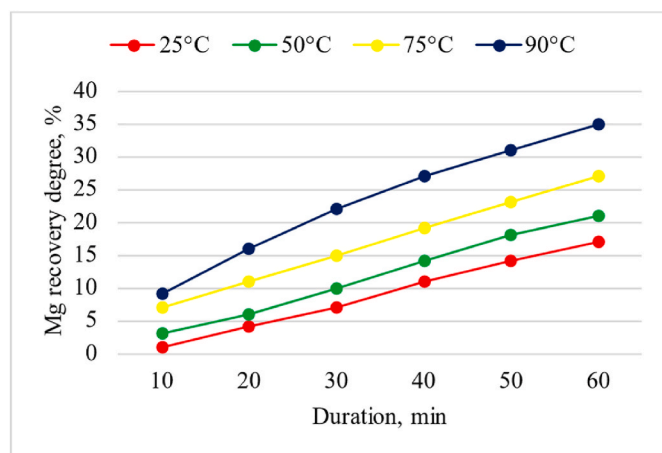


Fig. 3. The effects of leaching temperature and time on the Mg recovery degree.

In order to compare the results of this study with those from similar research, further references should be consulted. For example, Singh et al. [35] conducted a study on leaching of magnesium oxide using sulfuric acid and found that temperature had an optimum value at 70 °C for maximum extraction rate. This agrees with the findings of this study which showed that while increasing temperature up to 85 °C can result in increased extraction rate, beyond that point no additional benefit was obtained. Furthermore, Dönmez et al. [36] also investigated leaching of magnesium oxide but used acetic acid instead and achieved a higher extraction rate than that seen here when temperatures exceeded 55 °C. These studies suggest that different acids may have varying effects on magnesium extraction rates depending on temperature and other factors such as stirring speed or solid-liquid ratio which require further investigation into their influence on the reaction kinetics between acid and metal ions. Recent experimental research [37] has shown that chrysotile asbestos can be leached using oxalic acid, which aims to generate magnesium-rich solutions. The authors utilized a 10-day leaching process to achieve this goal. However, after carefully considering the objective of our research, which is to enhance the industrial production of magnesium, we believe that using hydrochloric acid as the leaching reagent would be a more practical choice.

4.2. The effects of acid concentration and time

The concentration of the leaching agent is a critical factor in determining the success of ore leaching. It has a direct impact on both the rate and extent of metal extraction from its raw materials, making it an important parameter to be considered when designing and executing leaching operations. Generally speaking, increasing the concentration of reagents used for leaching will result in higher rates of reaction due to increased reactant availability, but may also lead to excessive dissolution or poisoning effects if concentrations are too high. Therefore, selecting appropriate concentrations for optimal results requires careful consideration, based on factors such as solubility limits, toxicity thresholds, and other environmental constraints. Additionally, different metals require different levels of reagent concentration for successful extraction; thus it is essential that these be taken into account during process design and operation. Ultimately, understanding how varying concentrations influences the rate and efficiency of metal extraction from ore raw materials can help us ensure maximum yield while minimizing costs associated with reagent use.

The leaching rate of magnesium was studied with respect to the hydrochloric acid concentration in a range from 150 to 300 g/L. The results, as displayed in Fig. 4, demonstrate that the recovery degree of magnesium increases significantly with an increase in hydrochloric acid

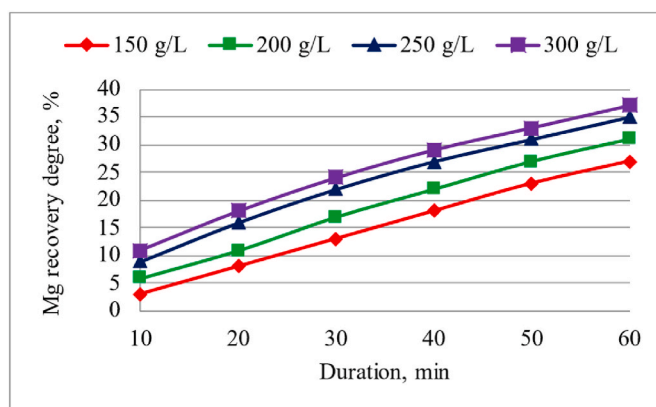


Fig. 4. The effects of HCl concentration and time on the Mg recovery degree.

concentration; specifically, 37% of magnesium is recovered after 60 min at an HCl solution concentration of 300 g/L. In order to evaluate the validity of our results, they were compared with those reported in other similar papers. For example, Yoo et al. [38] found that 39% of magnesium was recovered after 60 min at a solution concentration of 300 g/L using sulfuric acid as the leaching agent. This result is slightly higher than what has been obtained using hydrochloric acid as leaching agent; however, this difference can be attributed to the differences between HCl and H₂SO₄ solutions in terms of their chemical composition and reactivity towards different ores. A noteworthy recent publication authored by Gonzalez et al. [39] provides a comprehensive review of research that investigates the use of both organic and inorganic acids as leaching reagents. The authors meticulously analyze the relationship between reagent concentration and the efficiency of magnesium extraction. Their findings reveal that different acids can extract varying amounts of magnesium, ranging from 3% to 26% within a 1-h timeframe. Notably, sulfuric, hydrochloric, nitric, formic, and acetic acids demonstrate a descending order of effectiveness. However, it should be noted that although extraction with ammonium salts exhibits greater selectivity than acids, the overall degree of magnesium extraction is lower. Interestingly, alkaline solutions failed to dissolve magnesium, even after a 1-h leaching period. Based on their observations, the authors conclude that the leaching kinetics align with the unreacted core model, wherein the core gradually shrinks as the product layer develops. In a comparative leaching study conducted by Erlund et al. [40], various reagents, including hydrochloric acid, ammonium sulfate, and mixtures of ammonium bisulfate and ammonium sulfate, were utilized. Particularly notable were the findings regarding the effectiveness of ammonium bisulfate and bisulfate/sulfate mixtures in leaching serpentinite ores, resulting in recoveries surpassing 32%. Remarkably, ammonium bisulfate exhibited outstanding performance as a solvent for serpentine minerals, facilitating the recovery of over 32% magnesium within a 1-h timeframe. Moreover, the study revealed that up to 50% of the bisulfate component could be substituted with sulfate, yielding similar results. This suggests that utilizing pure ammonium bisulfate may not be necessary considering ammonium sulfate's availability as a more cost-effective alternative.

4.3. The effects of solid - liquid ratio and time

The ratio of liquid to solid phase during leaching is an important factor in determining the efficiency and effectiveness of the process [41]. This ratio affects the rate at which solutes are extracted from the raw materials, as well as their distribution between liquid and solid phases. In addition, it can influence factors such as mixing effect, heat transfer rates, and chemical reaction kinetics within a leaching system. Therefore, studying this ratio is essential for optimizing leaching operations and ensuring the desired outcome are achieved with maximum

efficiency.

The change in the ratio of solid to liquid from 1:2 to 1:5 (Table 3) has little effect on the extraction of magnesium into solution, which is consistent with findings reported in other studies. For example, a study by Yoo et al. [38] found that, when S:L ratios were decreased below 1:5, there was a reduction in magnesium extracted into solution and an increase in material flows and cost for evaporating this solution. Furthermore, another study conducted by Fedorocková et al. [42] showed similar results - decreasing S:L ratio below 1:5 led to reduced efficiency of extracting magnesium into solution and therefore increased the costs associated with evaporation processes. These findings suggest that reducing the S:L ratio beyond 1:5 may not be economically favorable due to reduced extraction efficiency as well as higher costs associated with additional evaporation processes.

4.4. Effect of stirring speed

Stirring speed during leaching can have a major impact on the rate of extraction and overall efficiency of the process. The mixing rate affects both the contact between solids and solution, as well as mass transfer rates within the system. Understanding how different stirring rates affect leaching processes is essential for optimizing leaching operations, minimizing energy costs, and ensuring that desired levels of product recovery are achieved. Bearing this reality in mind, the influence of stirring speed on the film thickness around asbestos wastes particles suspended in a reaction vessel has been studied. The results are presented in Fig. 5.

An increase in stirring speed reduces the diffusion layer thickness, thus improving mass transfer rates through the diffusion layer. Magnesium extraction rates can be used to distinguish between particle and film diffusion-controlled reactions, since intraparticle diffusion-controlled reactions have ion exchange rates that are independent of stirring speed. To further investigate this phenomenon, an experiment was conducted to examine magnesium recovery from an asbestos waste under varying stirring speeds (150–300 rpm) over 60 min. The results were then presented in Fig. 5., showing that increasing the agitation rate had a direct effect on metal leaching efficiency so that higher agitation rates resulted in more efficient extractions than lower ones. Thus, it is clear that adjusting stirring speeds can play an important role when attempting to maximize magnesium recovery from asbestos waste suspended within reaction vessels. Singh et al. [35] investigated the effects of varying agitation rates on metal leaching efficiency in a series of experiments involving different types of asbestos waste and found results similar to those presented here. In addition, research conducted by Demir et al. [43] also demonstrated stirring speed's importance for increasing magnesium extraction rate in an experiment with a specific type of asbestos-containing material. Comparing the findings in the present study to those obtained in previous studies can provide additional insight into how adjusting stirring speeds can yield more efficient extractions when attempting to maximize magnesium recovery from asbestos waste suspended within reaction vessels.

5. Comparative fitness of the kinetic models

Leaching of solid particles is a process which involves the removal of elements from raw materials through contact with a liquid phase. It can

Table 3

The effects of solid-liquid ratio on the recovery of magnesium.

S:L ratio	Mg recovery degree, %
1:2	29
1:3	36
1:4	37
1:5	37

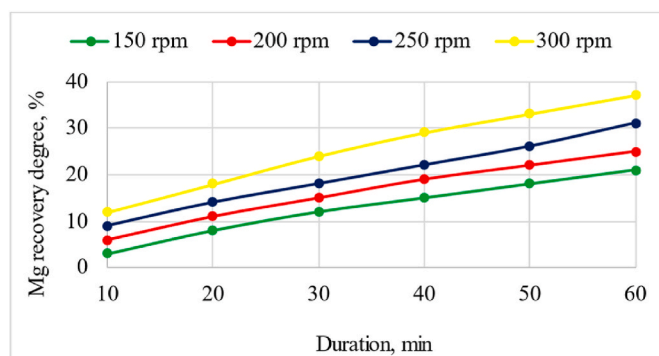


Fig. 5. The effects of stirring speed and time on the Mg recovery degree.

be described by various models and different mechanisms, such as diffusion through the liquid film, diffusion through the product layer, or chemical reaction. To identify the rate controlling step in leaching, all possible mechanisms must be tested. This can be done by plotting each respective equation in conversion-side versus time and then comparing their correlation coefficients to find out which one fits best to a studied equation line. The mechanism whose equation exhibits the highest linear relationship with time is considered as being rate controlling step. In addition, visual inspection may also help to determine which equation shows this linear relation most accurately.

The graph (Fig. 6a) obtained in processing data on the kinetic equation $1-(1-a)^{1/3} = k\tau + b$ shows the results of the leaching process at temperatures of 25, 50, 75 and 90 °C in relation to the duration (τ) of the leaching process. The y axis illustrates the data according to the expression $[1-(1-a)^{1/3}]^2$ and x axis represents τ which is a measure of time. The R^2 value for each temperature indicates that there is an unsatisfactory correlation between these two variables and it can be seen from the values (0.9805, 0.9864, 0.9801, and 0.9897, respectively). Such results indicate that the process does not proceed strictly by the tested kinetic model as expected; thus, necessitating further investigation into other equations like Yander diffusion equation, etc. to explain how this reaction proceeds under different temperature conditions over specific durations of time periods (τ). In order to compare the results obtained in this study with those of other similar papers, it is important to refer to relevant literature and consider the differences between their experimental data and those reported in the current study. For instance, one can look at studies [2] such as where similar leaching processes were conducted under different temperature conditions and durations. The comparison should focus on how the kinetic equations used in each study differ (e.g., Yander diffusion equation vs $1-(1-a)^{1/3} = k\tau + b$) and how these differences affect the exported results from the two experiments (Fig. 6a). This will help us better understand why there is an unsatisfactory correlation between these two variables seen in the experiments performed in the present study, as well as provide insight into more efficient methods for conducting future leaching studies.

When evaluating a chemical reaction's kinetics experimentally, temperature effects and acid concentration changes must be taken into account due to their significant influence on rate constants through alteration of intermolecular interactions. This can lead to discrepancies between experimental outcomes and those predicted theoretically using a single parameter analysis alone. Therefore, comprehensive analysis involving multiple parameters such as temperature effects and acid concentration changes should be undertaken before any definite conclusion regarding the reaction mechanism is reached.

The plots of $1-(1-a)^{1/3}$ versus $t^{0.5}$ demonstrate a good correlation between the two sets of data (Fig. 6b). This is evidenced by the values of correlation coefficients R^2 , which are 0.9994, 0.9953, 0.9936 and 0.9957, respectively, as shown in Fig. 6b. This phenomenon can be explained by Fick's law which states that diffusion rate is directly proportional to concentration gradient at any point in space or time (Fick's

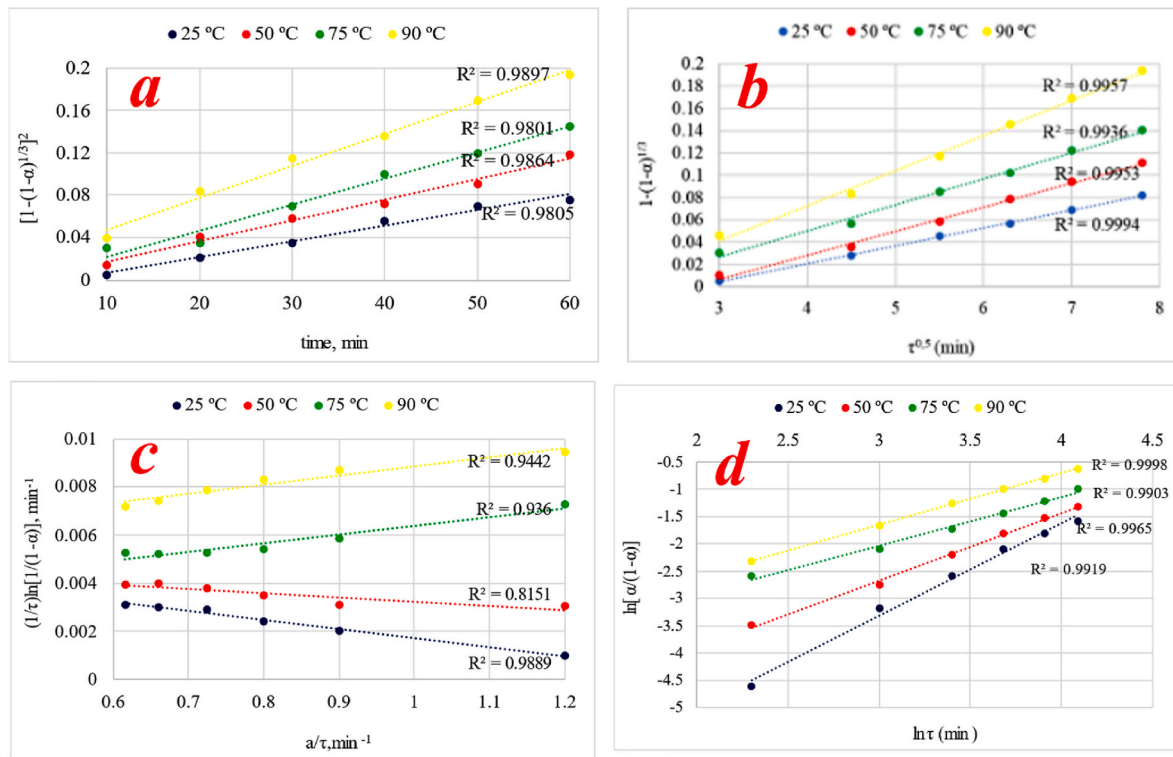


Fig. 6. Fitness of different kinetic equations with experimental data:

- (a) Plots of $[1-(1-\alpha)^{1/3}]^2$ versus dissolution time at different temperatures (kinetic model graphs);
 (b) Plots of $1-(1-\alpha)^{1/3}$ versus $\tau^{0.5}$ (min) at different temperatures (diffusion model graphs);
 (c) Plots of $(1/\tau)\ln[1/(1-\alpha)]$ versus a/τ (min⁻¹) at different temperatures (Drozdov-Rotinyan kinetic equation graphs);
 (d) Plots of $\ln[\alpha/(1-\alpha)]$ versus $\ln\tau$ at different temperatures (Prout-Tompkins kinetic equation graphs).

first law). In this case, the dissolution rate could be assumed to follow Fick's first law, since it involves transfer of solute through an interface where there exists a concentration gradient for mass transfer process to occur. The plot obtained for $1-(1-\alpha)^{1/3}$ suggests that the observed trend follows linearity over square root transformation which further reinforces its consistency with Fick's law as well as other empirical models such as Noyes Whitney equation [44].

In addition to being consistent with theoretical laws governing diffusional phenomena, this result also has practical implications for environmental remediation technologies involving leaching from asbestos waste deposits. An accurate understanding of these processes are helpful in making informative engineering decisions regarding efficient removal strategies while minimizing undesired effects on nearby ecosystems.

As it is known, the Drozdov-Rotinyan (4) and Prout-Tompkins (5) equations are used to process data from ore leaching processes. These two equations provide a comprehensive model for understanding the behavior of liquid-solid systems, which is essential for efficient extraction and utilization of resources. The use of these equations enables engineers to accurately predict the rate at which metals or other materials can be extracted from ores using chemical leaching methods.

The Drozdov-Rotinyan equation (4) provides insight into how various parameters influencing extraction efficiency change with time. It models kinetics associated with dissolution reactions taking place between solid surfaces and solutions containing dissolved metals or other elements being released during leaching operations. By incorporating information about reaction orders, activation energy values, bulk concentrations and surface properties this equation can help optimize conditions for maximum resource recovery rates while minimizing costs.

The Fig. 6c shows the relationship between $(1/\tau)\ln[1/(1-\alpha)]$ and a/τ , min⁻¹. The correlation coefficients (R^2) are 0.9889, 0.8151, 0.9360, and

0.9442 which indicate that there is a poor suitability between these two variables. This means that there is not much of a linear relationship between them as the data points do not align perfectly on one line. Also, it means that the mechanism and kinetics of the leaching process when data is poorly correlated with the Drozdov-Rotinyan equation (4) can be quite complex. The main factor that affects the rate of dissolution in this case is the surface area available for interaction between solids and liquids, as well as other factors such as temperature, acid concentration, and stirring speed. In addition to these parameters, there are a number of chemical processes which may come into play during leaching. It could also indicate that there are two parallel pathways involved in leaching; one being fast enough to provide significant amounts of material released from solids over time while another pathway takes longer to reach completion and, thus, providing lower concentrations at any given time point than would be expected using just one pathway alone. Therefore, determining what processes are occurring can help better understand why data is not correlating properly with existing equations used for predicting rates of dissolution.

As mentioned in section 3.1.4, the Prout-Tompkins equation (5) is an empirical model that describes the diffusion of solutes through a porous medium such as ore particles in a suspension. It takes into account factors like particle size, porosity, saturation level, and temperature gradient to calculate the diffusion coefficient (a measure of how quickly substances move through a material).

The diagram plots the relationship between $\ln[\alpha/(1-\alpha)]$ and $\ln\tau$ (min⁻¹) at different temperatures based on the Prout-Tompkins kinetic equation (Fig. 6d). The leaching experiments were conducted at 25, 50, 75, and 90 °C, respectively. By analyzing the data from these experiments, it was found that there is a strong correlation between $\ln[\alpha/(1-\alpha)]$ and $\ln\tau$ (min⁻¹), with R^2 values of 0.9925, 0.9965, 0.9973, and 0.9998, for the studied temperatures respectively, indicating a high degree of accuracy in this model's prediction of reaction rate as a

function of temperature. This suggests that, according to Prout–Tompkins kinetic equation (5), increasing temperature will increase reaction rate proportionally with respect to time taken for completion of reaction process. This is also known as Arrhenius' law or activation energy theory which states that increasing temperature increases molecular motion, thus resulting in increased reactant collision frequency leading to an overall faster rate of chemical reactions occurring within given period of time [45].

The reaction rate constant k at different temperatures obtained by fitting data into the Arrhenius equation (7) is used to model the diffusion control of hydrochloride acid through solid product layers at this time.

$$k = A \exp \frac{-E_a}{RT} \quad (7)$$

where:

A - is the frequency factor;

E_a - is apparent activation energy, (kJ/mol);

R - is gas equilibrium constant, $R = 8.314$ (J/mol);

T - is the absolute temperature, (K)

The Arrhenius graph for magnesium dissolution in the diffusion-controlled stage is a plot with $1000/K$ on the x axis and $\ln k$ on the y axis (Fig. 7). The graph displays a positive correlation between temperature and rate constant, indicated by a acceptable R^2 value of 0.9633. This indicates that, as temperature increases, so does the rate constant exponentially; in other words, when temperatures increase, the speed of magnesium dissolution also increases significantly. In general terms, this means that higher temperatures result in faster rates of reaction than lower ones.

The apparent activation energy of a process can be used to gain insight into the reaction mechanism. In this case, the magnesium dissolution process has an apparent activation energy of 19.8 kJ/mol, which confirms that it proceeds in a diffusion mode. This is because diffusion-controlled processes usually have higher activation energies. Diffusion processes involve molecules moving through space without undergoing any chemical transformation; therefore, they require more energy for their reactants to overcome the barrier between them and reach an activated complex state before reacting with each other. The value of 19.8 kJ/mol indicates that such mechanism is indeed the case for the magnesium dissolution process being studied here - confirming that it follows a diffusion mode.

If the leaching process proceeds in a diffusion mode, it means that the rate of chemical or mineral extraction from a solid is determined by the concentration gradient between two solutions. In this method, solutes are transferred from one solution to another as molecules diffuse through or barrier.

6. Conclusion

Using agitation leaching process, asbestos waste was successfully was recruited for extraction of magnesium. A comprehensive study was conducted to analyze the composition of asbestos waste, investigate optimal parameters for the leaching process (such as temperature, duration, solid to liquid ratio, and mixing rate), and analyze the thermodynamics and kinetics involved. The study found that increasing temperature resulted in higher dissolution rates and solubility degrees. It is recommended to use a temperature of 90 °C for efficient extraction and higher yields. The study also determined that a solid-to-liquid ratio of 1:3 is the most effective in terms of yield, concentration levels, cost-effectiveness, and quality balance. Keeping the mixing rate within a range of 300 rpm ensures optimal conditions for the dissolution process. The research revealed that the leaching kinetics of asbestos wastes in hydrochloric acid align with diffusion mode, with an apparent activation energy of 19.8 kJ/mol. This research provides valuable insights into how these factors interact and influence leaching rates, which can contribute to more effective management strategies for the disposal of

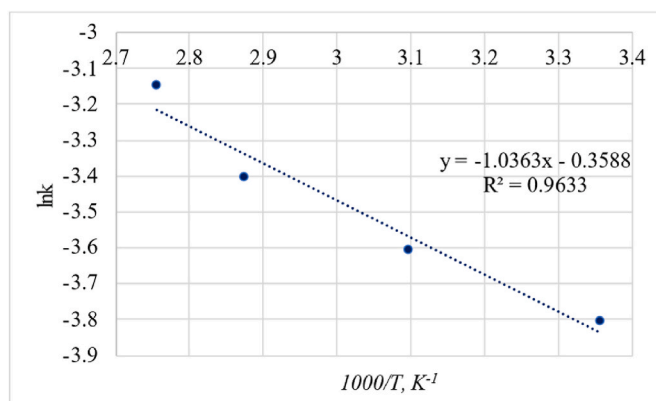


Fig. 7. Arrhenius plot for magnesium dissolution in the diffusion-controlled stage.

hazardous asbestos waste.

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CRediT authorship contribution statement

O.S. Baigenzhenov: Writing – review & editing, Conceptualization. **T.A. Chepushtanova:** Methodology. **A. Zh Altmysbayeva:** Data curation. **I.A. Temirgali:** Investigation. **G. Maldybayev:** Methodology, Investigation. **R.H. Sharipov:** Investigation, Data curation. **B.T. Altai-bayev:** Writing – original draft, Investigation. **A.T. Dagubayeva:** Visualization, Methodology, Data curation.

Declaration of competing interest

The authors declare that they have no known competing financial interests or personal relationships that could have appeared to influence the work reported in this paper.

Data availability

Data will be made available on request.

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