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TECHNICAL ARTICLE

DTPA-Assisted Selective Leaching of Mo, Co, Ni, and Al from Spent Hydrodesulfurization Catalysts

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The extraction of cobalt (Co), nickel (Ni), molybdenum (Mo), and aluminum (Al) from an alumina-supported hydrodesulfurization (HDS) spent catalyst was examined using diethylene triamine pentaacetic acid (DTPA) as a chelating agent. To assess the impact of different metals on leaching efficiency, a roasting pretreatment was performed on powdered catalyst samples at various temperatures (300–700°C) and durations (15–360 min). The morphological and textural modifications before and after roasting were characterized using scanning electron microscopy (SEM) and Brunauer–Emmett–Teller (BET) analysis. The optimal roasting parameters were determined to be 600°C for 180 min, under which the maximum metal extraction efficiencies were obtained: 71.05% for Mo, 80.06% for Co, 73.86% for Ni, and 15.33 % for Al. Leaching experiments were conducted with a particle size range of + 75 to − 30 μm, a liquid-to-solid ratio of 15 mL/g, a DTPA concentration of 0.2 M, a leaching temperature of 60°C, a duration of 180 min, and a stirring rate of 200 rpm. The findings highlight that both roasting temperature and time play a crucial role in enhancing metal dissolution from the spent catalyst.

INTRODUCTION

Highly selective solid catalysts are widely used in oil refinery plants to obtain high-quality and clean products.[1] These catalysts lose their catalytic activity as a result of the accumulation of various pollutants (impurities) on their surfaces during the process and become unusable after a while.[2] It is estimated that approximately 170×10^3 kg of catalysts are released as waste in the world every year.[3] These high amounts of waste catalysts, which are released both due to the metal content in their structure and the accumulation of other pollutants, especially sulfur, on the catalyst surface during refining processes, today pose a danger to the environment and human health. For this reason, the storage of waste catalysts, which are classified as hazardous waste, and the recovery of the metals in their structure, are very important.[4] Storage of waste catalysts is generally not preferred due to the need for large volumes and the damage they may cause to the environment. The number of studies on the recovery of metals from waste catalysts is increasing day by day in order to minimize environmental pollution and storage space requirements, and to meet the market demand for metals.[5–9] In the literature, there are studies in which pyrometallurgy, hydrometallurgy, and a combination of these two methods are applied together in the recovery of valuable metals from waste catalysts.[10–12] When these two methods are compared, the field of application of hydrometallurgical processes is much wider due to the low energy consumption and low gas emission compared to the pyrometallurgical method.[13] Therefore, leaching processes have been carried out by directly using various inorganic acids or alkaline solvents, such as nitric acid, hydrochloric acid, and sulfuric acid, of the waste catalyst.[1,2,14] On the other hand, in order to convert metal oxides in the complex structure of the waste catalyst into a more easily soluble form and to ensure that the metals pass into the solution

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with high efficiency, pretreatments such as oxidation and roasting can be applied.[15,16] In addition to the acids or bases mentioned above, complexing agents are also used as leaching reagents in metallurgical processes. As shown in Fig. 1, diethylene triamine pentaacetic acid (DTPA) is an organic compound containing four carboxylate and two amino groups, enabling it to form complex structures with metal ions. It is commonly used as a complexing agent in leaching processes.[17–20] Since DTPA has different affinity for metals, metals can be leached selectively by precipitating each metal at its own pH values. Studies have shown that DTPA–metal complex structures are thermodynamically stable and that DTPA has a higher tendency to form complexes, especially with cationic metals.[21,22] Therefore, it is possible to leach Mo, Co, Ni, and Al metals from alumina-based waste catalyst using DTPA.[23]

This study investigates the effects of roasting temperature (300°C, 400°C, 500°C, 600°C, and 700°C) and roasting time (15, 30, 60, 90, 120, 180, 240, and 360 min) on the extraction of Mo, Co, Ni, and Al from hydrodesulfurization (HDS) waste catalysts using DTPA as a complexing agent.

MATERIALS AND METHODS

Material Supply and Characterization

The spent HDS catalyst (Mo-Co-Ni/Al_2O_2) used in this study was sourced from a petrochemical facility in Romania in 2018. DTPA ($C_{14}H_{22}N_2O_{10}$), an analytical-grade complexing agent, was supplied by Merck. Before the experiments, the waste catalyst underwent an initial size reduction using a jaw crusher, followed by further grinding in a ball mill. The resulting powdered material was then sieved into distinct particle size fractions (20, 30, 75, 150, 300, and 600 μm). The prepared samples were dried at 105°C and stored in airtight plastic containers for subsequent use. The roasting process was carried out in a controlled-atmosphere furnace, maintaining a heating rate of 10°C/min, while varying the temperature (300–700°C) and roasting duration (15–240 min).

The chemical composition of the unroasted waste HDS catalyst samples, presented in Table I, was analyzed using microwave-assisted techniques. The surface morphologies of the samples, both before and after roasting, as well as the solid residues obtained after leaching, were examined using scanning electron microscopy (SEM; MAIA3 XMU;

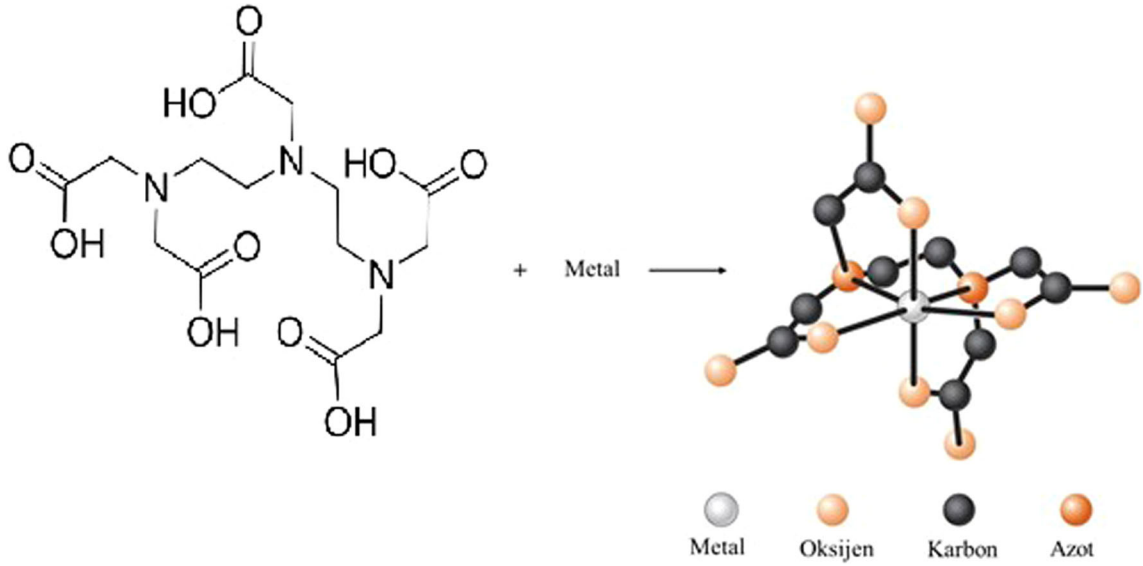

Fig. 1. DTPA–metal complex formation.

Table I. Characterization of the elemental composition of the waste HDS catalyst

Component (%)									
Al	**Mo**	**Co**	**Ni**	**Ca**	**C**	**S**	**P**	**Other elements**	**Loss of ignition**
37.94	9.35	2.18	1.72	0.34	13.7	0.73	0.28	0.051	33.709

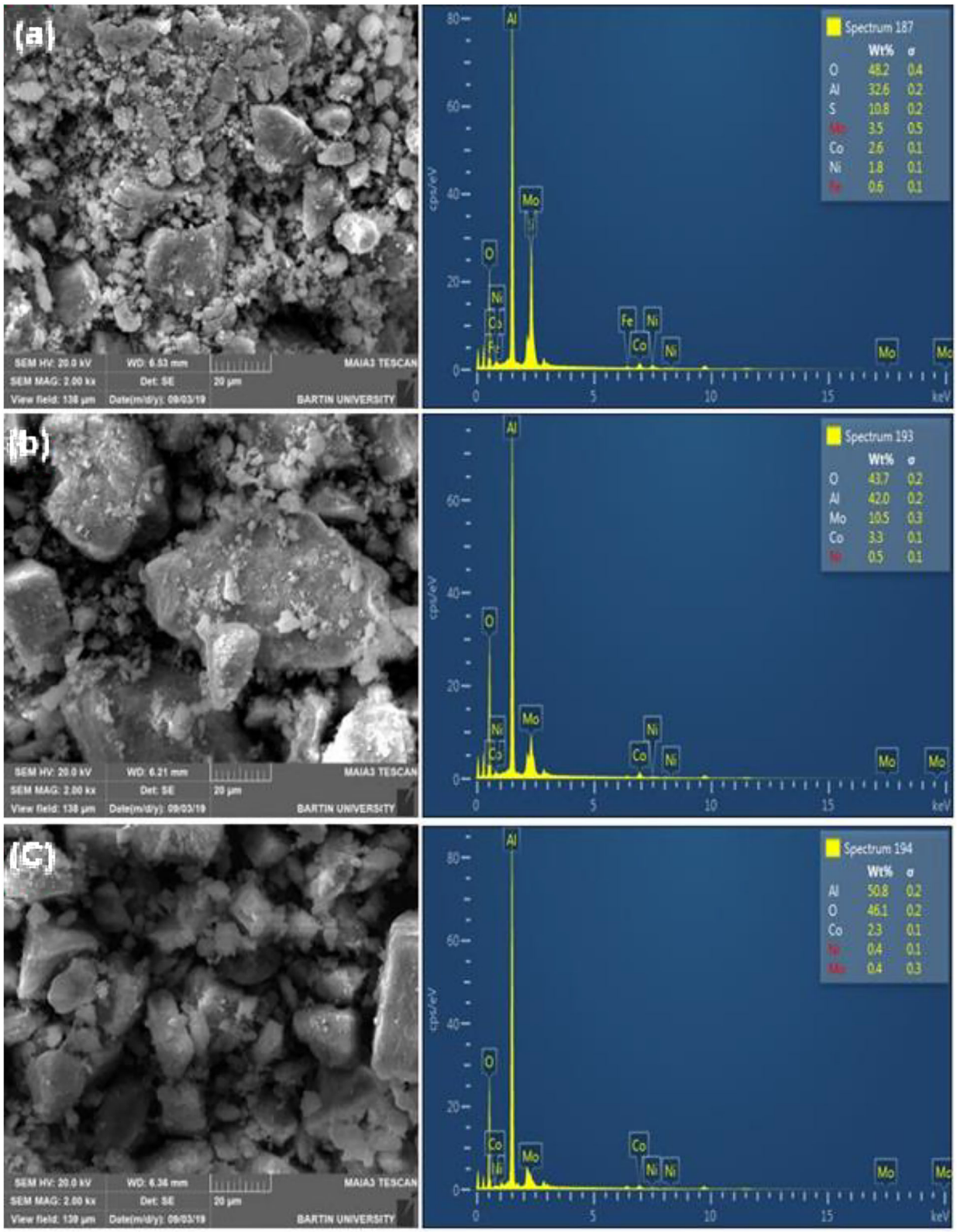

Fig. 2. SEM-EDS analysis results of (a) the unprocessed spent catalyst, (b) the roasted spent catalyst, and (c) the leaching residue of the spent catalyst.

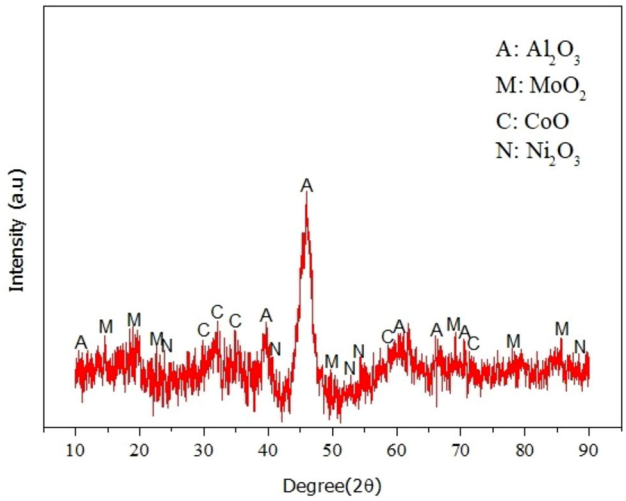

Fig. 3. XRD results of the roasted sample.

Tescan). As observed in the SEM results shown in Fig. 2, the particle sizes of the samples vary before and after roasting, and the distribution is not homogeneous. The SEM micrographs illustrate morphological changes (increased porosity, particle fragmentation) after roasting, which facilitate DTPA penetration. The literature, e.g., Liu et al.[35] showed similar structural changes in roasted catalysts enhancing leaching efficiency.

The surface area and pore volume of the powder samples before and after roasting were measured using the BET device. The surface areas of the unroasted and roasted samples were determined as 131.9 m^2/g and 198.3 m^2/g, respectively. On the other hand, the pore volume values of the roasted and unroasted samples were determined as 0.876 mL/g and 0.614 mL/g, respectively. According to the results, the surface area increases with the roasting process while the pore volume decreases. Although BET data (surface area, pore volume) are briefly mentioned, comparison with literature would enhance context. For example, Cheng et al.[36] showed that surface area increases up to a certain roasting temperature but declines if sintering occurs.

Figure 3 presents the XRD pattern of the roasted catalyst sample, but the main text lacks a detailed interpretation of this figure. The XRD analysis is crucial because it reveals phase transformations that occur during the roasting process, which directly affect the subsequent leachability of target metals (Mo, Co, Ni, and Al). Disappearance or reduction of MoS_2 peaks indicates that MoS_2 is oxidized to more soluble molybdates (e.g., MoO_3). The presence of spinel structures (e.g., $NiAl_2O_4$, $CoAl_2O_4$) or other mixed oxides suggests that some transition metals may become less soluble due to complexation. Peaks corresponding to γ-Al_2O_3 or amorphous alumina may remain, influencing Al extraction. As shown in Fig. 3, the XRD pattern of the roasted sample reveals the disappearance of MoS_2 diffraction peaks, suggesting successful oxidation of MoS_2 to MoO_3 or other molybdenum oxides during the roasting step. New diffraction peaks corresponding to $NiAl_2O_4$ and $CoAl_2O_4$ indicate the formation of spinel-type structures, which are known to be more resistant to leaching. This implies that roasting promotes both the liberation and stabilization of certain metal phases, which may have opposing effects on overall metal recovery efficiency. These findings are consistent with previous studies, such as those by Xie et al.[33] and Wang et al.[34] who reported similar transformations in spent HDS catalysts after thermal treatment.

Performance of Leaching Experiments

To assess the influence of roasting conditions on the dissolution efficiency of Mo, Co, Ni, and Al, powdered catalyst samples were subjected to roasting at different temperatures and durations. Leaching experiments were conducted in sealed 250-mL glass Erlenmeyer flasks, placed inside a temperature- and agitation-controlled incubator (ZCHENG 200D), following the process outlined in Fig. 4. A 0.2-M DTPA solution was first introduced into the flasks, and the initial pH of the solutions was recorded. The flasks were then transferred to an incubator, where the temperature was adjusted to the desired setpoint. Once the solution reached the target temperature, precisely measured powdered samples were added, and stirring was initiated. After the predetermined leaching period, the mixtures were filtered through filter paper. The resulting filtrates were collected in sealed plastic containers for pH measurement, and metal concentrations in the solutions were analyzed using atomic absorption spectrophotometry (Perkin Elmer AAnalyst-400) and ICP-MS (Agilent 7500ce). The leaching process was carried out under specific experimental parameters, including an DTPA concentration of 0.2 M, a particle size range of + 75 to − 30 μm, a leaching temperature of 60°C, a solid-to-liquid ratio of 15 mL/g, a leaching duration of 180 min, and a stirring speed of 200 rpm.

RESULTS AND DISCUSSION

Effect of Roasting Temperature

During the desulfurization processes in oil refinery plants, metal sulfate compounds are formed in the catalyst as a result of the accumulation of high amounts of sulfur on the Mo-Co-Ni/Al_2O_3 catalyst, which causes a decrease in catalytic activity. The metal sulfates formed are converted into metal oxides during the roasting process. The primary objective of applying roasting pretreatment to the samples is to eliminate the sulfur from the metal sulfate compounds by releasing sulfur dioxide (SO_2) gas under the influence of heat, and to transform the metal sulfates into metal oxides, which are more easily soluble.

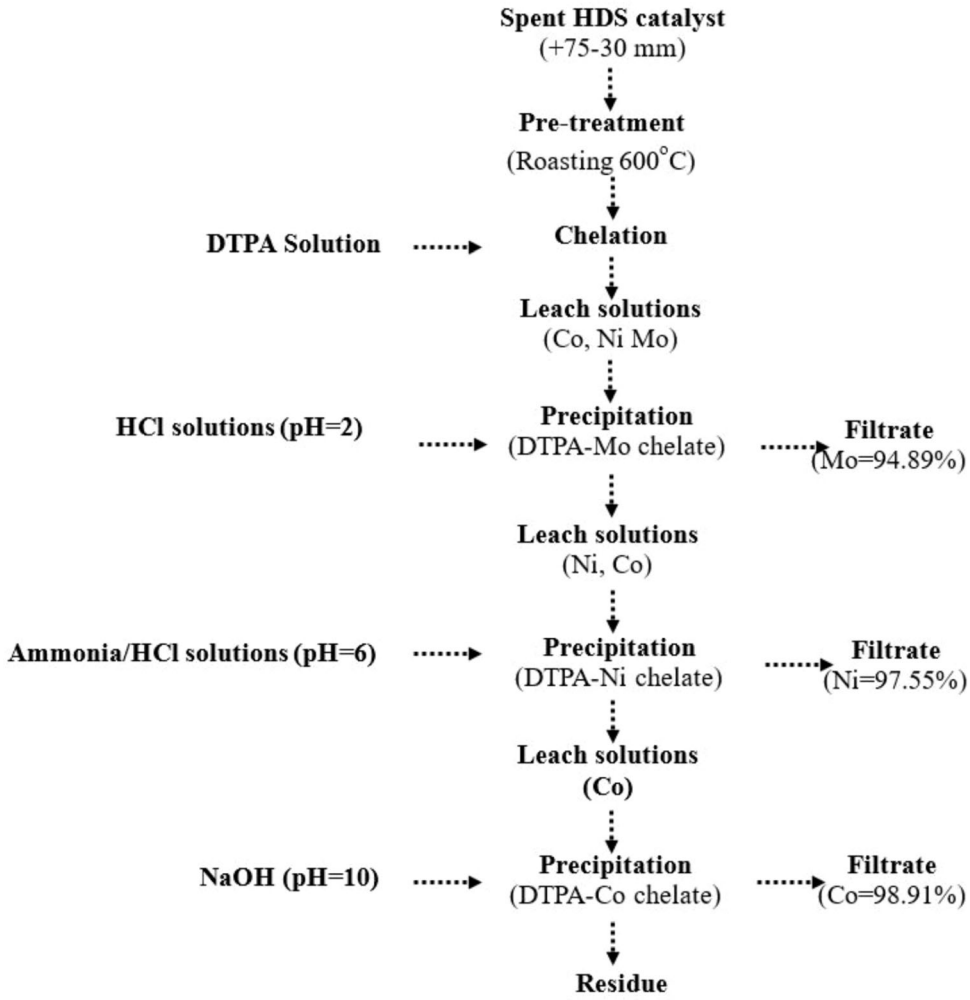

Fig. 4. The process of metal extraction through chelation.

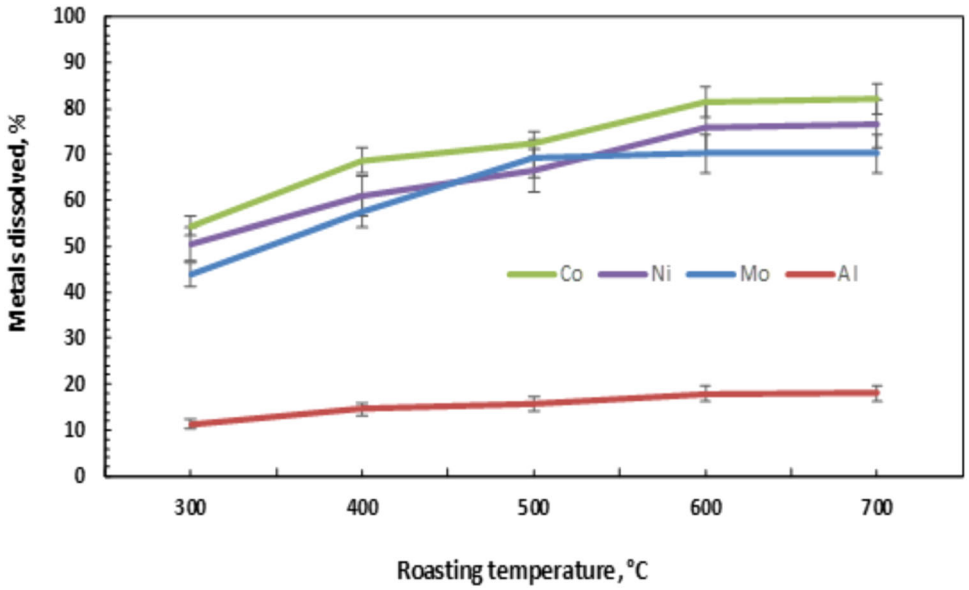

Fig. 5. Influence of roasting temperature on metal leaching efficiency: roasting temperature: 180 min; particle size: – 75 to + 30 μm; solid-to-liqid ratio: 12.5 mL/g; DTPA concentration: 0.2 M; leaching temperature: 20°C; external temperature: 60 min; agitation speed: 200 rpm.

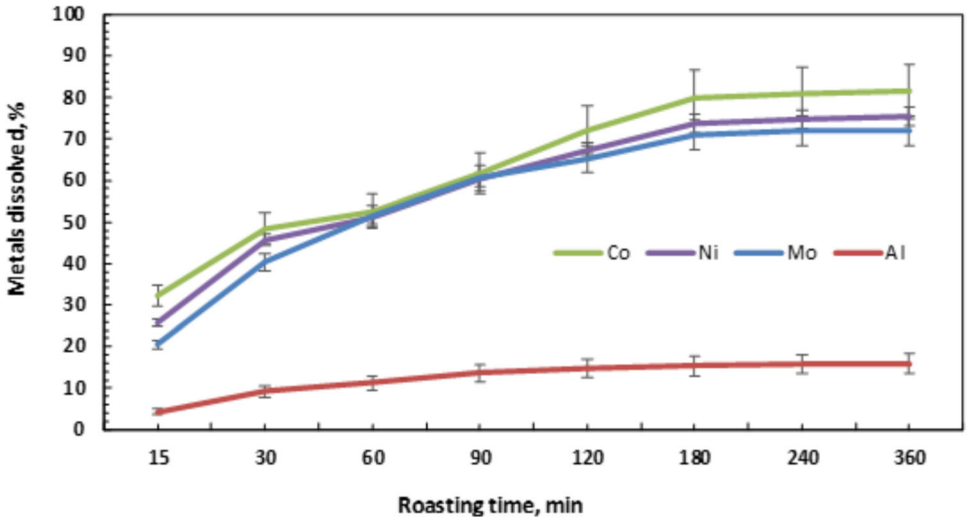

Fig. 6. Influence of roasting duration on metal leaching efficiency: roasting temperature: 600°C; particle size:− 75 to + 30 μm; liquid-to-solid ratio: 12.5 mL/g; DTPA concentration: 0.2 M; leaching temperature: 20°C; leaching duration: 60 min; stirring speed: 200 rpm.

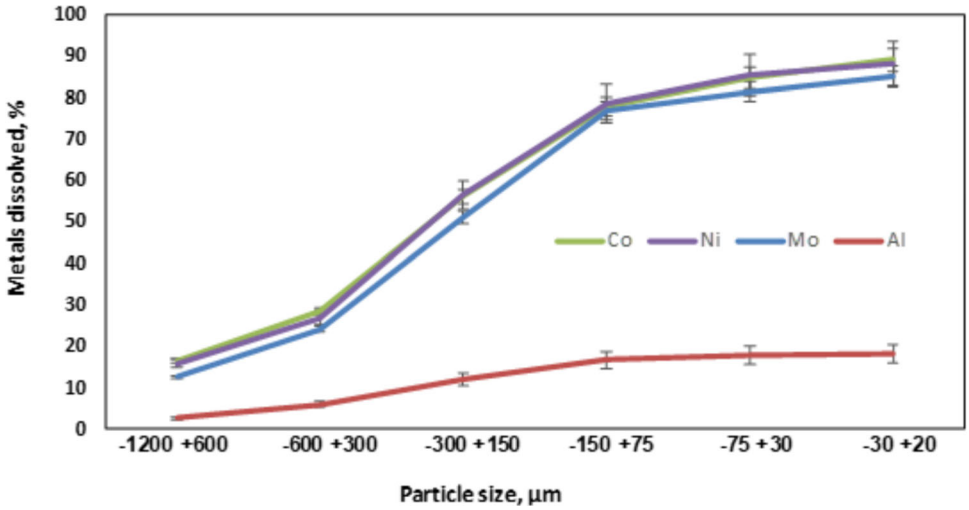

Fig. 7. The influence of particle size on metal dissolution: roasting temperature: 600°C;roasting time: 180 min; liquid-to-solid ratio: 12.5 mL/g; DTPA concentration: 0.2 M;leaching temperature: 20°C; leaching time: 60 min;agitation speed: 200 rpm.

The variation in metal dissolution rates with roasting temperature is presented in Fig. 5. The results clearly indicate that the extraction efficiency of all metals improves as the roasting temperature increases. The highest extraction values for Mo, Al, Co, and Ni were achieved at a roasting temperature of 600°C, with values of 70.22%, 17.99%, 81.32%, and 75.83%, respectively. At a roasting temperature of 700°C, no significant improvement in the dissolution rates of the metals was observed compared to the 600°C condition (Mo: 70.55%, Al: 18.46%, Co: 82.77%, and Ni: 76.99%).

Effect of Roasting Time

Figure 6 shows the results of the change in the dissolution rates of the metals with roasting time. Accordingly, a significant increase was observed in the extraction values of the metals up to 180 min of roasting time. At the end of 180 min of roasting time, it was determined that Mo, Al, Co and Ni metals passed into solution at 71.0%, 15.33%, 80.06% and 73.86%, respectively. No significant increase in the amount of metal leached into the solution was observed with longer roasting times (Mo: 72.55%, Al: 15.72%, Co: 81.99%, and Ni: 75.74%). Thus, the optimal roasting temperature and duration were established as 600°C and 180 min, respectively.

To assess the impact of particle size on metal dissolution efficiency, leaching experiments were conducted across two distinct particle size ranges:− 1200 to + 600 μm and − 30 to + 20 μm. The findings presented in Fig. 7 indicate a clear enhancement in leaching efficiency with decreasing particle size. Notably, reducing the particle size

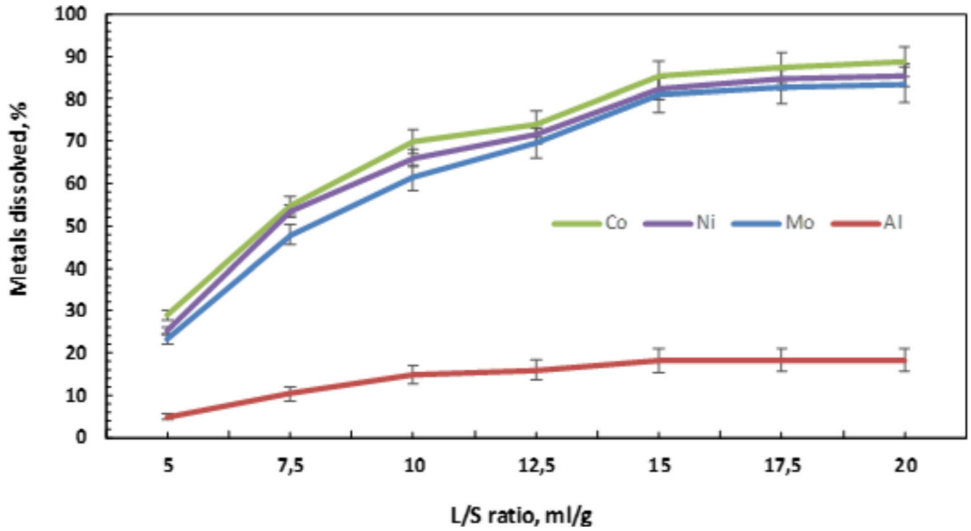

Fig. 8. The effect of liquid-to-solid (*L/S*) ratio on metal dissolution:roasting temperature: 600°C;roastnig time: 180 min;particle size: – 150 to + 75 μm; DTPA concentration: 0.2 M;leaching temperature: 20°C;leaching time: 60 min; agitation speed: 200 rpm.

from 150 μm to 30 μm led to a significant rise in the dissolution rates of Mo, Al, Co, and Ni, increasing from 12.55%, 2.77%, 16.99%, and 15.11% to 76.55%, 16.89%, 77.52%, and 78.57%, respectively. This enhancement in metal dissolution can be attributed to the increased rate of mass transfer that occurs as smaller particles provide a greater surface area for the solvent to interact with the active sites on the solid particle surface, leading to more efficient leaching.[24] However, for particle sizes smaller than 150 μm, no substantial improvement in the dissolution rates was observed. For instance, the dissolution rates at smaller particle sizes were Mo: 85.24%, Al: 18.39%, Co: 88.55%, and Ni: 88.74%. These findings suggest that, beyond a certain point, further decreasing the particle size does not result in a significant increase in dissolution efficiency. Similar outcomes were reported in a study focusing on the leaching of nickel from spent catalysts using EDTA,[25] where particle size was also found to influence the leaching efficiency, albeit with diminishing returns beyond a certain size reduction.

Figure 8 presents the results of experiments conducted at varying liquid-to-solid ratios (5, 7.5, 10, 12.5, 15, 17.5, and 20 mL/g) to explore the effect of this ratio on the extraction efficiency of Mo, Al, Co, and Ni from the spent HDS catalyst. As shown in the figure, the metal dissolution rates increased as the liquid-to-solid ratio was raised. Specifically, at a liquid-to-solid ratio of 15 mL/g, the leach efficiencies for Mo, Al, Co, and Ni were observed to be 80.99%, 18.24%, 85.43%, and 82.43%, respectively. However, when the liquid-to-solid ratio was increased beyond 15 mL/g, no substantial improvements were observed in the amount of metals leached into the solution. For instance, at higher ratios, the metal extraction values for Mo, Al, Co, and Ni were Mo: 83.57%, Al: 18.46%, Co: 88.11%, and Ni: 85.03%. This plateau in the extraction efficiency can likely be attributed to the limited availability of solvent to effectively dissolve the metals in the leach medium, which in turn limits the amount of DTPA present. As a result, the amount of DTPA becomes a constraining factor for further increasing the extraction efficiency, preventing significant improvements in metal dissolution. The trends observed in this study align with those of a previous investigation by Arslanoğlu and Altundoğan (2013), who reported similar findings regarding the impact of liquid-to-solid ratios on leaching efficiency. Additionally, the experimental data corroborate the results obtained by El-Okazy and colleagues,[26] supporting the conclusion that, beyond a certain point, increasing the liquid-to-solid ratio does not significantly enhance metal extraction.

Figure 9 presents the results of leaching experiments conducted using varying concentrations of DTPA (0.025, 0.05, 0.1, 0.15, 0.2, and 0.25 M) as the complexing agent. Since the reaction rate is dependent on the concentration of reactants, it is anticipated that an increase in the concentration of DTPA would lead to a corresponding increase in the dissolution rate. As expected, the data in Fig. 9 clearly indicate that the dissolution rates of all the metals improved as the concentration of DTPA was increased. The metal extraction efficiencies gradually rose with increasing DTPA concentration, reaching their peak at 0.15 M. At this concentration, the leaching efficiencies for Mo, Al, Co, and Ni were found to be 83.26%, 18.63%, 89.13%, and 87.60%, respectively. However, when the DTPA concentration was further increased beyond 0.15 M, no significant improvements in the amount of metal dissolved into the solution were observed. At higher concentrations (0.2 M and 0.25 M), the extraction values were Mo: 89.57%, Al: 19.54%, Co: 95.27%, and Ni: 92.13%, indicating that the increase in

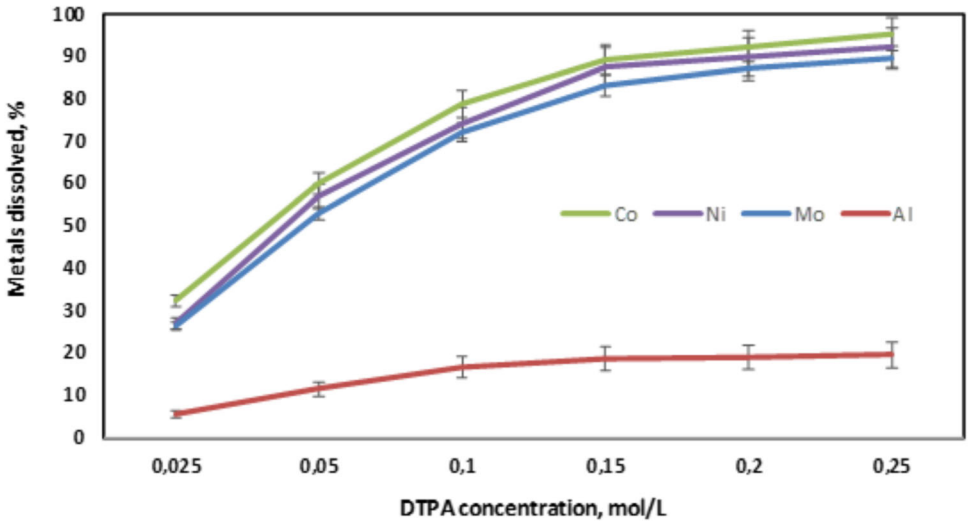

Fig. 9. The impact of DTPA concentration on metal dissolution: roasting temperature: 600°C; roasting time: 180 min; particle size:– 150 to + 75 μm; liquid-to-solid ratio: 15 mL/g; leaching temperature: 20°C; leaching time: 60 min;agitation speed: 200 rpm.

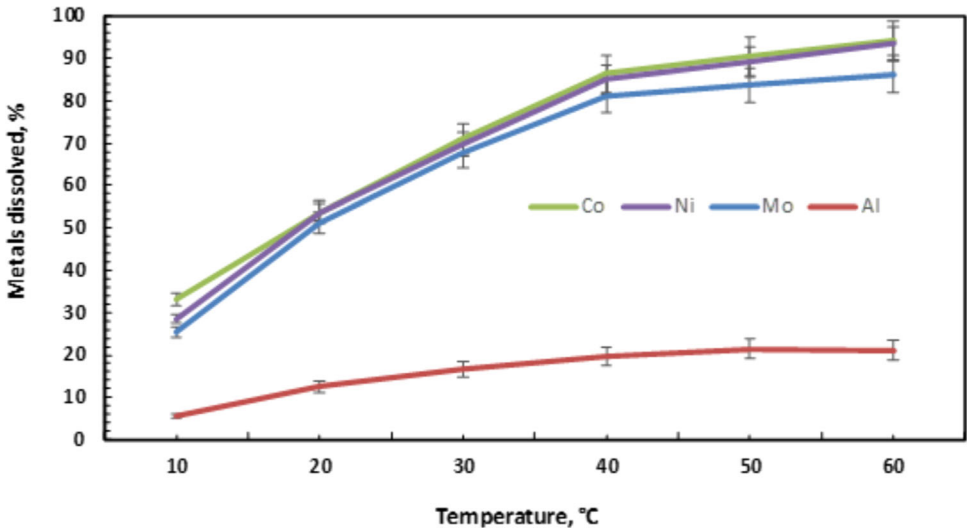

Fig. 10. The effect of leaching temperature on metal dissolution: roasting temperature: 600°C; roastine time: 180 min; particle size: – 150 to + 75 μm; liquid-to-solid ratio: 15 mL/g; DTPA concentration: 0.15 M; leaching time: 60 min; agitation speed: 200 rpm.

metal dissolution had plateaued. These results suggest that, while higher DTPA concentrations initially boost the dissolution rates, there is a threshold beyond which additional increases in DTPA concentration do not lead to further enhancement in metal extraction. This pattern aligns with findings from other studies in the literature, which also report similar trends regarding the effect of DTPA concentration on the dissolution rates of metals.[25−29] The data emphasize the importance of optimizing DTPA concentration to maximize metal recovery without unnecessary excess.

To examine the effect of temperature on the efficiency and kinetics of the leaching process, experiments were conducted at six different leaching temperatures: 10, 20, 30, 40, 50, and 60°C. The results clearly demonstrate that, as both leaching temperature and time increased, the metal extraction values also showed a significant rise. As illustrated in Fig. 10, the dissolution percentages for Mo, Al, Co, and Ni metals increased rapidly during the first 60 min of the leaching process, reaching 87.57%, 20.46%, 95.96%, and 94.13%, respectively. Beyond 60 min, however, there was no substantial increase in the metal extraction rates, with the dissolution percentages being Mo: 90.10%, Al: 21.12%, Co: 97.03%, and Ni: 94.27%. This rapid increase in metal extraction during the first 60 min can be attributed to the dissolution of the metal oxides present in the catalyst. The slower reaction rate observed after 60 min is likely due to the presence of metal sulfides and elemental sulfur within the catalyst structure, which does not fully dissolve even after the roasting pretreatment.[30]

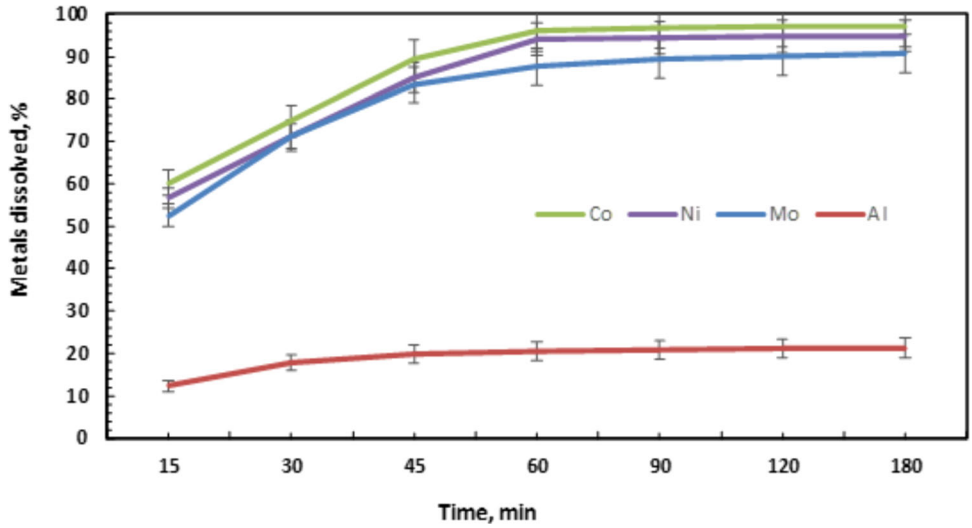

Fig. 11. The effect of leaching time on metal dissolution: roasting temperature: 600°C; roasting time: 180 min; particle size: − 150 to + 75 μm; liquid-to-solid ratio: 15 mL/g; DTPA concentration: 0.15 M; leaching temperature: 40°C; agitation speed 200 rpm.

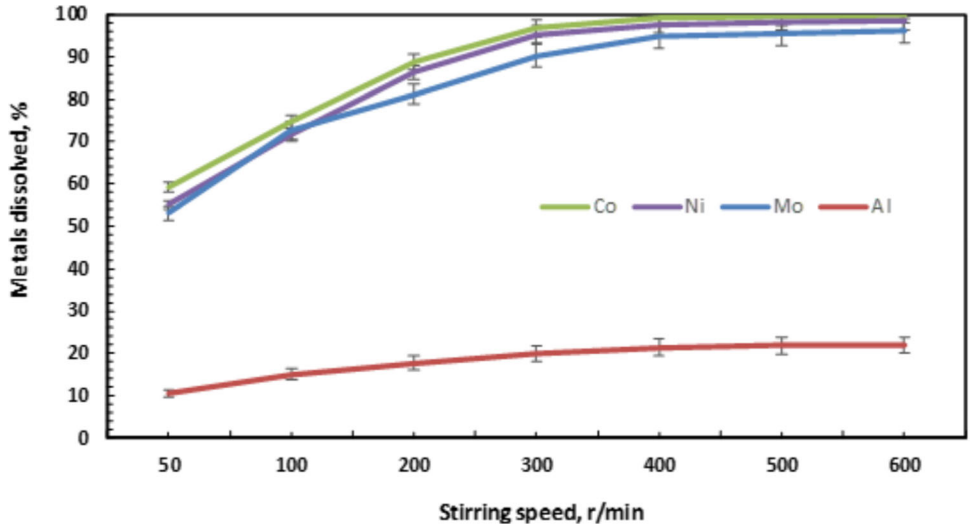

Fig. 12. The effect of stirring speed on metal dissolution: roasting temperature: 600°C; roasting time: 180 min; particle size: − 150 to + 75 μm; liquid-to-solid ratio: 15 mL/g; DTPA concentration: 0.15 M; leaching temperature: 40°C; leaching time: 60 min.

Furthermore, when the results shown in Fig. 11 were examined, it was observed that, at a leaching temperature of 40°C, the dissolution rates for Mo, Al, Co, and Ni were 81.75%, 19.14%, 86.23%, and 85.21%, respectively. However, when the leaching temperature was increased above 40°C, no significant improvement in metal extraction was noted, with the dissolution values at 50°C and 60°C being Mo: 86.01%, Al: 21.75%, Co: 94.51%, and Ni: 93.03%. Based on these findings, it was concluded that the optimal leaching temperature and time for the subsequent experiments would be 40°C and 60 min, respectively, as this combination provided efficient metal recovery without any substantial benefit from further increasing the temperature or extending the leaching duration.

The results of the experiments investigating the effect of stirring speed on the metal dissolution efficiencies are presented in Fig. 12. The data clearly indicate that the extraction efficiencies of Mo, Al, Co, and Ni metals increased as the stirring speed was raised from 50 rpm to 300 rpm. At a stirring speed of 600 rpm, the extraction efficiencies of Mo, Al, Co, and Ni metals were found to be 99%, 98%, 96%, and 20%, respectively. This improvement in extraction efficiency can be attributed to the reduction in the boundary layer thickness around the solid particles as the stirring speed increases. The thinner boundary layer allows for more efficient contact between the solid particles and the solvent, which enhances the dissolution of metals. However, it was observed that no significant improvement in

metal extraction occurred at stirring speeds higher than 300 rpm. This suggests that a stirring speed of 300 rpm is sufficient to ensure effective mixing and contact between the solid particles and the solvent, beyond which there is little benefit. This trend is consistent with findings in similar studies in the literature. For instance, a study examining the solubility of Mo and Co from HDS catalysts using sulfuric acid (H2SO4) reported that metal extraction efficiency increased with stirring speeds up to 500 rpm. However, the study also indicated that there was no substantial increase in the amount of dissolved metal at higher stirring speeds.[24,30] This suggests that, while higher stirring speeds can improve the dissolution process up to a certain point, there is a limit beyond which further increases in stirring speed do not significantly affect the extraction efficiency.

When the chelating agent (L-5) is introduced at the solid–liquid interface during the metal extraction process, a metal–chelate complex (Mi-L-2) forms on the catalyst surface. This complex then transitions from the catalyst surface into the solution, as illustrated in Eq. 1. In this reaction, {[HDS]-O} refers to the active sites on the catalyst surface that interact with the chelating agent. The term Mi + 2 represents the metal ion that binds with the ligands from the chelating agent, forming a stable complex. Additionally, Mii + 2 signifies the other metals that are present in the solution, which may also participate in the dissolution process. When metal oxides and hydroxides are simultaneously present in the catalyst structure, they can undergo partial dissolution when they interact with the chelating agent. This occurs due to the ability of the chelating agent to break down these metal oxides and hydroxides, facilitating the extraction process. The corresponding chemical reactions that describe this partial dissolution are shown in Eqs. 3 and 4. These equations demonstrate how the chelating agent (L-5) not only forms complexes with the metal ions but also contributes to the dissolution of metal oxide and hydroxide precipitates, thereby enhancing the overall metal extraction efficiency. This process highlights the role of the chelating agent in breaking the strong bonds between the metal ions and the solid catalyst, allowing for improved metal recovery from the catalyst surface. The dissolution mechanism facilitated by the chelating agent is crucial in the extraction of metals, especially when they are present in more stable, insoluble forms such as oxides and hydroxides. This concept is well-documented in the literature, particularly in studies that explore the interaction between chelating agents and metal-bearing catalysts:[31,32]

$$\{[\mathrm{HDS}]-\mathrm{O}\}-\mathrm{M_i^{+2}}+\mathrm{L^{-5}}+\mathrm{H_2O} \leftrightarrow \mathrm{HDS}-\mathrm{OH}+\mathrm{M}_i-\mathrm{L^{-2}}+\mathrm{OH^-} \quad (1)$$

$$\mathrm{M_i(OH)_2}+\mathrm{L^{-5}} \leftrightarrow \mathrm{M_i}-\mathrm{L^{-2}}+2\mathrm{OH^-} \quad (2)$$

$$\mathrm{M_iO}+\mathrm{L^{-5}}+\mathrm{H_2O} \leftrightarrow \mathrm{M_i}-\mathrm{L^{-2}}+2\mathrm{OH^-} \quad (3)$$

The formation of metal–DTPA complex structures on the catalyst surface can be dynamic, as these complexes can undergo exchanges with other metals that are adsorbed on the catalyst surface. This exchange process can be represented in Eqs. 4 and 5, where the metal– DTPA complex (Mi- DTPA) is either replaced by other absorbed metals or can be reabsorbed by the surface, leading to the formation of new complex structures. The stability of the metal- DTPA complex and the specific bonding interactions between the metal ions (Mi + 2) and the chelating agent play a key role in determining whether this exchange occurs. In addition to the formation of metal–DTPA complexes, the complex structures can also be reabsorbed onto the catalyst surface (Eq. 3). The activity of the metal ions in the solution, represented as Mii + 2, also influences the reabsorption and exchange processes. If the metal ions in the solution have a higher activity or stability, they can displace the original metal ions from the DTPA complex, leading to a change in the complex composition. The effectiveness of this exchange and reabsorption process is governed by several factors, including the stability constant of the metal–DTPA complex, the concentration of metal ions in the solution, and the relative affinity of the catalyst surface for different metal ions. The chelating agent's ability to form strong complexes with metal ions is crucial in facilitating these reactions, which can lead to the selective extraction or reabsorption of specific metals. In summary, these processes—exchange, reabsorption, and formation of new complexes—are central to the metal extraction mechanism. The reactions are influenced by the stability of the metal–DTPA complex, the bonding capacity of the metal ions (Mi + 2), and the activity of the other metals in the solution (Mii + 2). These interactions contribute to the overall efficiency of the extraction process, determining which metals are more readily extracted from the catalyst surface:

$$[\mathrm{HDS}]-\mathrm{OH}+\mathrm{M_i}-\mathrm{L^{-2}} \leftrightarrow [\mathrm{HDS}]-\mathrm{L}-\mathrm{M_i}+\mathrm{OH^-} \quad (4)$$

$$[\mathrm{HDS}]-\mathrm{L}-\mathrm{M_i}+[\mathrm{HDS}]-\mathrm{O}-\mathrm{M_{ii}^+} \leftrightarrow [\mathrm{HDS}]-\mathrm{L}-\mathrm{M_{ii}}+[\mathrm{HDS}]-\mathrm{OH}+\mathrm{M_i^+} \quad (5)$$

$$[\mathrm{HDS}]-\mathrm{L}-\mathrm{M_{ii}}+\mathrm{OH^-} \leftrightarrow [\mathrm{HDS}]-\mathrm{OH}+\mathrm{M_{ii}}-\mathrm{L^{-2}} \quad (6)$$

CONCLUSION

This study investigated the impact of various roasting temperatures and times on the selective leaching of Mo, Co, Ni, and Al from hydrodesulfurization (HDS) waste catalyst, utilizing DTPA (diethylene triamine pentaacetic acid) as a complexing agent. The objective was to optimize the conditions under which each of the metals could be selectively leached from the catalyst. The selective leaching process was carried out at pH values that were carefully chosen to favor the precipitation of specific metals. In particular, Mo, Ni, and Co were effectively precipitated at pH values of 2, 6, and 10, respectively, ensuring that each metal could form its corresponding precipitate under the desired conditions. The study's results revealed that the optimum roasting temperature and time for maximizing metal extraction were 600°C and 180 min, respectively. Under these conditions, the highest extraction efficiencies were obtained for the metals of interest. Specifically, the metal extraction yields were 71% for Mo, 80.06% for Co, 73.86% for Ni, and 15.33% for Al. The results from the experiments clearly demonstrate that both roasting temperature and roasting time play crucial roles in determining the overall metal extraction yields. The roasting temperature significantly influences the degree of metal dissolution, while the roasting time determines the extent to which metal oxides and sulfides can be effectively broken down to facilitate leaching. These findings underscore the importance of carefully controlling these parameters to achieve optimal metal recovery from HDS waste catalyst. Additionally, the study highlights the effectiveness of DTPA as a complexing agent in selectively extracting valuable metals from complex catalyst materials.

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AUTHOR CONTRIBUTIONS

Feride N. Türk: Writing, Acquisition of data, Analysis of data, Approval of the version of the manuscript to be published, review and editing, Hasan Arslanoğlu: Writing, Revising the manuscript critically for important intellectual content, Approval of the version of the manuscript to be published, review and editing.

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DATA AVAILABILITY

The data that support the findings of this study are available from the corresponding author, upon reasonable request.

CODE AVAILABILITY

Not applicable.

CONFLICT OF INTEREST

The authors declare that there are no conflicts of interest regarding the publication of this paper.

CONSENT FOR PUBLICATION

Not applicable.

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