Research Article

Recovery and Enrichment of Platinum from Spent Al$_2$O$_3$ Carrier Catalysts by Matte Smelting-Acid Leaching Process

Lida Sun [ID],[1,2,3] **Yan Jiang**,[1,2,3] **Mengyang Huang**,[1,2,3] **and Xingxiang Fan** [ID][1,2,3]

[1]*School of Chemistry and Resources Engineering, Honghe University, Mengzi 661199, Yunnan, China*
[2]*Engineering Research Center for Clean Extraction and High Value of Waste Precious Metal Catalysts of Yunnan Province Higher Education Institutions, Mengzi 661199, Yunnan, China*
[3]*China Renewable Resources Industry Technology Innovation Strategic Alliance "Joint Innovation Laboratory for Recycling Waste Precious Metals Catalyst", Mengzi 661199, Yunnan, China*

Correspondence should be addressed to Xingxiang Fan; fanxingxiang@uoh.edu.cn

Recovery of platinum from spent catalysts can meet the comprehensive recycling of secondary resources. Recovery and enrichment of platinum from the spent Al$_2$O$_3$ carrier catalyst using matte smelting for the pyrite-acid leaching process were studied. In melting process, the effects of pyrite, CaO, quartz sand, and Na$_2$B$_4$O$_7$ dosage on the recovery rate of Pt were investigated. In leaching process, the effect of H$_2$SO$_4$ concentration, temperature, time, and liquid/solid (L/S) ratio on Pt enriching efficiency was also investigated. The slag, iron matte, and leaching residue were analyzed using XRD. The molten platinum content in the slag was measured using an atomic absorption spectrometer. The experimental results showed that 98% recovery of platinum was obtained under the following optimum conditions: 1400°C temperature, 50 min smelting time, 1 : 2 mass ratio of pyrite to spent catalysts, 0.5 : 1 ratio of CaO to spent catalysts, and 2 : 1 ratio of quartz sand to the raw material with optimum conditions, which include 15% dosage of Na$_2$B$_4$O$_7$. Moreover, the acid leaching was more efficient under the following optimum conditions: H$_2$SO$_4$ concentration of 1.8 mol/L, time of 70 min, temperature of 60°C, and L/S ratio of 10 : 1. The enrichment and recovery of Pt were achieved at the disappearance of FeS during the acid-leaching process.

1. Introduction

Platinum is one of the most widely used platinum group metals with a high catalytic activity advantage, which is widely used in petroleum, automobile, and pharmacy industry [1, 2]. As technology has improved, platinum consumption has increased, resulting in scarcity of platinum resources. The resources are often in the form of paragenetic ore dispersed in nature. The resources of platinum group metals are extremely unevenly distributed, being concentrated in South Africa, Russia, the United States, and Canada. As a result of these few reserves, the supply of Pt to other parts of the world is restricted. Platinum is a crucial catalytic element in the fluid catalytic cracking petrochemical conversion process; it is also used in catalytic dehydrogenation, reforming, and hydrogenation processes of petroleum refining [3–5]. The catalytic

performance of platinum/rhenium Al$_2$O$_3$ catalyst in the previously mentioned processes gradually decreases. The gradual decrease of catalytic properties is due to catalyst poisoning, which results from sulfidation, carbon deposition, PGM oxidation, and Al$_2$O$_3$ support structural change, and crystallite size changes. Thus, the catalyst becomes ineffective and is eventually deactivated in the process. As the catalyst becomes ineffective and scrap, a large number of by-products, such as spent automobile catalyst [6, 7] and petroleum catalysts [8], are generated. From the point of view of resource reuse and environmental protection, the recovery of platinum from spent catalysts has both important economic and social benefits. China has limited platinum resources, which account for 0.6% of the world's total reserves. Therefore, the development of platinum advanced recovery and recycling technologies from spent catalysts is highly necessary.
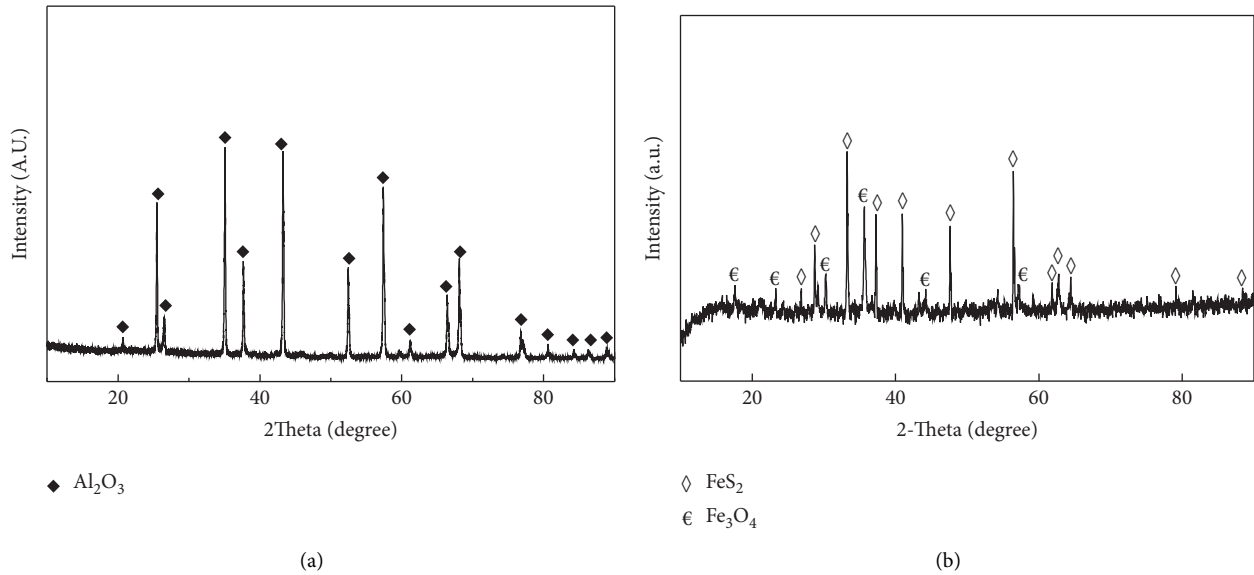
FIGURE 1: XRD pattern of (a) spent Al_2O_3 carrier and (b) Pt/Re catalyst and pyrite.

Pyrometallurgical and hydrometallurgical recovery methods have been used to recover precious metals from spent Al_2O_3 carrier catalysts. Pyrometallurgical processes are usually used to trap platinum since platinum is present in very low levels in spent catalysts. In this process, spent catalysts are crushed and mixed with trapping agents, slagging agents, and reductants and are smelted in medium frequency and electric furnaces. The carrier material is oxidized or directly transformed into the slag phase, while the platinum forms an alloy of metal phases. Platinum can be trapped with several trapping agents such as copper [9], lead [10], iron [11, 12], and nickel matte [13, 14]. Pt capture has been applied on a large scale by using the eutectic copper method in Tanaka Precious Metals in Japan and in some Chinese companies which has been employed in large-scale applications by Tanaka Precious Metals in Japan and China [15, 16]. Molten iron collection is performed using plasma and electric arc furnaces in the operating temperature range of 1600 to 2000°C. The high temperatures lead to the reduction of silicon and the formation of iron-silicon alloys. Although iron-silicon alloys are difficult to dissolve, silicon is easily generated, resulting in less efficient recovery of PGMs [17]. In recent years, matte capture has also been commonly used to recover PGMs [18]. The capturing matte efficiency depends on the nature of the slag and temperature. The hydrometallurgical process recovers platinum by leaching the carrier or platinum into an acidic or alkaline solution. However, this method has numerous drawbacks, such as wastewater generation, leaching material cost, poor adaptability, and high recovery time of platinum material [5].

In this study, we proposed a matte smelting-acid leaching process to efficiently recover Pt from spent Al_2O_3 carrier catalysts with consideration of environmental impacts. Pyrite was cracked to form FeS, which can effectively capture platinum at a relatively low temperature (1400°C) and prevent the formation of iron-silicon alloys. The effect of pyrite, CaO, quartz sand, CaF_2, and $Na_2B_4O_7$ dosages and

metal basicity on Pt recovery rate were investigated. Then, the trapping efficiency of FeS by leaching, using sulfuric acid along with the effect of H_2SO_4 concentration, temperature, time, and liquid/solid (L/S) ratio on Pt enriching efficiency, was studied. Pyrite can easily be obtained as a trapping agent with a high collection efficiency. This process has advantages such as simple process, simple operation, high enrichment ratio of Pt, and low technical difficulty.

2. Methodology

The experimental raw material was spent Al_2O_3 carrier Pt/Re catalysts. The content of Pt in the spent catalysts was analyzed using ICP-OES of 1780 g/t. The trapping agent used was pyrite. X-ray diffraction (XRD) pattern of the spent Al_2O_3 carrier Pt/Re catalyst is shown in Figure 1(a). The main component of the spent Al_2O_3 carrier catalyst was Al_2O_3 of high melting point. But the platinum can be discovered by SEM-EDS in the spent Al_2O_3 catalyst. From Figure 2(b), it can be seen that the spent Al_2O_3 catalyst mainly contains the oxygen and aluminium. The platinum element was also found in catalyst, which is very small. In Figure 1(b), the main components of the pyrite were FeS_2 and a small amount of Fe_3O_4.

All chemical reagents used in the leaching experiments of this study were analytically pure. All raw materials were dried at 375 K for two hours to remove moisture before melting. The smelting equipment used was an intermediate frequency furnace (LSZ-110). The graphite crucibles of size $\Phi80 \, mm \times 130 \, mm$ were used for the collected melting materials. The FeS traps, molten slag, and leaching residue phase compositions were analyzed using X-ray diffraction (X'pert3 Powder). The molten platinum in the form of slag was measured using an atomic absorption spectrometer (Type: Varian).

Figure 3 shows a comprehensive process for the recovery of platinum from spent Al_2O_3 carrier catalysts. The process includes five sections: fine grinding and mixing of the catalyst,
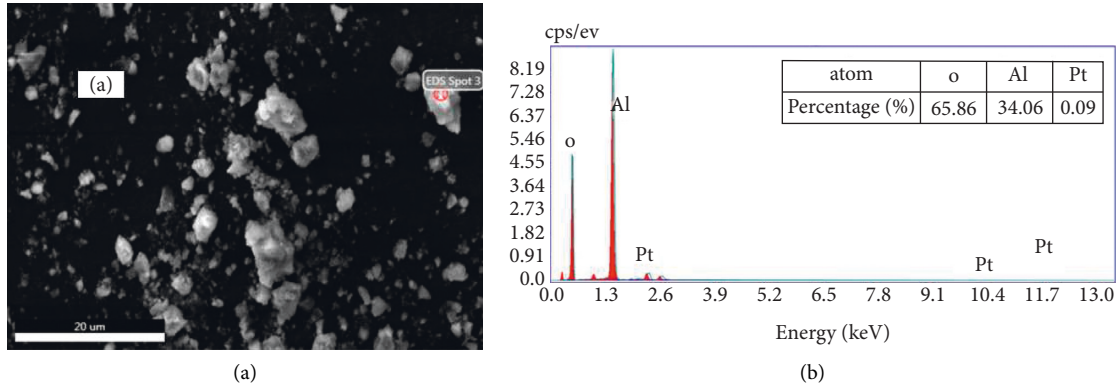
FIGURE 2: (a) SEM image and (b) EDS spectrum of spent Al_2O_3 carrier Pt/Re catalyst.
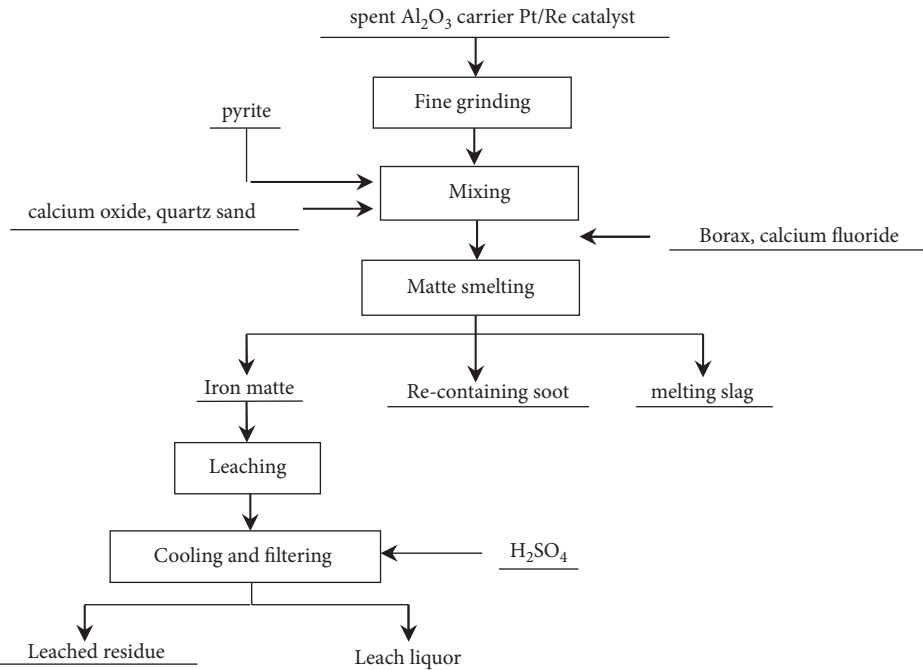

FIGURE 3: The process for the recovery of platinum from spent Al_2O_3 carrier Pt/Re catalyst leach liquor.

matte smelting, acid leaching, and filtration. The spent Al_2O_3 carrier catalyst was ground to 60–100 μm sizes in a mill. The ground spent catalyst, pyrite, CaO, SiO_2, $Na_2B_4O_7$, and carbon powder were evenly mixed of different proportions in the graphite crucible. The graphite crucible was placed in an intermediate frequency furnace (IFF) to matte smelting the mixtures at a controlled temperature of 1400°C and melting time of 50 min. After completing the melting, the graphite crucible was removed, and the melting slag was separated from the FeS traps during and after melting. The melting slag and iron traps were sampled, and the Pt content was analyzed. The recovery efficiency of Pt can be expressed as follows:

$$1 - \frac{m_2 \times c_2}{m_1 \times c_1}, \quad (1)$$

where m_1 is spent Al_2O_3 carrier catalyst weight (g), m_2 is melting slag weight (g), c_1 is Pt content in spent Al_2O_3 carrier catalyst (g/t), and c_2 is Pt content in melting slag (g/t).

According to the corrosion resistance of PGMs, the enrichment and recovery of Pt from iron matte were obtained via sulfuric acid leaching. Platinum was enriched in leaching residue to attain high-grade PGMs concentrations. Mixing of the iron matte and leachate were carried out in a 1000 ml beaker, which was placed in a thermostatically regulated water bath. A magnetic stirrer was used to stir the mixture. After the leaching is completed, the leachate is filtered, separated, dried, and weighed. The enriching efficiency of Pt can be expressed as follows:

$$\frac{m_4}{m_3}, \quad (2)$$

where m_4 is iron matte weight (g) and m_3 is leached residue weight (g).

FeS_2 in pyrite cracked into FeS at high temperature and formed matte after smelting. The reaction occurred as follows:
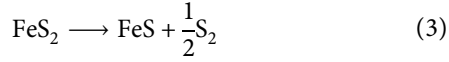
$$FeS_2 \longrightarrow FeS + \frac{1}{2}S_2 \qquad (3)$$

Table 1 shows thermodynamic data for calculating the relationship between the $\Delta G_T^\ominus$ and T, which can be obtained as follows:

$$\Delta G_T^\theta = 135457 - 121.377T \text{J}. \qquad (4)$$

It can be seen from the previously mentioned thermodynamic equation that when $\Delta G_T^\ominus$ is equal to 0, the reaction temperature will be 1116 K. At 1400°C temperature, $\Delta G_T^\ominus$ in (4) becomes −67.607 kJ, which is below zero. The negative value indicates that the reaction occurred thermodynamically. The thermodynamic calculation shows that (3) can be realized at 1400°C.

3. Results and Discussion

3.1. Trapping Experiment

3.1.1. Effect of Pyrite Dosage and Spent Catalyst on the Recovery Rate of Pt. Figure 4(a) shows the effect of pyrite dosage on platinum recovery. The graphite crucibles loaded with about

200 g of spent catalyst material mixed with a certain dosage of pyrite, 50 wt% CaO, 200 g quartz sand, 10 wt% C, 15 wt% CaF_2, and $Na_2B_4O_7$ at the melting temperature and time of 1400°C and 50 min, respectively. The recovery rate ratio of Pt increased with an increase in pyrite dosage from 0.1 : 1 to 0.5 : 1. As shown in Figure 3(a), the recovery rate of Pt can be enhanced from 89.54% to 97.92% by increasing the pyrite dosage from 0.1 : 1 to 0.6 : 1. When the pyrite dosage and spent catalyst was 0.1 : 1, the recovery rate of Pt was only 89.54%.

The matte was characterized using XRD. Figure 4(a) shows that the newly generated phases in leaching contained only FeS with sharp phase peaks. Low production of the FeS (trapping agent), metal content in the molten, and trapping efficiency of Pt during the smelting process were due to the limited quantity of pyrite. When a large quantity of pyrite was used, more FeS trapping agent was generated in the melting process. However, the content of Pt in the raw material was 1780 g/t. When the amount of trapping agent reached a certain limit, the effect of trapping remained uniform, and the total amount of alloy increased to some extent while the recovery rate unconspicuous. The recovery rate of Pt gradually increased as the quantity of pyrite increased. Therefore, 0.5 : 1 was determined as the ratio of the pyrite dosage to the spent catalyst of the recovery process.

There are few reports on the capture of platinum group metals by matte. Chen Jing assumed that the principle of capturing PGMs by matte depends on the matte metalloid property and the ability to form alloys. For the matte phase, because matte phase has high electrical conductivity at higher temperatures and its temperature coefficient is

TABLE 1: Thermodynamic data.

Material name	FeS_2	FeS_2	S_2
$\Delta S_{298}^\ominus$/J. K^{-1}	52.928	60.291	228.028
$\Delta H_{298}^\ominus$/J	−171544	−100416	−128658

negative, its conductance belongs to the electron conductance. The properties of matte are actually similar to those of the metal in the molten state; therefore, during the matte-smelting process, the precious metal atoms will enter the ice copper phase, rather than the slag phase.

3.1.2. Effect of CaO Dosage and Spent Catalyst Ratio on the Recovery Rate of Pt. To study effect of CaO dosage and spent catalyst on recovery rate of Pt, a series of matte smelting experiments were performed under the matte smelting temperature and time of 1400°C and 50 min, respectively, with other matte smelting conditions kept constant. The effect of CaO dosage on the recovery rate of Pt is shown in Figure 4(b). It shows that when the ratio of calcium oxide dosage to spent catalyst was increased from 0.25 : 1 to 0.5 : 1, the recovery rate of Pt increased from 95.21% to 97.85%. But the recovery rate of Pt decreased when the amount of calcium oxide dosage increased further. When the ratio of calcium oxide dosage to spent catalyst was 1 : 1, the recovery rates of Pt decreased to 92.33%. The reason is that the quantity of CaO affects the amount of slag basicity. The CaO can form a low melting point slag phase with SiO_2 and Al_2O_3 in the raw material and break the Si-O chemical bond, reduce the viscosity of the slag, and improve the recovery of Pt. It was observed that the slag basicity became low at a lower quantity of CaO, resulting in poor platinum trapping from the slag by FeS. With the increase in basicity, the viscosity significantly decreased, the slag fluidity improved considerably, and the platinum particles in the slag were fully trapped by the FeS, which led to the significant increase in the recovery of platinum. However, when excess CaO was used, the viscosity of the slag became so low that the trapping agent FeS_2 fell to the bottom of the graphite crucible and decomposed completely. The suspended platinum metal particles cannot be fully trapped by FeS, resulting in a poor trapping effect and a decrease in the recovery of Pt. Consequently, CaO and the spent catalyst ratio were determined to be 0.5 : 1.

3.1.3. Effect of Quartz Sand and Spent Catalyst Ratio on the Recovery Rate of Pt. The effect of quartz sand and spent catalyst ratios on the platinum recovery was investigated under the same conditions as previously mentioned. The results are shown in Figure 4(c). The recovery rate of Pt increased linearly with the ratio of quartz sand to spent catalyst between 1.2 : 1 and 2 : 1. When the ratio of quartz sand to spent catalyst exceeded 2 : 1, the recovery rate of Pt gradually increased as the amount of quartz sand increased. Thus, the excessive slagging agent will undoubtedly reduce the processing efficiency of smelting equipment and increase the processing cost in production.

The melting slag was characterized using XRD. The results shown in Figure 5(b) demonstrate that the major phases in residue contain $CaO \cdot Al_2O_3 \cdot 2SiO_2$. The melting slag and iron matte phases appeared in the graphite crucible during the process. The melting slag was composed of gangue minerals SiO_2, Al_2O_3, and CaO, forming CaO-SiO_2-Al_2O_3 slag. The quantity of catalyst was fixed so that the viscosity and surface tension of the melting slag could be influenced directly by the CaO/SiO_2. During the slagging reaction, SiO_2 reacted with O^{2-} to form a tetrahedral structure ($SO4_4^{4-}$): silica anion silicon oxide complex anion. The slag acidic increased at high content of SiO_2, which enhanced the decomposition of Al_2O_3 and production of O^{2-}. The increase in the number of free O^{2-} will inhibit the production of ($SO4_4^{4-}$). At this point, high melting point intermediate products will be formed, which will reduce the fluidity of slag and the recovery rate. Therefore, the ratio of quartz sand to spent catalyst was determined to be 2 : 1.

3.1.4. Effect of $Na_2B_4O_7$ Dosage on Recovery Rates of Pt.

The effect of $Na_2B_4O_7$ dosage on recovery rates of Pt was investigated at the same previously mentioned existing conditions. The relationship between the recovery of platinum and the amount of $Na_2B_4O_7$ is shown in Figure 4(d).

The recovery of platinum increased from 96.27% to 97.74% when the $Na_2B_4O_7$ dosage increased from 10% to 15%. The recovery rates of Pt were found to increase dramatically when $Na_2B_4O_7$ dosage was set within 15%. However, increasing trend becomes mild when the $Na_2B_4O_7$ dosage exceeded 15% by weight of spent catalyst; hence, the recovery rates of Pt increased gradually. When the weight ratio of borax and the spent catalyst was 0.2 : 1, the recovery rates of Pt became 98.02%. The $Na_2B_4O_7$ is is an important flux, which can form borate with CaO, SiO_2, and Al_2O_3 with low melting point (1154°C) and viscosity such as $CaO \cdot B_2O_3$. Also, $Na_2B_4O_7$ can significantly reduce the melting point of the slag. Conversely, there was no significant effect on the recovery of platinum as the amount of $Na_2B_4O_7$ was increased. Therefore, the optimum condition of $Na_2B_4O_7$ was 15%.
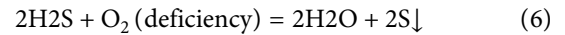
3.2. Leaching

3.2.1. Effect of H_2SO_4 Concentration on Pt Enriching Efficiency.

The effect of H_2SO_4 concentration on Pt enrichment efficiency and the dissolution of iron matte was investigated under the following conditions: L/S ratio of 10 : 1, leaching temperature of 60°C, and leaching time of 70 min (Figure 6(a)). The H_2SO_4 concentration varied from 1.2 to 2.0 mol/L. The enriching efficiency of Pt increased linearly with an increase in H_2SO_4 concentration. Higher concentration of H_2SO_4 improved the leaching efficiency of FeS and the enriching efficiency of platinum. Conversely, FeS dissolves less in lower concentrations of H_2SO_4. The enrichment of platinum in the residue increases from 7.43 to 22.41 times when the H_2SO_4 concentration increases from 1.2 mol/L and 1.8 mol/L, respectively. It can be found that the enrichment of platinum increases negligibly when the H_2SO_4 concentration exceeds 1.8 mol/L.

During the leaching process, sulfuric acid reacted with FeS to form $FeSO_4$ and H_2S. The chemical reaction of the equation is as follows:

$$FeS + H2SO_4 = FeSO4 + H2S\uparrow \tag{5}$$
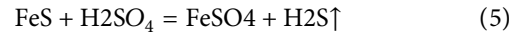
Hydrogen sulfide gas dissolves in water to form hydrosulfuric acid. In the absence of oxygen, the chemical reaction equation becomes

$$2H2S + O_2 \,(deficiency) = 2H2O + 2S\downarrow \tag{6}$$

Therefore, there is the formation of sulfur in the reaction process.

XRD analyses of acid leaching residue with different concentrations are shown in Figure 6(b). The physical phase of leaching residue changed with the increasing H2SO4 concentration. There were no noticeable changes at the concentrations of 1.2 mol/L and 1.4 mol/L. The FeS was the main phase of leaching residue, and a small S phase was observed. When the H_2SO_4 concentration increased to 1.6 mol/L, noticeable characteristic peaks of S were observed, and the characteristic peaks of FeS dropped a little. Moreover, when the H_2SO_4 concentration increased to 1.8 mol/L, the main phase was sulfur, which led to the disappearance of characteristic peaks of FeS. Therefore, the optimum H_2SO_4 concentration was 1.8 mol/L.

3.2.2. The Effect of Leaching Temperature on Pt Enriching Efficiency.

The leaching process can be influenced by temperature. Thus, the optimization of the leaching process temperature was investigated at a varied temperature from 30°C to 70°C with other conditions such as H_2SO_4 concentration 1.8 mol/L, liquid/solid (L/S) ratio of 10 : 1, and leaching time of 70 min. The effect of leaching temperature on the platinum enrichment efficiency is shown in Figure 7(a). The leaching temperature ranged from 30°C to 60°C, and the enriching efficiency of Pt increased from 6.27 to 18.86. When the leaching temperature was increased to 70°C, the enriching efficiency of Pt dropped to 17.39. The decrease in enriching efficiency can be attributed to the large number of sulfur elements formed in the process.

The leaching residue with different leaching temperatures was characterized using XRD (Figure 7(b)). As shown in Figure 7(b), no significant changes in the phases were observed when the leaching temperatures were 40°C and 60°C. Moreover, the FeS was observed as the main phase of leaching residue, and a small amount of the S phase formed. When the temperature is increased to 70°C, the diffraction peaks of FeS disappear, while the diffraction peaks of S appear. The S phase diffraction peak of the leaching residue obtained at 80°C was stronger than other leaching temperatures. Thus, 60°C was selected as the optimal temperature for acid leaching. Under this condition, the enriching efficiency increased to 18.86. Therefore, a temperature of 60°C is suitable for the leaching process and is the optimum leaching temperature.

3.2.3. The Effect of Leaching Time on Pt Enriching Efficiency.

The influence of leaching time on Pt enriching efficiency was investigated under the following conditions: leaching
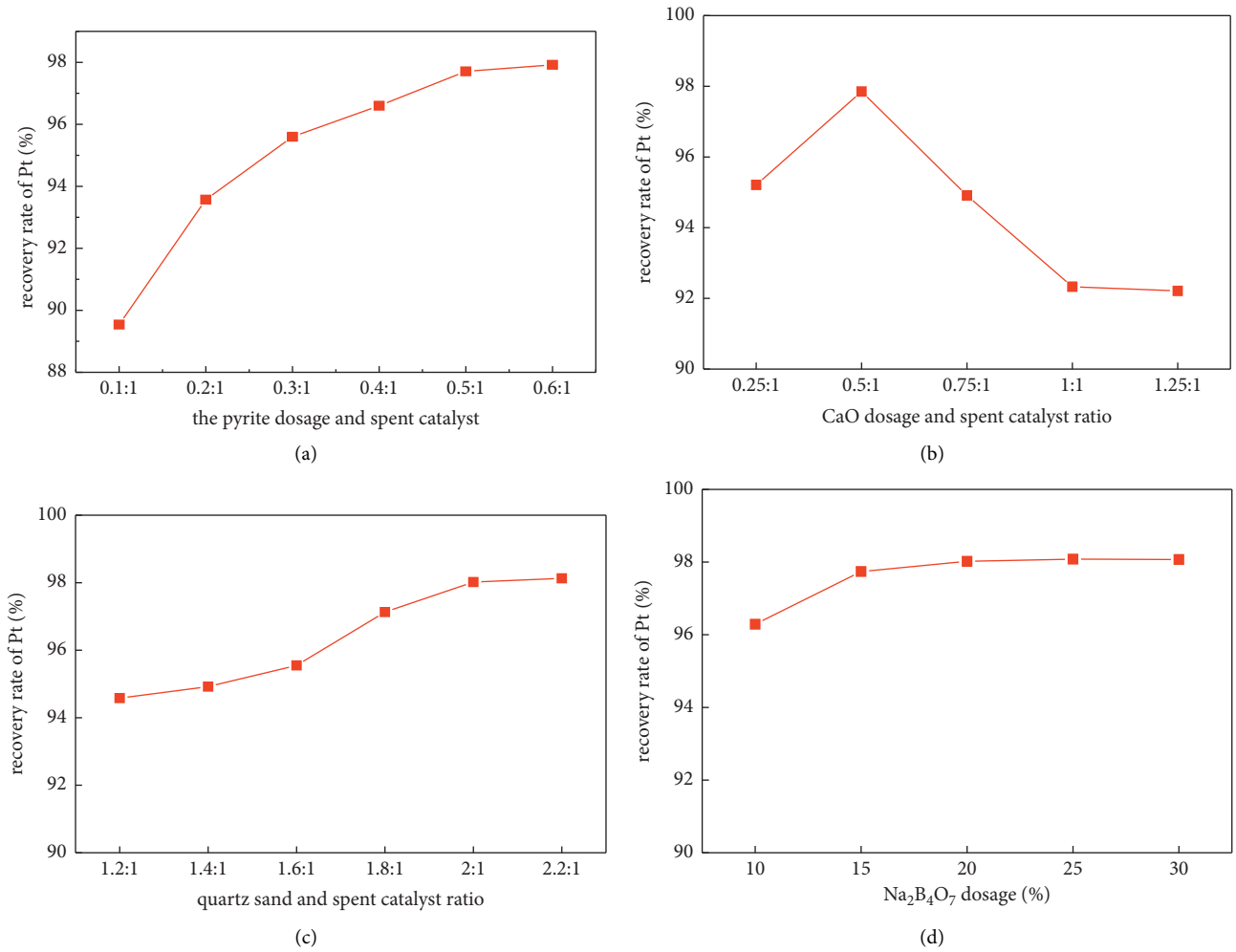
Figure 4: Pt recovery efficiency under different melting parameters. (a) Effect of trapping agent on recovery rates of Pt. (b) Effect of CaO dosage on recovery rates of Pt. (c) Effect of quartz sand on recovery rates of Pt. (d) Effect Na_2B4O_7 dosage on recovery rates of Pt.
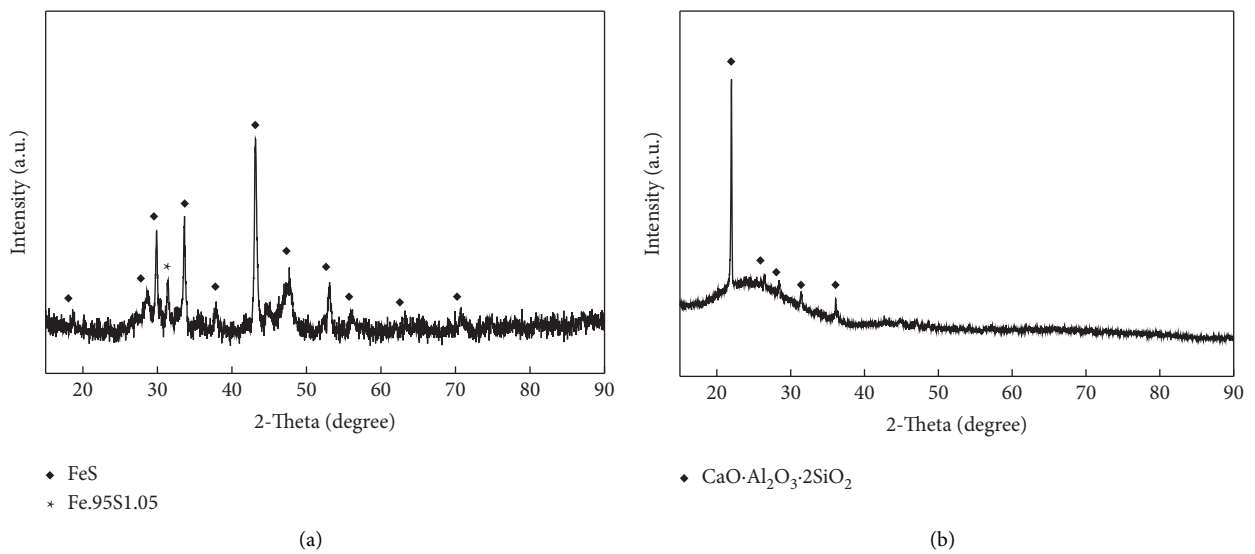

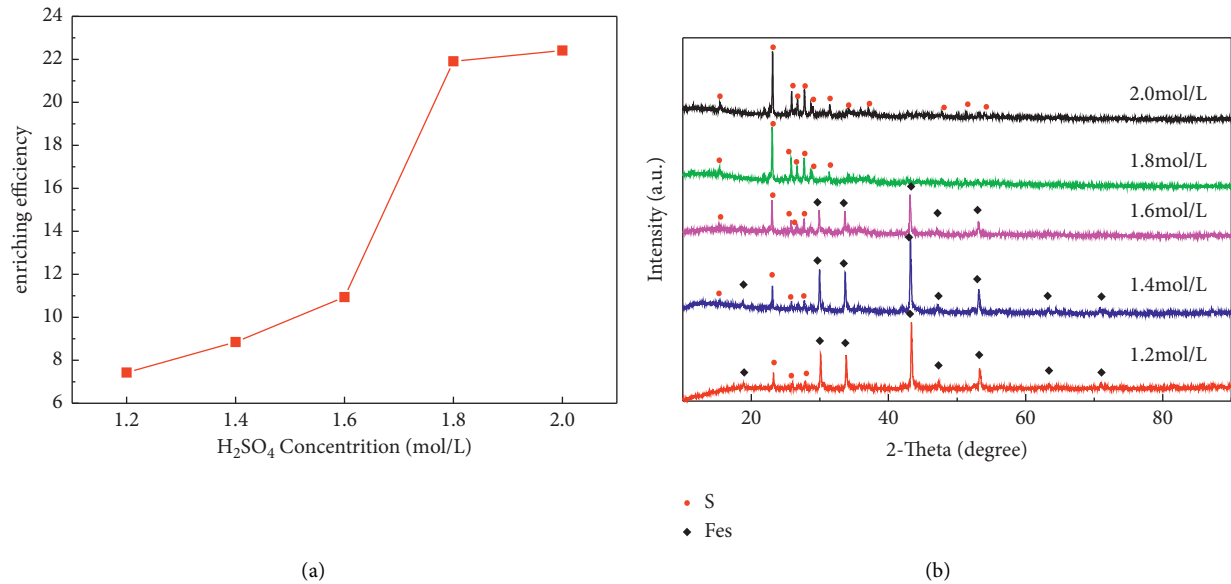Figure 5: XRD pattern of iron matte and smelting slag.

(a)

(b)

FIGURE 6: Effects on (a) Pt enriching efficiency and (b) XRD analysis of acid leaching residue with different concentrations.
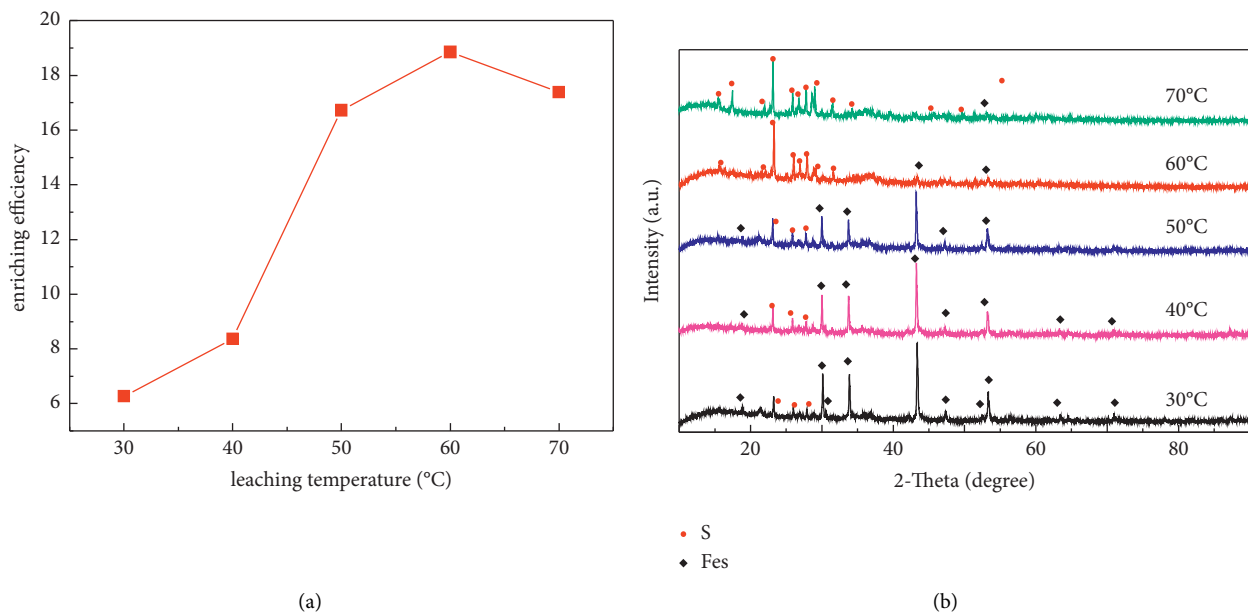

(a)

(b)

FIGURE 7: Effects on (a) Pt enriching efficiency and (b) XRD analysis of leaching residue with different temperatures.

temperature of 60°C, H_2SO_4 concentration of 1.8 mol/L, and liquid/solid (L/S) ratio of 10 : 1, at leaching times 30, 40, 50, 60, and 70 min. The effect of leaching time on Pt enriching efficiency is illustrated in Figure 8(a). The leaching acid attained high enriching efficiency of Pt when the leaching was 70 min (Figure 8(a)). The enriching efficiency gradually increased from 6.27 to 19.11 as the leaching time increased from 30 min to 70 min.

The leaching residue with different leaching times was characterized using XRD (Figure 7(b)). As shown in Figure 8(b), no significant changes in the phases were observed when leaching time was 40 min; thus, this was observed to be the main phase of leaching residue. However, a significant change was observed when leaching time increased from 40 min to 70 min. Moreover, the diffraction strength of FeS decreased, and S was observed to be the main phase of leaching residue. The diffraction peaks of FeS disappeared, and the remaining phases were S when the leaching time was adopted as 70 min. This indicated that the iron matte had been leached out. The results show that the leaching time can best influence the iron matte leaching within 60 min. Consequently, the leaching time was determined to be 60 min.

3.2.4. Effect of L/S Ratio on Pt Enriching Efficiency. The L/S ratio usually plays an important role in leaching. The effect of the L/S ratio on the enriching efficiency of Pt was also investigated. The L/S ratio varied from 4 : 1 to 12 : 1, and the
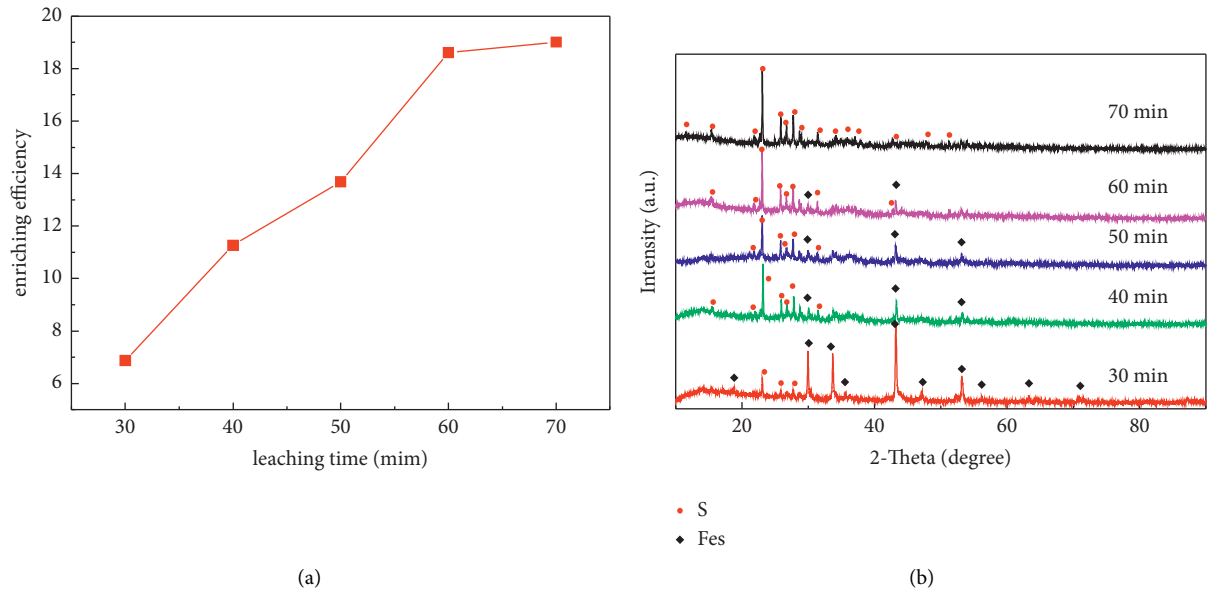
(a)

(b)

FIGURE 8: Effects on (a) Pt enriching efficiency and (b) XRD analysis of leaching residue with different time periods.
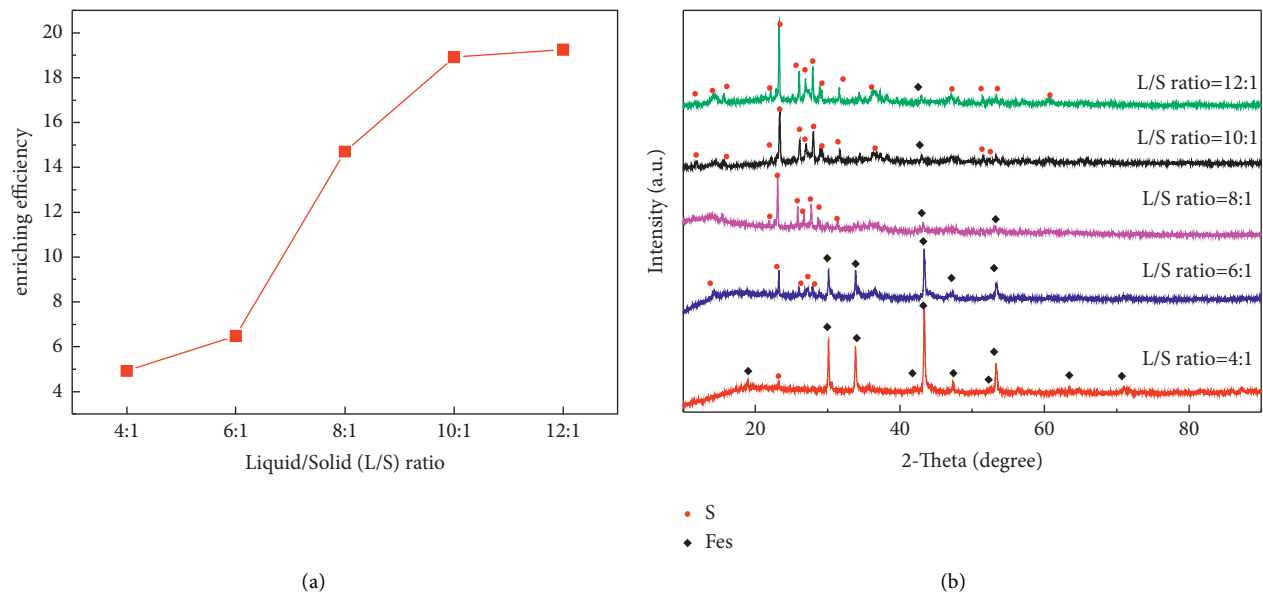

(a)

(b)

FIGURE 9: Effects on (a) Pt enriching efficiency and (b) XRD analysis of leaching residue with different L/S ratios.

other conditions were H_2SO_4 concentration 1.8 mol/L, leaching temperature of 60°C, and leaching time of 70 min. Other experimental conditions remain constant. Figure 9(a) shows the effect of the L/S ratio on the enriching rate of Pt by using H_2SO_4. As shown in Figure 9(a), the enriching rate of Pt significantly increased as the L/S ratio increased from 4 : 1 to 12 : 1. It was observed that only a 4.92 enriching rate of Pt at the L/S 4 : 1 was obtained. As the L/S ratio increases to 10 : 1, the enriching rate becomes 18.91. The enriching rate gradually increased when the L/S ratio exceeded 10 : 1. The enriching rate became only 19.24 when the L/S ratio was 12 : 1. Figure 9(b) illustrates the results obtained from the XRD of leaching residue with different L/S ratios. There was no

significant change observed when the L/S ratio was 4 : 1; this indicated the presence of FeS in the main phase of leaching residue. When the L/S ratio was 6 : 1, the intensity of the diffraction peaks of FeS was reduced, and a small diffraction peak of S appeared. When the L/S ratio was 8 : 1, a significant change was observed, and the diffraction peaks of FeS disappeared, indicating the presence of S only in the phase. There was no obvious difference when the L/S ratio was within 10 : 1 and 12 : 1; the main phase of leaching residue was S. At a certain concentration of the leaching agent, the concentration of metal ions in the leaching solution was reduced at a high L/S ratio. Therefore, increasing the mass transfer rate between the solid and liquid phase is beneficial,

thereby increasing the leaching efficiency. Furthermore, if the L/S ratio is too high, the loss of leaching agent and a load of liquid-solid separation equipment cost will increase, which may not be economically beneficial. Consequently, the optimum L/S ratio was determined to be 10 : 1.

4. Conclusion

The enrichment effect of platinum on the spent Al_2O_3 carrier catalyst using matte smelting of the pyrite-acid leaching process was significant. Pyrite was cracked to form FeS (trapping agent), which can effectively trap platinum. Through the experimental study, the matte smelting-acid leaching method was determined for the enrichment and recovery of platinum. The recovery rate of platinum was 98% under the optimum conditions, including a temperature of 1400°C, reacting time of 50 min, mass ratio of pyrite to spent catalysts of 1 : 2, a ratio of CaO to spent catalysts of 0.5 : 1, the ratio of quartz sand to the raw material of 2 : 1, and 15% (by weight of the spent catalyst) dosage of borax and calcium fluoride. The acid leaching was more efficient under the following optimum conditions: H_2SO_4 concentration of 1.8 mol/L, leaching time of 70 min, leaching temperature of 60°C, and L/S ratio of 10 : 1. Furthermore, the enrichment of Pt under the optimum condition (1.8 mol/L concentration of H_2SO_4) was 19 times higher than in other conditions (such as 1.2 mol/L concentration of H_2SO_4). Finally, at the optimum conditions, S was the only main phase in leaching residue, and the enrichment effect was observed to be significant.

Data Availability

The datasets used and/or analyzed during the current study are available from the corresponding author on reasonable request.

Conflicts of Interest

The authors declare that they have no conflicts of interest.

Acknowledgments

References

[1] H. B. Trinh, J. C. Lee, Y. J. Suh, and J. Lee, "A review on the recycling processes of spent auto-catalysts: towards the development of sustainable metallurgy," *Waste Management*, vol. 114, pp. 148–165, 2020.

[2] C. Saguru, S. Ndlovu, and D. Moropeng, "A review of recent studies into hydrometallurgical methods for recovering PGMs from used catalytic converters," *Hydrometallurgy*, vol. 182, pp. 44–56, 2018.

[3] H. Dong, J. Zhao, J. Chen, Y. Wu, and B. Li, "Recovery of platinum group metals from spent catalysts: a review," *International Journal of Mineral Processing*, vol. 145, pp. 108–113, 2015.

[4] L. A. Diaz, T. E. Lister, J. A. Parkman, and G. G. Clark, "Comprehensive process for the recovery of value and critical materials from electronic waste," *Journal of Cleaner Production*, vol. 125, pp. 236–244, 2016.

[5] M. K. Jha, J. C. Lee, M. S. Kim, J. Jeong, B. S. Kim, and V. Kumar, "Hydrometallurgical recovery/recycling of platinum by the leaching of spent catalysts: a review," *Hydrometallurgy*, vol. 133, pp. 23–32, 2013.

[6] Y. Chen, Q. Qiao, J. Cao, H. Li, and Z. Bian, "Precious metal recovery," *Joule*, vol. 5, no. 12, pp. 3097–3115, 2021.

[7] D. Jimenez de Aberasturi, R. Pinedo, I. R. Larramendi, J. I. R. D. Larramendi, and T. Rojo, "Recovery by hydrometallurgical extraction of the platinum group metals from car catalytic converters," *Minerals Engineering*, vol. 24, no. 6, pp. 505–513, 2011.

[8] Y. J. Ding and S. G. Zhang, "Status and research progress on recovery of platinum group metals from spent catalysts," *Chinese Journal of Engineering*, vol. 42, no. 3, pp. 257–269, 2020.

[9] J. C. Zhao, H. Chui, S. M. Bao, W. F. Tong, Y. D. Wu, and H. G. Dong, "Theory and practice on recovery of platinum group metals from spent auto catalysts by reduction-smelting copper trapping method," *Chinese Journal of Nonferrous Metals*, vol. 29, no. 12, pp. 2819–2825, 2019.

[10] H. X. Xiao, "Determination of gold and palladium in printed circuit board by plumbum assaying-flame atomic absorption spectrometry method," *Materials Research and Application*, vol. 6, no. 3, pp. 215–218, 2012.

[11] M. Benson, C. R. Bennett, M. K. Patel, J. E. Harry, and M. Cross, "Collector-metal behaviour in the recovery of platinum-group metals from catalytic converters," *Mineral Processing and Extractive Metallurgy*, vol. 109, no. 1, pp. 6–10, 2000.

[12] J. C. Zhao, H. Chui, S. M. Bao, W. F. Tong, and Y. D. Wu, "Recovery of platinum group metal secondary resource by iron trapping method based on solid state," *Chinese Journal of Nonferrous Metals*, vol. 24, no. 10, pp. 2692–2697, 2014.

[13] Y. Liu, Q. Song, L. Zhang, and Z. Xu, "Novel approach of in-situ nickel capture technology to recycle silver and palladium from waste nickel-rich multilayer ceramic capacitors," *Journal of Cleaner Production*, vol. 290, Article ID 125650, 2021.

[14] H. Tang, Z. Peng, Z. Li et al., "Recovery of platinum-group metals from spent catalysts by microwave smelting," *Journal of Cleaner Production*, vol. 318, Article ID 128266, 2021.

[15] L. Zhang, Q. Song, Y. Liu, and Z. Xu, "Novel approach for recovery of palladium in spent catalyst from automobile by a capture technology of eutectic copper," *Journal of Cleaner Production*, vol. 239, Article ID 118093, 2019.

[16] J. C. Zhao, H. Chui, S. M. Bao, W. F. Tong, and H. G. Dong, "Recovery of Pt, Pd and Rh from spent auto catalysts by copper collection method," *Precious Metals*, vol. 39, no. 1, pp. 56–59, 2018.

[17] H. Zheng, Y. Ding, Q. Wen et al., "Slag design and iron capture mechanism for recovering low-grade Pt, Pd, and Rh from leaching residue of spent auto-exhaust catalysts," *The Science of the Total Environment*, vol. 802, Article ID 149830, 2022.

[18] M. H. Morcali, "A new approach to recover platinum-group metals from spent catalytic converters via iron matte," *Resources, Conservation and Recycling*, vol. 159, Article ID 104891, 2020.