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Pt and Al Recovery from a Spent Pt/Al_2O_3 Catalyst via an Integrated Soda Roasting–Alkaline Leaching–Carbonization Process

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Abstract: An integrated soda roasting–alkaline leaching–carbonization process was developed to recover platinum (Pt) and aluminum (Al) from a spent Pt/Al_2O_3 catalyst. A product with the main component of $NaAlO_2$ was obtained under the optimal roasting conditions of the mass ratio of Na_2CO_3 to spent catalyst 1, roasting time 2 h and roasting temperature 900 °C. The Al in the roasted residue was effectively leached in a dilute NaOH solution, while Pt was enriched in the leached residue with a Pt content that reached 29.4%, and the calculated concentration factor of Pt yielded 79.4. After carbonization was performed for the leached solution, 99.0% of the Al was recovered as the pseudo-boehmite product, which could be used as the feed for preparing the Al_2O_3 carrier. Pt and Al_2O_3 in the spent catalyst were selectively separated and enriched using the above process, which thus has good application prospects.

Keywords: spent Pt/Al_2O_3 catalyst; soda roasting; alkaline leaching; carbonization; pseudo-boehmite

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1. Introduction

Platinum (Pt) is an important precious metal, and it has strong catalytic activity [1–3]. Al_2O_3 is usually used as the carrier where Pt is loaded to enhance its catalytic performance [4,5]. Pt/Al_2O_3 catalysts are widely used in the chemical industry. After being used for a certain amount of time, the catalytic performance of the catalysts will evidently be decreased and spent Pt/Al_2O_3 catalysts will be produced [6–9]. The spent Pt/Al_2O_3 catalysts have very high economic value. Therefore, recovering platinum (Pt) and aluminum (Al) from spent Pt/Al_2O_3 catalysts is imperative [10–13].

During their use, some organic substances and carbon will be deposited on the surface of the catalyst. In order to eliminate their negative effects on Pt and Al recovery, the spent Pt/Al_2O_3 catalysts are generally subjected to roasting to remove these substances [14]. Hydrometallurgical processes are the main routine to recover Pt and Al in industrial practice [15,16]. In some studies, the spent Pt/Al_2O_3 catalyst is dissolved in H_2SO_4 or NaOH to separate Al_2O_3, and Pt is concentrated in the leached residue, which was recovered by leaching with aqua regia [17,18]. Some studies [19,20] have suggested that Pt in the spent catalyst can be leached in a HCl medium in the presence of oxidizing agents. The Pt in the leaching solution was then recovered via direct reduction using active metals or solvent extraction followed by refining treatment. The Pt recovery was over 98%, and a platinum product with 99.95% purity was obtained. In a recent study [21], aqua regia leaching with the help of microwaves was adopted to recover Pt from a spent Pt/Al_2O_3 catalyst. Pt was recovered in the form of $(NH_4)_2PtCl_6$ with a recovery of 98.3%. A complete dissolution method was used to dissolve Pt and Al_2O_3 from a spent Pt/Al_2O_3 catalyst, followed by ion

exchange and refining [22]. Pure platinum and aluminum polychloride were produced, and the recoveries of Pt and Al were over 99% and 98%, respectively. However, the above-mentioned processes still face some issues, such as high reagent consumption, unstable recovery percentage and potential harm to the environment [23]. Thus, it is essential to develop an eco-friendly process to recover Pt and Al. In order to realize the high-efficiency separation and enrichment of Pt and Al_2O_3 in the spent catalyst, a novel routine is to selectively transform the Al_2O_3 carrier into a form that can be easily leached/dissolved while Pt still exists in its initial form.

In this study, a soda roasting–alkaline leaching–carbonization process was investigated to recover Pt and Al from a spent Pt/Al_2O_3 catalyst. Using Na_2CO_3 roasting, the Al_2O_3 in the catalyst was first converted into soluble $NaAlO_2$, while Pt was not transformed and remained in the roasted residue. After the roasted residue was subjected to NaOH leaching, Al was dissolved in the leached solution, and it was recovered via the carbonization process for preparing the pseudo-boehmite product. Pt was not lost during the roasting and leaching, and it was concentrated in the leached residue and can be further recovered.

2. Experimental Work

2.1. Materials and Reagents

The spent Pt/Al_2O_3 catalyst used in this study was from a Chinese petrochemical company, and its chemical composition is shown in Table 1. The main components of the spent catalyst were Pt, Al_2O_3, Fe_2O_3 and C, of which their contents reached up to 0.37%, 88.00%, 2.00% and 6.5%, respectively. The X-ray diffraction (XRD) pattern of the spent catalyst is displayed in Figure 1. It can be seen that Al_2O_3 was the only phase in the spent catalyst. No Pt phase was observed because the Pt content in the catalyst was below the detection limit. The reagents used in this work, including Na_2CO_3 and NaOH, were of analytical grade. Ultrapure water was used in all the experiments.

Table 1. Chemical compositions of the spent Pt/Al_2O_3 catalyst.

Element	Pt	Al_2O_3	Fe_2O_3	K_2O	S	C
Content (%)	0.37	88.00	2.00	1.80	0.30	6.50

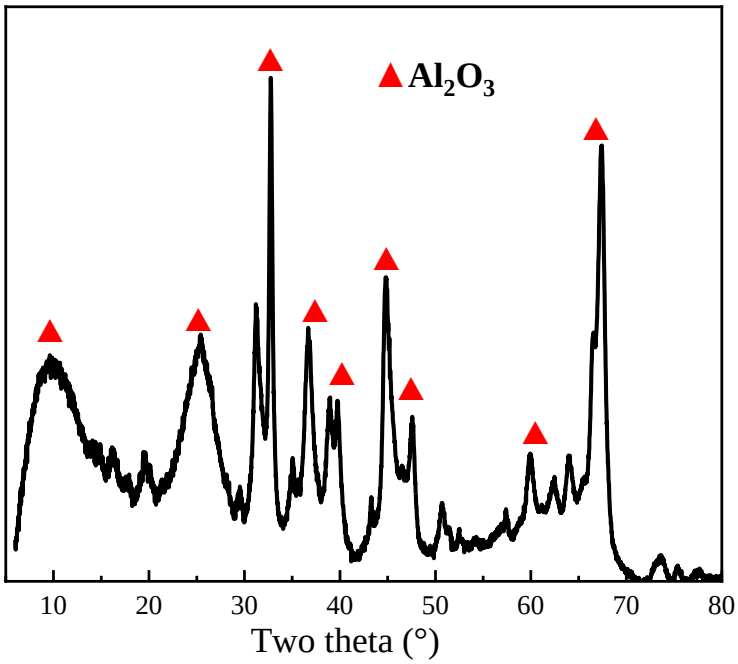


Figure 1. XRD pattern of the spent Pt/Al_2O_3 catalyst.

2.2. Experimental Methods

The flowchart of the proposed integrated process is shown in Figure 2. The experiments include three aspects, i.e., soda roasting, alkaline leaching and carbonization. Soda roasting experiment was conducted in a muffle furnace. Before roasting, a certain amount of spent Pt/Al_2O_3 catalyst was fully mixed with Na_2CO_3, and the furnace was heated to a predetermined temperature. Afterward, the mixture was put in a nickel crucible, which was roasted in the furnace. When the roasting was finished, the power supply was cut off, and the roasted residue was cooled and collected for further analyses and leaching tests.

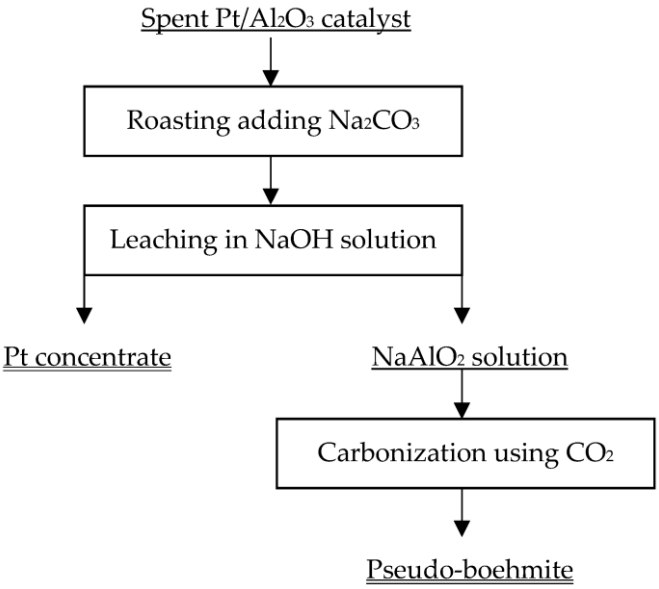


Figure 2. Experimental flow of recovering Pt and Al from a spent Pt/Al_2O_3 catalyst.

All the leaching tests were performed in a 250 mL beaker, which was equipped with an electric agitator, and the reaction temperature was controlled via an electro-thermostatic water bath. For each test, ultrapure water and NaOH were sequentially added into the beaker and heated to a predetermined temperature. After that, the roasted residue was added, and then it was immediately agitated at a constant speed of 400 rpm. When the reaction was completed, the pulp was filtrated using a vacuum filter. The obtained solution was used for Al recovery, and the leached residue was washed and dried for subsequent detection. The concentration factor of Pt was calculated according to Equation (1):

$$CF = \frac{C_1}{C_0} \quad (1)$$

where CF is the concentration factor, C_1 (wt. %) is the Pt content in the leached residue and C_0 (wt. %) is the Pt content in the spent Pt/Al_2O_3 catalyst.

The leached solution containing $NaAlO_2$ was used to produce pseudo-boehmite via the carbonization method, and the detailed experimental procedure was described in our previous work [24]. First, CO_2 gas was injected into the leached solution, which was stirred at the speed of 200 rpm until the pH value was up to 10.5. Then, the solution was filtered, and a filter cake was obtained. Afterward, the cake was heated to 90 °C and aged for 4 h in an oven, and the pseudo-boehmite product was obtained.

2.3. Analytical Methods

The element content in the spent catalyst was determined using an inductively coupled plasma optical emission spectrometer (ICP-OES, Optima 5300DV, PerkinElmer, WA, USA). The mineral compositions in the spent catalyst and pseudo-boehmite product were analyzed via X-ray diffraction (XRD, D8 Advance, Bruker, Karlsruhe, Germany). The

surface morphology of the pseudo-boehmite product was characterized with a scanning electron microscope (SEM, MIRA3 LMS, TESCAN Brno, s,r.o., Brno, Czech Republic). To provide theoretical guidance for sodium roasting of the spent Pt/Al_2O_3 catalyst, the equilibrium component distribution of each phase at different roasting temperatures and Na_2CO_3 dosages was studied using the thermodynamic software Factsage 7.0 (Thermfact/CRCT, Montreal, QC, Canada; GTT-Technologies, Ahern, Germany) [25–28]. Factsage supports the computation and manipulation of phase diagrams, complicated phase equilibria and predominance diagrams. The species and phases that were either present in the original material or potentially formed during the process were included in the input data. The initial pressure was set to 0.1 MPa, and the phase formation was simulated in the temperature range of 100–1100 °C and $Na_2CO_3/(Na_2CO_3 + Al_2O_3)$ molar ratio range of 0–1.

3. Results and Discussion

3.1. Soda Roasting of Spent Pt/Al_2O_3 Catalyst

3.1.1. Effect of Roasting Temperature

In this study, the effects of important roasting conditions, including roasting temperature, mass ratio of Na_2CO_3 to spent catalyst and roasting time, on the concentration factor of Pt were systematically investigated. To investigate the suitable roasting conditions, a thermodynamic analysis of sodium roasting was performed. The phase diagram of the Na_2CO_3-Al_2O_3 system under the air atmosphere is displayed in Figure 3. When the temperature is lower than 360 °C, Na_2CO_3 does not react with Al_2O_3, even at a high Na_2CO_3 dosage. As the temperature is between 360 °C and 750 °C, Na_2CO_3 and Al_2O_3 begin to react, but the reaction is incomplete, and only the intermediate product $NaAl_9O_{14}$ is formed. With the increase in temperature to 750–1100 °C, the $NaAlO_2$ phase starts to appear at the $Na_2CO_3/(Na_2CO_3 + Al_2O_3)$ molar ratio of 0.15–0.5, but the reaction is still incomplete. Only when the molar ratio is greater than 0.5 can Al_2O_3 be entirely transformed into $NaAlO_2$.

The effect of roasting temperature is displayed in Figure 4. Clearly, the concentration factor of Pt was only 14.1 when the spent catalyst was roasted at 700 °C. With the increase in roasting temperature from 700 °C to 900 °C, the concentration factor also rose from 14.1 to 79.4. The chemical reaction between Al_2O_3 and Na_2CO_3 belongs to a solid–solid reaction at low temperatures because the melting point of Na_2CO_3 is 851 °C. As the roasting temperature was over 900 °C, the roasting reaction became a liquid–solid reaction. As a result of this, the reaction between Al_2O_3 and Na_2CO_3 was easier, contributing to the increase in the concentration factor. The phase diagram of the Na_2CO_3-Al_2O_3 system in the air condition presented in Figure 3 can support the above result. Clearly, with the increase in roasting temperature, the contents of Al_2O_3, Na_2CO_3 and intermediate products ($Na_2Al_{12}O_{19}$ and $NaAl_9O_{14}$) decreased, while that of $NaAlO_2$ increased, indicating that the reaction between Al_2O_3 and Na_2CO_3 was more complete.

The phase change rule of roasted residue with the increase in roasting temperature was also researched, and the results are indicated in Figure 5a–d. Clearly, when the roasting temperature was below 900 °C, the main phases in the roasted residue were Al_2O_3 and Na_2CO_3, as well as a small amount of $NaAlO_2$ (Figure 5a,b). As the roasting temperature rose to 900~1000 °C, the only phase was $NaAlO_2$ (Figure 5c,d). The phase change rule was consistent with the result of the thermodynamic analysis of sodium roasting in Figure 3. Under the standard state, the initial reaction temperature of Equation (2) is about 700 °C. When the roasting temperature was between 700 °C and 800 °C, most of the Al_2O_3 did not react with Na_2CO_3, resulting in a low concentration factor of Pt. A higher roasting temperature made the chemical reaction more complete. It can be seen that when the roasting temperature was 900 °C, the concentration factor was the biggest, which indicated that this roasting temperature is optimal for the formation of soluble $NaAlO_2$.

$$Al_2O_3 + Na_2CO_3 = 2NaAlO_2 + CO_2 \quad (2)$$

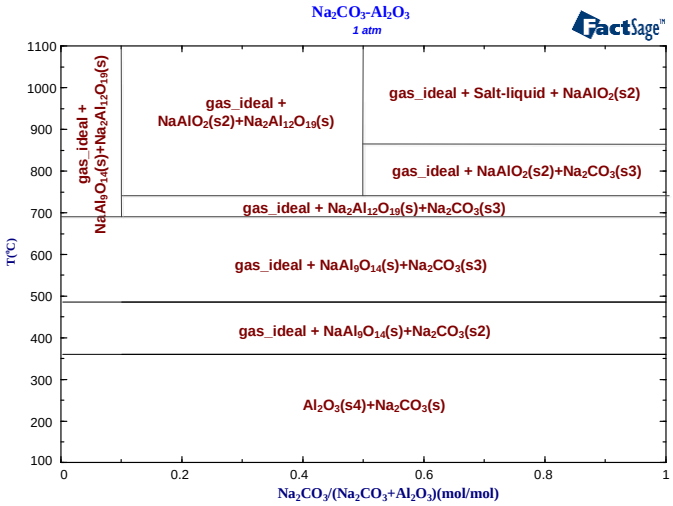


Figure 3. Phase diagram of the Na_2CO_3-Al_2O_3 system in the air condition.

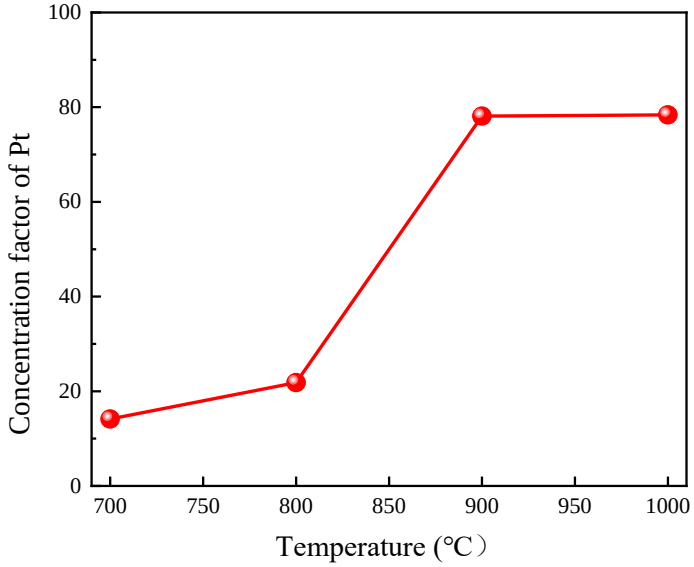


Figure 4. Effect of roasting temperature on the concentration factor of Pt. Roasting conditions: mass ratio of Na_2CO_3 to catalyst of 1, time of 2 h. Leaching conditions: temperature of 80 °C, time of 10 min, liquid-to-solid ratio of 5, stirring speed of 240 r/min, NaOH concentration of 6%.

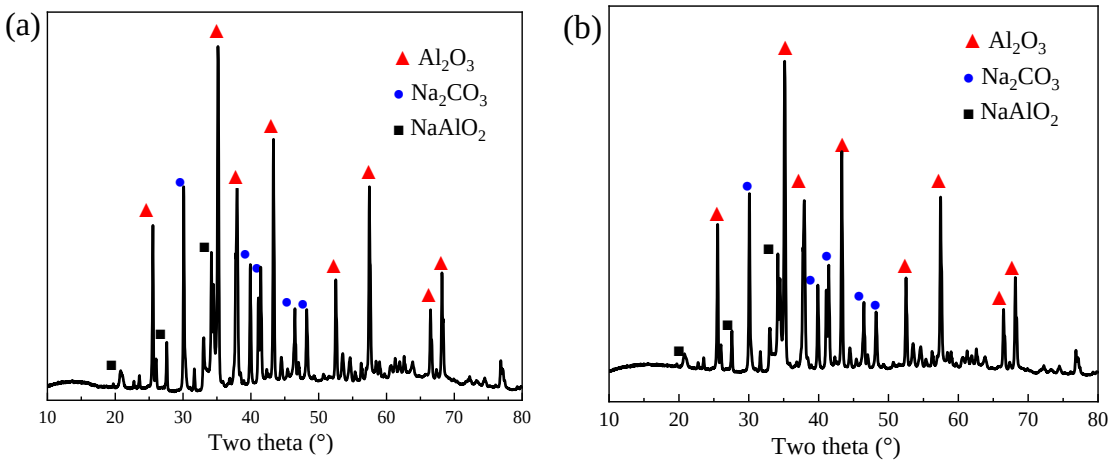


Figure 5. *Cont.*

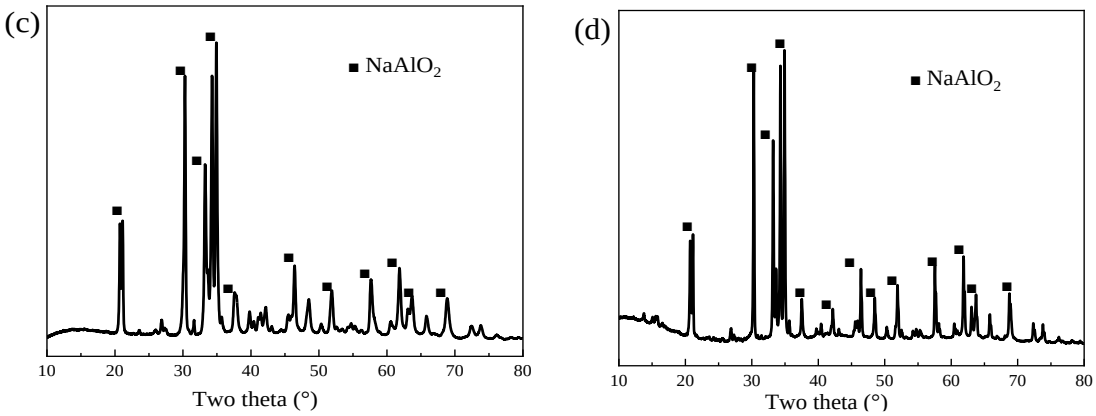


Figure 5. XRD patterns of the roasted residues under the roasting temperatures of 700 °C (**a**), 800 °C (**b**), 900 °C (**c**) and 1000 °C (**d**). Roasting conditions: mass ratio of Na_2CO_3 to catalyst of 1, time of 2 h. Leaching conditions: temperature of 80 °C, time of 10 min, liquid-to-solid ratio of 5, stirring speed of 240 r/min, NaOH concentration of 6%.

3.1.2. Effect of Roasting Time

The effect of roasting time on the concentration factor of Pt is indicated in Figure 6. The concentration factor of Pt was only 38.8 when the roasting time was 0.5 h. As the roasting time increased to 2 h, the concentration factor also rose to 79.4. Longer roasting times did not contribute to the evident increase in the Pt concentration factor, indicating that the reaction was complete at 2 h. In this process, it should be noted that Pt may be dissolved in the leaching stage, which will affect its subsequent recovery. Therefore, the Pt concentration in the leached solution was also determined. The result showed that Pt was not detected in the leached solution, which indicated that Pt was not dispersed in the leaching process. The leached solution containing $NaAlO_2$ can be used as the feed for preparing pseudo-boehmite.

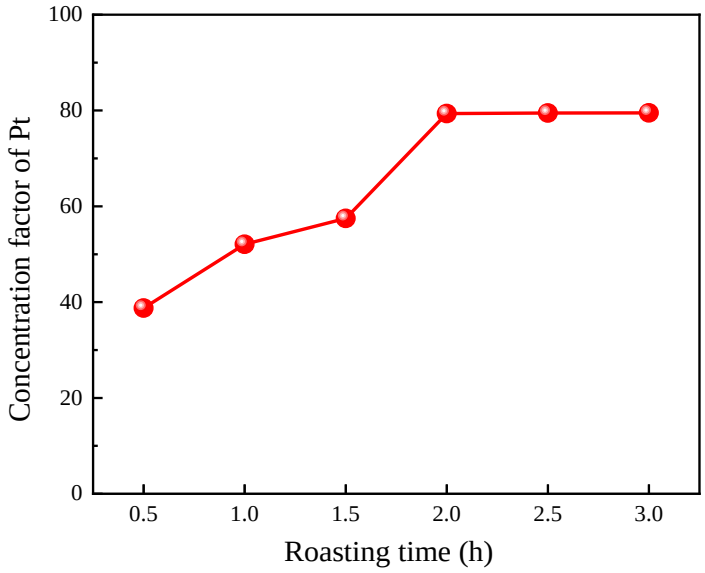


Figure 6. Effect of roasting time on the concentration factor of Pt. Roasting conditions: mass ratio of Na_2CO_3 to catalyst of 1, temperature of 900 °C. Leaching conditions: temperature of 80 °C, time of 10 min, liquid-to-solid ratio of 5, stirring speed of 240 r/min, NaOH concentration of 6%.

3.1.3. Effect of Na_2CO_3 Dosage

The effect of the Na_2CO_3 dosage on the concentration factor of Pt is shown in Figure 7. When the mass ratio of Na_2CO_3 to spent catalyst was 0.5, the concentration factor of Pt

was only 8.9. As the mass ratio rose to 1, the concentration factor also increased to 79.4. The reason for this is that the increase in the ratio promoted the reaction between Al_2O_3 and Na_2CO_3, which was favorable to the transformation of Al_2O_3 in the spent catalyst into $NaAlO_2$. The phase diagram of the Na_2CO_3-Al_2O_3 system in the air condition in Figure 3 also supports the above result. Obviously, when the roasting temperature was over 900 °C, the contents of the intermediate products ($Na_2Al_{12}O_{19}$ and $NaAl_9O_{14}$) gradually dropped, while those of $NaAlO_2$ rose with the increase in the Na_2CO_3 dosage, implying the easier conversion of Al_2O_3 into $NaAlO_2$. There was no obvious increase in the concentration factor with the further increase in the mass ratio to 1.2. This is because Al_2O_3 could be fully transformed into $NaAlO_2$ at the mass ratio of 1, and excess Na_2CO_3 did not evidently improve the reaction. From the above result, it can be concluded that when the mass ratio of Na_2CO_3 to the spent catalyst was 1, the Al_2O_3 carrier in the spent Pt catalyst was effectively converted into leachable $NaAlO_2$, and Pt was concentrated in the leached residue.

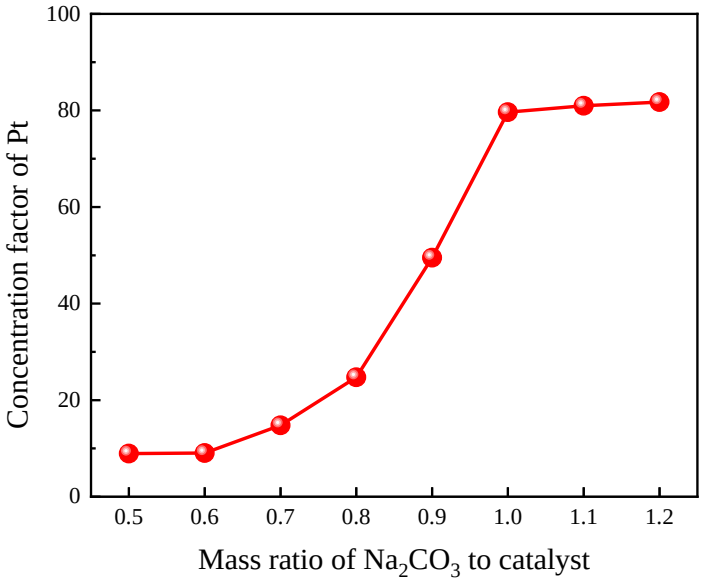


Figure 7. Effect of Na_2CO_3 dosage on the concentration factor of Pt. Roasting conditions: temperature of 900 °C, time of 2 h. Leaching conditions: temperature of 80 °C, time of 10 min, liquid-to-solid ratio of 5, stirring speed of 240 r/min, NaOH concentration of 6%.

3.2. Alkaline Leaching of Roasted Residue

3.2.1. Effect of Leaching Temperature

In this work, the spent catalyst was roasted in the presence of Na_2CO_3, and $NaAlO_2$ was newly formed while the Pt phase remained. The roasted product was leached in a dilute NaOH solution, which avoided the form of $Al(OH)_3$ due to the hydrolysis of an AlO_2^- ion. Pt was not dissolved during Al leaching, and it remained in the residue and can be easily extracted in aqua regia or hydrochloric acid plus oxidants.

The effect of leaching temperature on the concentration factor of Pt is exhibited in Figure 8. When the roasted residue was leached at 50 °C, the concentration factor of Pt was only 24.2. As the leaching temperature increased, the concentration factor gradually increased, and it reached 79.4 at 80 °C. This is because the solubility of $NaAlO_2$ increased with the increase in leaching temperature, which was beneficial to its dissolution [29]. With the further increase in leaching temperature to 90 °C, the concentration factor basically remained unchanged. Thus, the optimal leaching temperature was 80 °C.

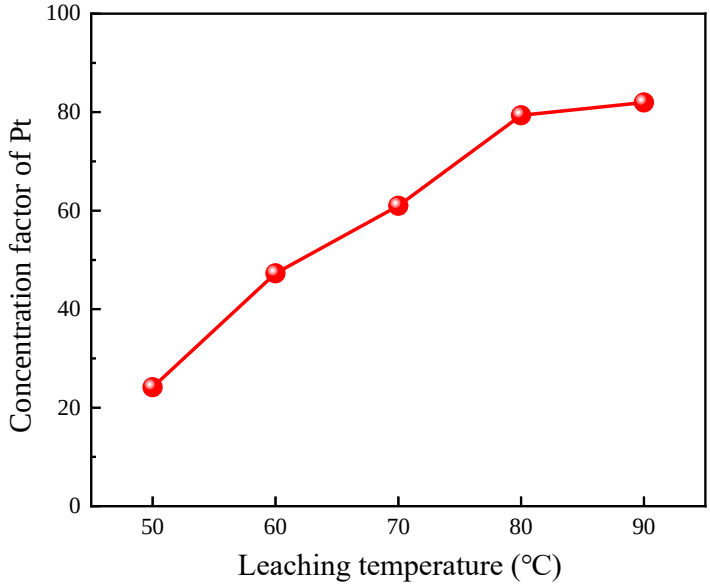


Figure 8. Effect of leaching temperature on the concentration factor of Pt. Leaching conditions: time of 10 min, liquid-to-solid ratio of 5:1, stirring speed of 240 r/min, NaOH concentration of 6%.

3.2.2. Effect of Leaching Time

The effect of leaching time on the concentration factor is shown in Figure 9. When the roasted residue was leached for 5 min, the concentration factor of Pt was only 24.2. As the leaching time increased, the concentration factor sharply increased, and it reached 79.4 when the roasted residue was leached for 10 min. When the leaching time increased to 15 min, the concentration factor only slightly increased to 82.0. On the contrary, the concentration factor dropped to 75.8 when the leaching time further rose to 20 min, indicating that the leaching percentage decreased. This is because the dissolved $NaAlO_2$ reacted with CO_2 in the air to generate $Al(OH)_3$ precipitate, and the reaction is shown in Equation (3). Thus, the optimal leaching time was 10 min.

$$2NaAlO_2 + CO_2 + 3H_2O = 2Al(OH)_3\downarrow + Na_2CO_3 \quad (3)$$

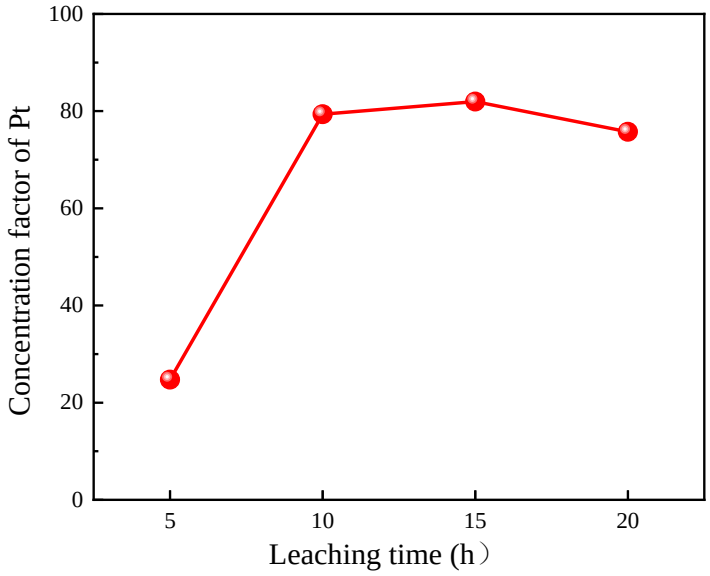


Figure 9. Effect of leaching time on the concentration factor of Pt. Leaching conditions: temperature of 80 °C, liquid-to-solid ratio of 5:1, stirring speed of 240 r/min, NaOH concentration of 6%.

3.2.3. Effect of Liquid-to-Solid Ratio

The effect of the liquid-to-solid ratio on the concentration factor is exhibited in Figure 10. When the liquid-to-solid ratio was 2, the concentration factor of Pt was only 5.9. As the liquid-to-solid ratio increased, the concentration factor gradually augmented, and it arrived at 79.4 at the liquid-to-solid ratio of 5. The reason for this is that the concentration of $NaAlO_2$ in the liquid decreased with the increase in the liquid–solid ratio [30]. As a result, the viscosity of the pulp dropped, and the diffusion rate of $NaAlO_2$ accelerated, which was conducive to its dissolution. With the further increase in the liquid-to-solid ratio to 6, the concentration factor only slightly increased to 82.0. Therefore, the optimal liquid-to-solid ratio was 5.

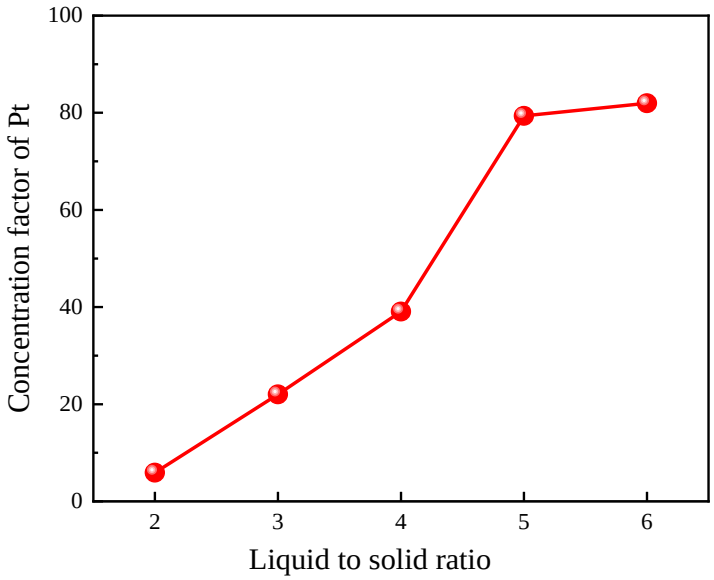


Figure 10. Effect of liquid-to-solid ratio on the concentration factor of Pt. Leaching conditions: temperature of 80 °C, time of 10 min, stirring speed of 240 r/min, NaOH concentration of 6%.

3.2.4. Effect of Stirring Speed

The effect of stirring speed on the concentration factor is displayed in Figure 11. When the stirring speed was 80 r/min, the concentration factor of Pt was only 59.6. With the increase in stirring speed, the concentration factor gradually augmented. This is because, from the kinetic point of view, increasing the stirring speed effectively reduced the resistance of external diffusion, which was conducive to the dissolution of $NaAlO_2$ in the roasted residue. The concentration factor reached up to 79.4 at the stirring speed of 240 r/min. With the further increase in stirring speed to 320 r/min, there was no obvious increase in the concentration factor. Thus, the optimal stirring speed was 240 r/min.

3.2.5. Effect of NaOH Concentration

The effect of NaOH concentration on the concentration factor is displayed in Figure 12. When the NaOH concentration was 0 (i.e., the roasted residue was leaching using water), the concentration factor of Pt was only 3.9. With the increase in the NaOH concentration, the concentration factor gradually augmented, and it arrived at 79.4 when the NaOH concentration was 6%. This is because AlO_2^- can exist stably at a high-solution pH, and it is easily hydrolyzed to generate $Al(OH)_3$ precipitate at a low NaOH concentration, which is unbeneficial to $NaAlO_2$ dissolution. With the further increase in the NaOH concentration to 8%, the concentration factor only slightly rose to 80.7. Therefore, the optimal NaOH concentration was 6%.

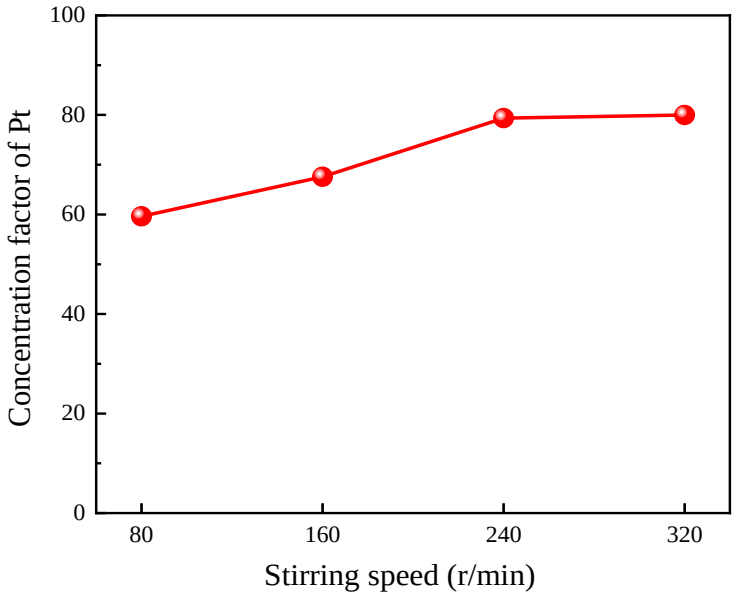


Figure 11. Effect of stirring speed on the concentration factor of Pt. Leaching conditions: temperature of 80 °C, time of 10 min, liquid-to-solid ratio of 5:1, NaOH concentration of 6%.

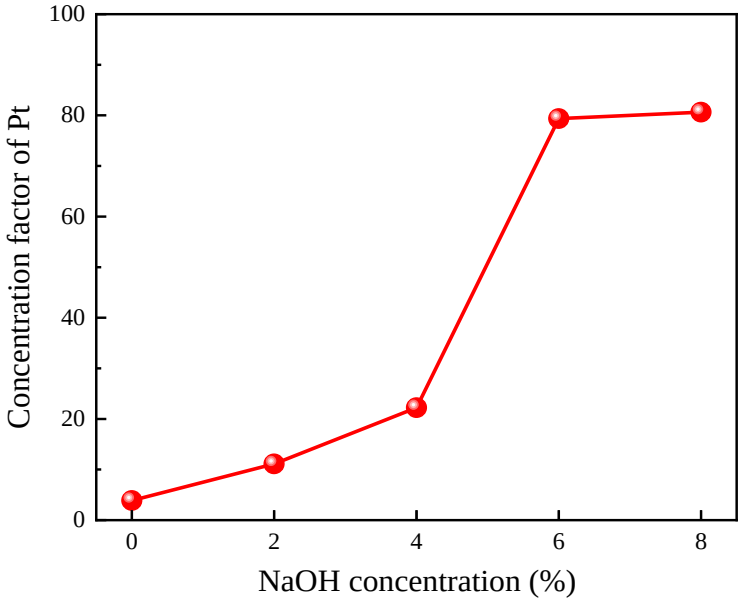


Figure 12. Effect of NaOH concentration on the concentration factor of Pt. Leaching conditions: temperature of 80 °C, time of 10 min, liquid-to-solid ratio of 5, stirring speed of 240 r/min.

3.3. Pseudo-Boehmite Preparation with Leached Solution

Pseudo-boehmite ($AlOOH{\cdot}nH_2O$, n = 0.08~0.62) is widely used in chemical, oil refining and petrochemical reactions as a catalyst carrier such as a hydrorefining catalyst carrier, reforming catalyst carrier, methanation catalyst carrier, etc. [24]. Pseudo-boehmite is dehydrated to become gamma alumina, which can also be used as a catalyst. As shown in Table 2, after NaOH leaching, the main elements in the leached solution were Na and Al, and it was suitable for preparing pseudo-boehmite via the carbonization method, as expressed in Equation (4).

$$2NaAlO_2 + CO_2 + (2n + 1)\ H_2O = 2AlOOH{\cdot}nH_2O + Na_2CO_3 \quad (4)$$

Table 2. Main components of the leached solution from alkaline leaching of the roasted residue.

Element	Na	Al	Si	Fe	Ca
Content ($mg \cdot L^{-1}$)	36,999	13,184	130	0.04	0.48

Using the proposed carbonization process, a pseudo-boehmite product with a specific surface area of 276 $g \cdot m^{-2}$ was obtained, and the Al recovery percentage arrived at 99.0%.

The appearance, XRD pattern and SEM images of the pseudo-boehmite product are exhibited in Figure 13a–d. It can be seen that the pseudo-boehmite product was a gray solid powder. Also, only the pseudo-boehmite phase was detected, and thus, the product had a high purity. In terms of microstructure, the particle was fluffy, and small and large particles clumped together. The pseudo-boehmite product can used as a material for the preparation carrier of a Pt/Al_2O_3 catalyst.

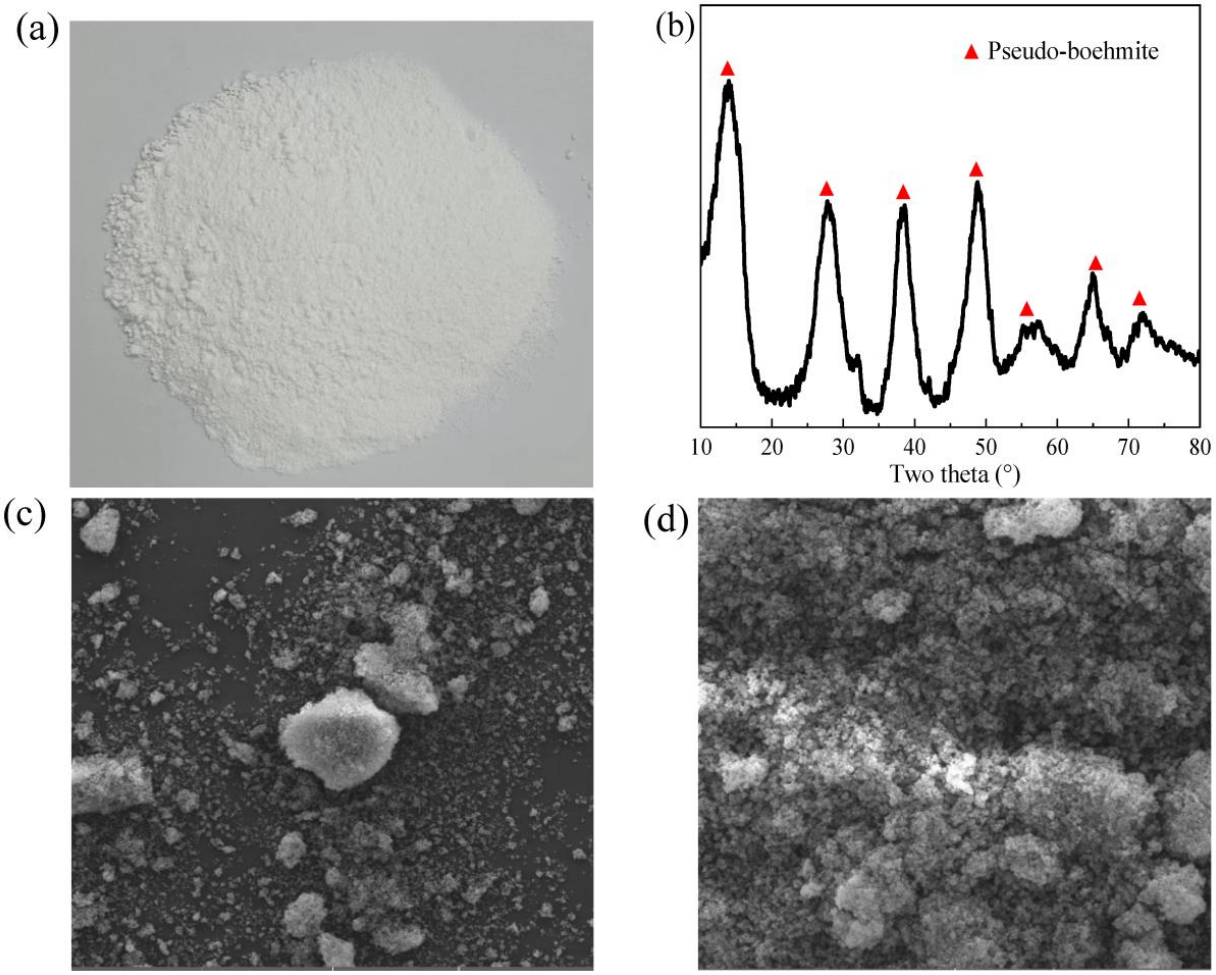


Figure 13. Appearance (**a**), XRD pattern (**b**) and SEM images (**c**,**d**) of the pseudo-boehmite product.

In addition, chemical composition analysis was performed for the leached residue, and the results indicated that the Pt content reached up to 29.4%. Thus, the leached residue is a Pt-containing concentrate. The Pt in the concentrate can be efficiently extracted via chloride leaching, as indicated in Equation (5). The dissolved Pt can finally be recovered via replacement with metal powders such as iron and zinc.

$$Pt + 16Cl^- + 2ClO_3^- + 12H^+ = 3PtCl_6^{2-} + 6H_2O \quad (5)$$

4. Conclusions

In this study, an integrated soda roasting–alkaline leaching–carbonization process was used to recover Pt and Al from a spent Pt/Al_2O_3 catalyst, and our main findings are summarized as follows.

(1) Al_2O_3 in the spent catalyst was transformed into soluble-water $NaAlO_2$ via soda roasting under the optimal conditions of the mass ratio of Na_2CO_3 to spent catalyst of 1, roasting temperature of 900 °C and roasting time of 2 h.

(2) Most of the generated $NaAlO_2$ in the roasted residue was dissolved with dilute NaOH solution, and Pt remained in the leached residue and its concentration factor achieved 79.4 under the optimal leaching conditions of temperature of 80 °C, time of 10 min, liquid-to-solid ratio of 5, stirring speed of 240 r/min and a NaOH concentration of 6%.

(3) After carbonization was performed for the $NaAlO_2$ solution, a pseudo-boehmite product with a specific surface area of 276 $g \cdot m^{-2}$ was obtained, which can used as a feed for preparing an Al_2O_3 carrier. The Al recovery percentage was up to 99.0%, and Pt was enriched in the leached residue, with a content that yielded 29.4%. Pt and Al in the spent Pt/Al_2O_3 catalyst were efficiently recovered via the proposed process, and thus, it has a bright prospect of industrial application.

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