Available online at www.sciencedirect.com
ScienceDirect
Trans. Nonferrous Met. Soc. China 32(2022) 345−353
Transactions of Nonferrous Metals Society of China
www.tnmsc.cn

Nickel and aluminium recovery from spent reforming catalyst through selective leaching, crystallization and precipitation

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Received 26 January 2021; accepted 3 September 2021

Abstract: An attempt has been made to investigate and optimize the recovery of Ni and Al through sulphuric acid (3.0−5.5 mol/L) leaching under different operating conditions. From the leaching experiments, it was possible to extract 98.5% of NiO and 40.7% of Al_2O_3 under the conditions of 5.5 mol/L H_2SO_4, reaction time of 4 h, solid-to-liquid ratio 0.2 g/mL, temperature of 358 K, particle size <100 μm, 200−250 r/min with 5.0 g catalyst dosage. The leached liquor Al was separated by selective crystallization using 1.4 mol/L KOH and Ni was separated by selective precipitation using 0.3 mol/L $H_2C_2O_4$. From the studies, it is possible to recover around 97.9% of NiO having 98.3% purity, around 25% of Al_2O_3 was also recovered as alum-(K) having 99% purity and 14.7% of Al_2O_3 as a salt of $Al-K-C_2O_4-SO_4$. Sulphuric acid was found to be a suitable leaching agent for selective leaching and it was also observed that alum-(K) can be selectively crystallized from sulphate solutions. The study also indicated the effective extraction and recovery of nickel and aluminium which were well supported by characterization studies using TG-DTA/DTG and XRD techniques.

Key words: spent catalyst; leaching; precipitation; crystallization; nickel; aluminium

1 Introduction

Reforming catalysts ($NiO-CaO-Al_2O_3$) are used in ammonia and petroleum industries for the conversion of steam reformation reaction in the primary reforming tubes and secondary reformer reactors. Most of these commercial catalysts consist of 10%−20% NiO, dispersed in a refractory support material Al_2O_3 and promoted with alkaline earth oxides such as CaO/MgO [1]. After periodical use, these catalysts have to be discarded as spent catalysts due to decrease in their conversion efficiency and mechanical strength. Generation of huge quantity (150000−170000 t/a) of the spent hydro-processing catalysts is also reported in the Refs. [2−5]. These spent catalysts being the rich source of Ni and other metals need to be recycled to recover the metal values. Although different pyro-based processes have been used earlier, recent developments involve hydrometallurgical processes for their recovery. In this regard many reagents, such as NaOH, H_2SO_4, NH_3, $(NH_4)_2SO_4$ and oxalic acid with H_2O_2 and $Fe(NO_3)_2$ have been employed for the extraction of nickel [6−8]. From the studies reported it may be seen that 73%−99% of crucial metals were extracted using ammonia, caustic and acid leaching and Na_2CO_3 [9]. CHAUDHARY et al [10] reported the extraction of nickel (~73%) at 80 °C from a spent catalyst (17.7% Ni) employing HCl leaching. From a study carried out by AL-MANSI and ABDEL MONEM [11], it was possible to obtain ~99% Ni extraction with 50% sulfuric acid at 100 °C in more than 5 h from a

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DOI: 10.1016/S1003-6326(22)65799-6

spent reforming catalyst. SAHU et al [12] achieved a high Ni leaching (~98%) with H_2SO_4 from a spent fertilizer catalyst ($NiO-SiO_2-Fe_2O_3-Al_2O_3$). A combined acid ($HNO_3/H_2SO_4/HCl$) leaching study carried out by LAI et al [13], indicated a recovery of 99% V and Ni, and ~90% Mo. In another study by LEE et al [14], it was possible to recover 98% NiO with 100% purity by the precipitation and separation of Al with Na_2CO_3 solution at pH ~5.4 from a sulphuric acid-leached solution of spent Raney-nickel catalysts ($NiO-Al_2O_3$). NAZEMI et al [15,16] showed a leaching process to recover about 99% Ni by H_2SO_4. Nickel ammonium sulphate could partly be precipitated by NH_3 and nickel in the supernatant was enriched using SX with D2EHPA. PARHI et al [17] proposed the diffusion-controlled model to represent the leaching kinetics, and they have also reported that over 99.9% Ni was leached using HCl at 50 °C from spent $Ni-Al_2O_3$ catalyst. SHEIK et al [18] and RAMOS-CANO et al [19] studied the dissolution kinetics of Ni from spent catalysts in 5 mol/L HNO_3 at 90 °C with 90% conversion and in 50% nitric acid at 65 °C with 85% extraction of NiO. ABDEL-AAL and RASHAD [20] studied nickel leaching kinetics on spent nickel catalysts ($NiO-Al_2O_3$) with 50% H_2SO_4 and extracted 94% NiO at 358 K. VALVERDE et al [21] studied the isolation of elements (>98%) through solvent extraction and precipitation from a sulphuric acid-leached solution of spent catalyst ($NiMo/Al_2O_3/CoMo$) at 363 K. CHAUHAN et al [3], VUYYURU et al [4], GOEL et al [22], and OZA and PATEL [23] have shown more than 95% Ni recovery as $NiSO_4$ or $Ni(NO_3)_2$ from spent nickel catalysts by EDTA leaching. MUZAYANHA et al [24] studied Li−Ni−Al oxide material from waste cathode via mixed metal hydroxide using strong acids as leaching agents and NaOH as a precipitant at pH 11−12. BATTI and MANDRE [25] investigated the spent methanation catalyst and recovered 99% NiO with 98.9% purity by selective and oxidative leaching with HNO_3; in the study, Fe (III) precipitated at pH~3.5 to 4.5 with an alkaline solution and Ni was selectively precipitated using oxalic acid.

From a detailed literature survey, it may be seen that there is a need to develop suitable leaching and separation of these minerals. Hence, in the present study, an attempt has been made to carry out selective leaching of Ni, Al and their separation in the acidic medium. The results obtained from the investigation were supported by the detailed characterization studies.

2 Experimental

2.1 Materials and chemicals

The spent reforming catalysts used in this study were collected from Rashtriya Chemicals and Fertilizers (RCF), Thal Unit (Mumbai, India). These spent catalysts were dried at 393 K in a hot air oven for 4 h to remove moisture, further ground in central peripheral ball mill and screened through a set of standard sieves to obtain the particle size <100 μm. During the study, analytical reagent grade chemicals such as sulphuric acid (3.0−5.5 mol/L) for selective leaching, 1.4 mol/L potassium hydroxide for selective crystallization of alum-(K), 0.3 mol/L oxalic acid for selective precipitation of nickel, 2% sodium hydroxide for neutralization of effluent and distilled water for preparation of reagent solutions were used throughout the process.

2.2 Analysis

2.2.1 X-ray diffraction (powder/single crystal) and thermal analysis

The phase identification of metal oxides present in spent reforming catalyst and recovered material was determined by powder XRD (Model: X’pert Pro, Panalytical) equipped with a Cu target (Cu $K_{\alpha 1}$ radiation, λ=1.540598 Å) operated at 45 kV. The diffraction peaks assigned to NiO (JCPDS No. 01−072−1464) [22,26], Al_2O_3 (JCPDS No. 43−1484), CaO, $NiAl_2O_4$, $CaAl_2O_4$ [27], and deposited carbon [26]. The diffraction spectrogram obtained from the study is given in Fig. 1. Further, single-crystal XRD (SuperNova, Single source at offset/far, EosS2; radiation type Mo K_α and wavelength is 0.71073) study was carried out to identify the crystal phase present in the recovered alum-(K), the thermal studies (thermogravimetry, TG; differential thermal analysis, DTA; and differential thermogravimetry, DTG) were carried out using $N_{2(g)}$ purge and protective gas with the heating rate at 10 K/min in the TG-DTA/DTG instrument (Model: No-NetzschSta 449F3) to obtain compound information of recovered nickel oxalate and alum-(K).

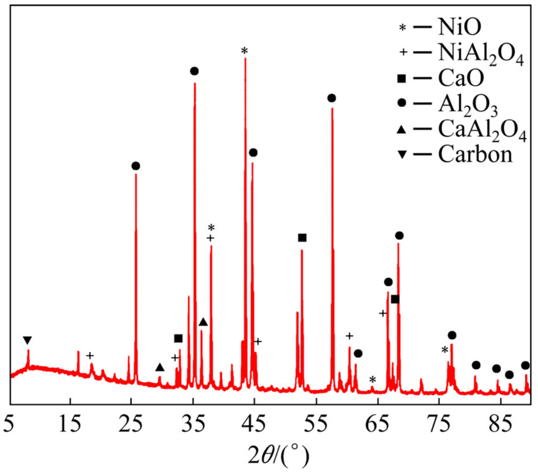

Fig. 1 XRD pattern of spent reforming nickel catalyst

2.2.2 Chemical analysis

Chemical analysis was done by using ED-XRF (Energy Dispersive X-Ray Fluorescence, PAN analytical, Epsilon-1), atomic absorption spectrophotometer (AAS; Thermo Fisher, Model No. Pu9000) and EDTA complexometric methods [25,28]. Sulphate was estimated by the gravimetric method with 5% $BaCl_2$ solution [29]. Results of the studies are given in Table 1.

2.2.3 Experimental methods of leaching and recovery

Isothermal leaching experiments were carried out in a 500 mL round bottom flask fitted with a reflux condenser which was dipped in an oil bath placed on a magnetic stirrer; and it was connected with an automatic temperature controller and indicator through resistance temperature detector (RTD) (accuracy ±1 K). The second end of RTD was immersed in an oil bath to measure the reaction temperature. In this process, the fixed conditions were as follows, i.e., spent catalyst particle size <100 μm, stirring speed 200−250 r/min and catalyst dosage of 5.0 g. In the first stage, nickel and aluminium were leached with sulphuric acid (3.0−5.5 mol/L). In the second stage, about 25% Al_2O_3 was selectively crystallized as alum-(K) from leached solution using 1.4 mol/L KOH and separated. In the third stage, Ni was selectively precipitated using 0.3 mol/L oxalic acid in an acidic medium by stabilizing the rest of aluminium in the aqueous state [30,31]. In the fourth stage, the rest of aluminium, sulphate, potassium and excess oxalate were recovered from the filtrate of nickel oxalate precipitate by open-pan crystallization process by maintaining the specific gravity of ~1.40.

3 Results and discussion

3.1 Leaching and separation of minerals

3.1.1 Leaching

Leaching experiments were performed with sulphuric acid in an isothermal reaction vessel. The NiO and Al_2O_3 were leached and generated a sulphate solution of Ni and Al according to Reactions (1) and (2).

$$NiO_{(s)}+H_2SO_{4(aq)}+5H_2O \rightarrow NiSO_4.6H_2O_{(aq)} \quad (1)$$

$$Al_2O_{3(s)}+3H_2SO_{4(aq)}+15H_2O \rightarrow Al_2(SO_4)_3\cdot 18H_2O_{(aq)} \quad (2)$$

From the experiments, it was observed that the leaching of Ni and Al increased with an increase in the concentration of sulphuric acid (3.0−5.5 mol/L), reaction time (1−4 h), temperature (333−358 K), and decreased with increasing solid-to-liquid ratio (0.20−0.58 g/mL); stirring speed was maintained at 200−250 r/min. From the studies suitable leaching conditions can be obtained under the conditions of 5.5 mol/L H_2SO_4, 0.2 g/mL solid-to-liquid ratio, stirring speed 200−250 r/min, particle size <100 μm at 358 K of temperature for 4 h of reaction time; the results obtained are given in Tables 2−5. From the results, it was possible to extract 98.5% of NiO and 40.7% of Al_2O_3. It was also noted that the calcium was not found in the leached solution, which may be attributed to the formation of acid-insoluble gypsum [32].

3.1.2 Selective crystallization of alum-(K)

On cooling the leached solution, salt deposition was observed, which may be due to the high concentration of aluminium present in the solution [33]. Further, the solution was diluted

Table 1 Chemical analysis of spent reforming catalyst by different methods (wt.%)

<table>
<tr><td rowspan="2">XRF</td><td>Ni</td><td>Ca</td><td>Al</td><td>Fe</td><td>Na</td><td>Si</td><td>P</td><td>Cr</td><td>Mg</td></tr>
<tr><td>8.94</td><td>1.18</td><td>44.68</td><td>0.24</td><td>0.51</td><td>0.43</td><td>0.25</td><td>0.03</td><td>0.16</td></tr>
<tr><td rowspan="2">Classical quantitative method</td><td colspan="2">NiO</td><td colspan="3">CaO</td><td colspan="2">Al_2O_3</td><td colspan="2">*LOI at 1173 K</td></tr>
<tr><td colspan="2">11.8</td><td colspan="3">1.7</td><td colspan="2">84.5</td><td colspan="2">0.14</td></tr>
</table>

* LOI: Loss on ignition

Table 2 Effect of acid concentration on leaching of NiO and Al_2O_3 under conditions of particle size <100 μm, stirring speed 200−250 r/min, sample dosage 5.0 g, temperature 358 K, time 4 h, and solid-to-liquid ratio 0.2 g/mL

Acid concentration/ $(mol \cdot L^{-1})$	Dissolution/%	
	NiO	Al_2O_3
3.0	65.8	26.9
3.5	74.8	28.4
4.0	80.4	31.8
4.5	87.9	34.5
5.0	94.1	38.2
5.5	98.5	40.7

Table 3 Effect of temperature on leaching of NiO and Al_2O_3 under conditions of particle size <100 μm, stirring speed 200−250 r/min, sample dosage 5.0 g, time 4 h, H_2SO_4 5.5 mol/L, and solid-to-liquid ratio 0.2 g/mL

Temperature/K	Dissolution/%	
	NiO	Al_2O_3
333	72.4	19.7
338	77.9	23.1
343	83.4	26.8
348	86.8	31.4
353	94.7	37.1
358	98.5	40.7

Table 4 Effect of reaction time on leaching of NiO and Al_2O_3 under condition of particle size <100 μm, stirring speed 200−250 r/min, sample dosage 5.0 g, temperature 358 K, H_2SO_4 5.5 mol/L, and solid-to-liquid ratio 0.2 g/mL

Time/h	Dissolution/%	
	NiO	Al_2O_3
1	65.4	19.4
2	82.9	25.6
3	93.5	33.7
4	98.5	40.7

using distilled water to prevent the deposition of unwanted salts and to promote the formation of a homogeneous solution. The analysis of the solution indicated 0.4% NiO (m/v), 1.4% Al_2O_3 (m/v), pH~1.5 having a specific gravity of 1.07 at 305 K. This indicated that, the concentration (m/v) of Al_2O_3 was higher than that of NiO (i.e. 1.4% $Al_2O_3 \gg 0.4\%$ NiO and the ratio of Al_2O_3 to NiO ≥ 3.5).

Table 5 Effect of solid-to-liquid ratio on leaching of NiO and Al_2O_3 under conditions of particle size <100 μm, stirring speed 200−250 r/min, sample dosage 5.0 g, temperature 358 K, time 4 h, and H_2SO_4 5.5 mol/L

Solid-to-liquid ratio/$(g \cdot mL^{-1})$	Dissolution/%	
	NiO	Al_2O_3
0.58	82.6	17.7
0.45	87.3	25.6
0.31	94.2	34.3
0.2	98.5	40.7

For selective crystallization of aluminium, the pH of the solution (100 mL) was increased from ~1.5 to ~2.81 using 1.4 mol/L KOH (~10 mL) in a graduated open-pan with a stirring rate around 200 r/min [34]. The mixed solution was evaporated and cooled for 24 h. Selective crystallization of alum-(K) was achieved by vaporizing ~27.3 vol.% of 110 mL mixed solution and the results obtained are given in Table 6. From this, it was possible to recover 25% of Al_2O_3 as alum-(K) having 99% purity. The formation of alum-(K) crystal is given in Reaction (3) and the chemical analysis of recovered alum-(K) is given in Table 7.

$$Al^{3+}_{(aq)}+2SO_4^{2-}{}_{(aq)}+K^+_{(aq)}+12H_2O_{(l)} \rightleftharpoons KAl(SO_4)_2 \cdot 12H_2O_{(s)} \quad (3)$$

3.1.3 Selective precipitation of nickel

After completion of the above tests, nickel was selectively precipitated from Ni−Al filtrate by using 0.3 mol/L oxalic acid as a precipitant at different stoichiometric concentrations of $C_2O_4^{2-}$ to Ni. For this purpose, pH of the solution was maintained at 1.50−2.0, in this pH range, Al has high solubility and is independent of the concentration of

Table 6 Selective crystallization of alum-(K) from leached Ni−Al−SO_4 solution (~100 mL) using 1.4 mol/L KOH (~10 mL) at pH ~2.81 by vaporization of 110 mL mixed solution

Volume vaporized/vol.%	Al_2O_3 recovered as alum-(K)/wt.%
10	6
15	12
20	18
25	23
27.3	25

Table 7 Chemical analysis of recovered alum-(K) using XRF (wt.%)

Ni	Ca	Al	K	S	Si	Fe	P
0.01	0	5.32	7.96	13.41	0.07	0.01	0.13

oxalic acid [34,35]. From the Ref. [36], these aqueous Al ions were noticed to have solubility properties similar to hydroxo aluminate $[Al(OH)_4^-]$ ions formed in the Bayers alkaline leaching of alumina. Nickel precipitation was carried out at 1:1, 1.5:1 and 2:1 of oxalate to metal stoichiometry with oxalic acid by maintaining a stirring speed of 200−250 r/min, temperature range from 303 to 343 K and reaction time of 1−1.5 h (Reaction (4)) [31,35]. From the tests, it was found that 2:1 of oxalate to nickel at 333 K for 1 h, 99.4% nickel was stoichiometry precipitated with high settling rate and the results obtained are given in Table 8. The precipitate was dried at 383 K for 4 h, then decomposed into nickel oxide at 873 K (Reaction (5)). From the study, it was found that about 97.9% NiO was recovered with 98.3% purity. Chemical analysis of recovered nickel oxide is given in Table 9.

$$NiSO_4+H_2C_2O_4\cdot 2H_2O\rightarrow NiC_2O_4\cdot 2H_2O+H_2SO_4 \tag{4}$$

$$NiC_2O_4\cdot 2H_2O\rightarrow NiO+2CO_2+2H_2O \tag{5}$$

Table 8 Selective precipitation and recovery of nickel oxide from Ni−Al−SO$_4$ solution obtained from filtrate of recovered alum-(K) using 0.3 mol/L oxalic acid at different oxalate-to-nickel stoichiometric ratios at 333 K for 1 h

Stoichiometric ratio of oxalate to Ni	NiO recovery/%
1:1	39.7
1.5:1	63.8
2:01	99.4

Table 9 Chemical analysis of recovered nickel oxide using XRF (wt.%)

Ni	Ca	Al	K	Fe	S	Cr	Mg
77.24	0.01	0.11	0.33	0.11	0.13	0.02	0.12

3.1.4 Crystallization of aluminium, sulphate, potassium and excess oxalate

Further, nickel-free Al filtrate (NiO<0.02×10^{-6}, pH ~1.5) was concentrated in the open-pan crystallization process under quiescent conditions and kept for 24 h without any disturbance to obtain crystals of Al compound. From this process, 14.7% Al_2O_3 was recovered and the chemical analysis results are given in Table 10.

Table 10 XRF analysis of recovered crystals of aluminium, sulphate, potassium and excess oxalate obtained from filtrate of nickel oxalate (wt.%)

Ni	Ca	Al	K	S	Si	Fe	Mg	Cl
0	0.04	9.61	6.98	11.24	0.01	0.01	0.03	0.08

3.2 Characterization of recovered materials

3.2.1 Thermogravimetric studies of nickel oxalate

Characterization of the recovered nickel oxalate was carried out by TG-DTA/DTG and the results are given in Fig. 2. From the figure, it may be seen that the mass loss observed up to 383.1 K and 523.9 K is attributed to the moisture loss and removal of water of crystallization, respectively. The endothermic peak found at 514.8 K with a mass loss of 18.3% is a close approach with the theoretical value of 19.7%, which may be attributed to dehydration of hydrated nickel oxalate (Reaction (6)).

$$NiC_2O_4\cdot 2H_2O_{(s)}\rightarrow NiC_2O_{4(s)}+2H_2O_{(g)} \tag{6}$$

In the second stage, mass losses of 39.6% and 33.3% of residue were observed between 523.9 and 662.2 K with an endothermic peak at 642.6 K corresponding to the decomposition of dehydrated nickel oxalate (Reaction (7)) [37]. On further heating around 1023.5 K, the residue mass was gained and remained constant at 41.1% which is nearer to the theoretical value of 40% (Reaction (8)) [27,31,38].

$$NiC_2O_{4(s)}\rightarrow Ni_{(s)}+2CO_{2(g)} \tag{7}$$

$$Ni_{(s)}+CO_{2(g)}\rightarrow NiO_{(s)}+CO_{(g)} \tag{8}$$

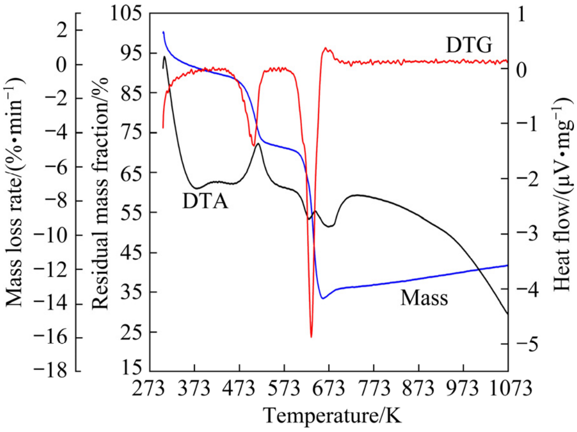

Fig. 2 TG-DTA/DTG curves of recovered nickel oxalate

3.2.2 X-ray diffraction studies of recovered nickel oxide

Apart from the above studies, characterization was also carried out by using XRD test and the result is given in Fig. 3. The diffraction peaks found with high intensity indicated bunsenite (NiO) phase [27,35], the low-intensity peaks indicated alum-(K) [39,40] and t-spinel ($NiAl_2O_4$) [26], it was also observed that, the absence of diffraction lines correspond to Al_2O_3 [35] and CaO [26]. The XRD pattern of recovered NiO is well-indexed to JCPDS. Here it may be observed that the above results supported the ED-XRF and TG-DTA/DTG studies for the effective separation and recovery of NiO from the spent reforming catalyst.

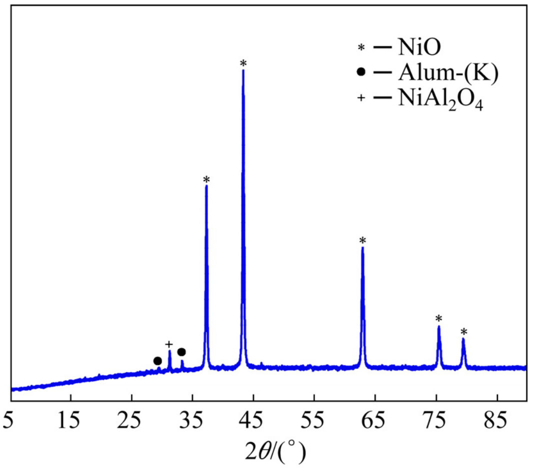

Fig. 3 X-ray diffraction pattern of recovered nickel oxide

3.2.3 Thermogravimetric studies of recovered alum-(K)

The results of the thermogravimetric studies carried out on recovered alum-(K) are given in Fig. 4. From the figure it may be seen that a mass loss of 43.2% in the temperature range from 344.1 (T_i) to 525.1 K (T_f) with peak temperature at 402.2 K has a close agreement with the theoretical value of 45.6%, which indicates the loss of water of crystallization (Reaction (9)). Further, in the second stage, mass loss of 20.2% and residue of 35.9% were found in the temperature range from 973.1 (T_i) to 1170.8 K (T_f) with a peak temperature at 1066.8 K, which may be attributed to the release of gases due to decomposition of $KAl(SO_4)_2$ [37]. The endothermic peaks of DTA curve at 404.6, 504.7 and 1069 K correspond to Reactions (9)−(11) respectively, as shown in Fig. 4.

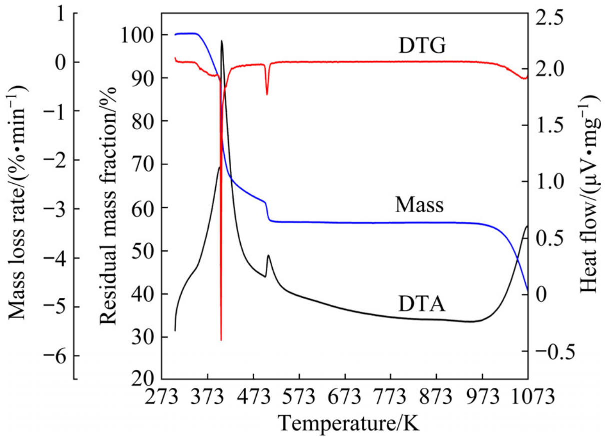

Fig. 4 TG-DTA/DTG curves of recovered potassium alum

$$KAl(SO_4)_2\cdot 12H_2O \rightarrow 12H_2O_{(g)}+KAl(SO_4)_2 \quad (9)$$

$$2KAl(SO_4)_2 \rightarrow Al_2(SO_4)_3+K_2SO_4 \quad (10)$$

$$2KAl(SO_4)_2 \rightarrow K_2SO_4+Al_2O_3+3SO_{2(g)}+3/2O_{2(g)} \quad (11)$$

3.2.4 X-ray diffraction studies of recovered alum-(K)

From the single crystal XRD study of recovered alum, axes angle (θ) and cell length/axes are well indexed to the standard cubic lattice parameters of alum-(K) (Table 11) [40,41]. This indicates that the XRD results support the ED-XRF and TG-DTA/DTG studies for the effective recovery and purity of alum-(K).

Table 11 Unit cell parameters of recovered alum-(K) using single-crystal XRD

Experimental parameter		Standard parameter	
Cell length/Å	Axes angle/(°)	Cell length/Å	Axes angle/(°)
12.1649 (*a*)	90.0 (α)	12.1640 (*a*)	90.0 (α)
12.1649 (*b*)	90.0 (β)	12.1640 (*a*)	90.0 (β)
12.1649 (*c*)	90.0 (γ)	12.1640 (*a*)	90.0 (γ)
Volume = 1800.2 $Å^3$		Volume = 1799.82 $Å^3$	

* alum-(K) = Potassium aluminum sulphate [$KAl(SO_4)_2\cdot 12H_2O$]

From the studies carried out, a schematic process flow diagram developed for the leaching and recovery of nickel and aluminium from spent reforming catalyst composed of NiO−CaO−Al_2O_3 is given in Fig. 5.

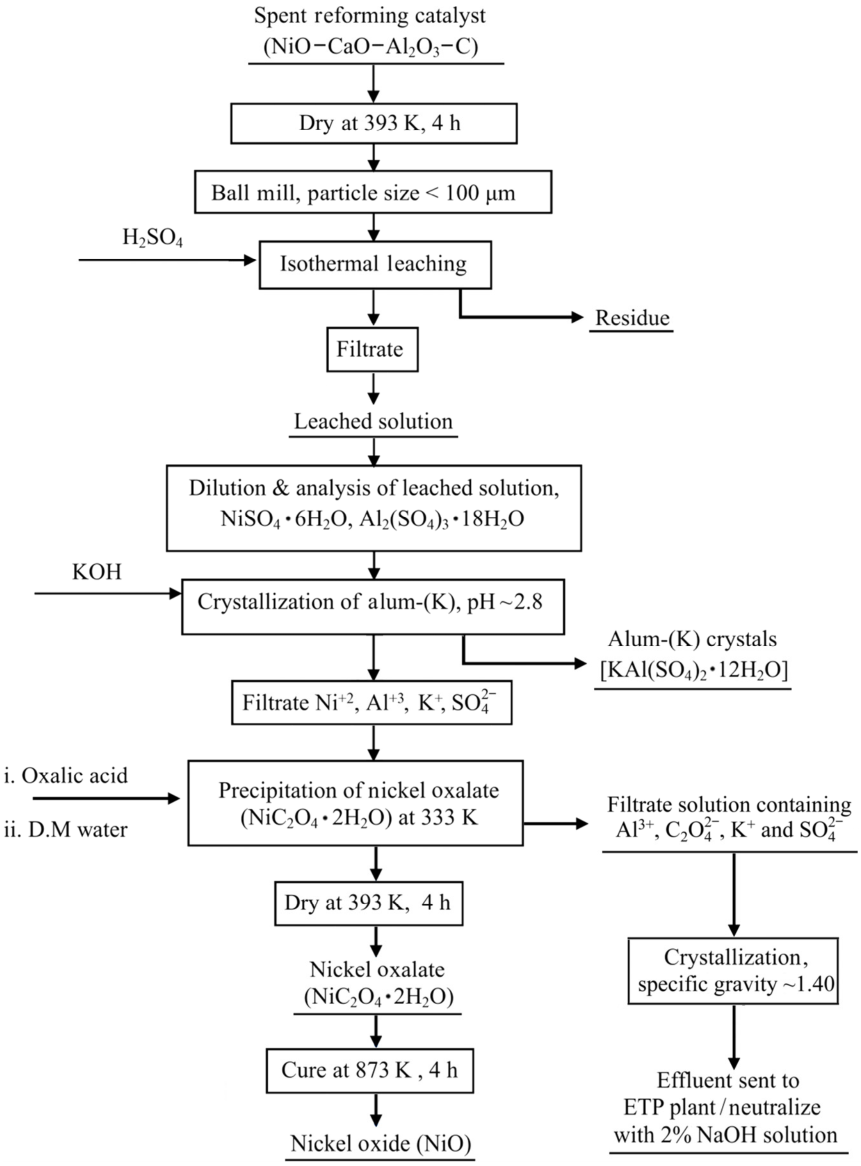

Fig. 5 Process flow diagram for recovery of nickel and aluminium from spent catalyst

4 Conclusions

(1) Sulphuric acid was found to be a suitable leaching agent for selective extraction of nickel and aluminium from a material composed of $NiO-Al_2O_3-CaO$. This may be attributed to the formation of acid-insoluble gypsum, soluble nickel and aluminium sulphates.

(2) Aluminium was selectively crystallized at pH ~2.8 as alum-(K). During the process, Ni has remained in the aqueous state due to the high concentration and low solubility of alum-(K) than nickel sulphate. Nickel was selectively precipitated and separated as $NiC_2O_4\cdot2H_2O$ from the filtrate of alum-(K).

(3) The aluminium, sulphate, potassium and excess oxalates present in the final filtrate were effectively recovered by the open-pan crystallization process.

(4) From the studies, it was possible to recover about 98.5% NiO and 40.7% Al_2O_3; about 97.9% NiO was recovered with 98.3% purity, 25% Al_2O_3 was recovered as alum-(K) with 99% purity and 14.7% Al_2O_3 was recovered as a salt of $Al-K-C_2O_4-SO_4$ with less than 1% of Ca, Ni, Fe and Mg impurities.

(5) Recovered $NiC_2O_4\cdot2H_2O$, NiO and alum-(K) were well characterized with TG-DTA/DTG and XRD studies. The study also indicated the effective extraction, separation and recovery of nickel and aluminium, which created the

opportunity to reuse/recycle the spent reforming catalyst.

Acknowledgments

The authors are thankful to the Projects & Development India Limited, Noida, and XRD Laboratory of IIT-Kanpur and Central Research Facility of IIT (Indian School of Mines), Dhanbad.

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通过选择性浸出、结晶和沉积回收废重整催化剂中的镍和铝

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摘　要：研究和优化不同工艺条件下硫酸(3.0~5.5 mol/L)浸出回收 Ni 和 Al 的工艺。浸出实验表明，在 H_2SO_4 浓度 5.5 mol/L、反应时间 4 h、固液比 0.2 g/mL、温度 358 K、粒径<100 μm、搅拌速度 200~250 r/min、催化剂用量 5.0 g 的条件下，可提取 98.5%的 NiO 和 40.7%的 Al_2O_3。浸出液中的 Al 用 1.4 mol/L KOH 选择性结晶分离，Ni 用 0.3 mol/L $H_2C_2O_4$ 选择性沉淀分离。此法可回收约 97.9%的 NiO，纯度达 98.3%；约 25%的 Al_2O_3 以明矾(纯度为 99%)形式回收，14.7%的 Al_2O_3 以 Al−K−C_2O_4−SO_4 盐的形式回收。研究结果表明，硫酸是一种合适的选择性浸出溶剂，而且可以从硫酸盐溶液中选择性结晶出明矾。TG-DTA/DTG 和 XRD 表征研究证明，本工艺可以有效地提取和回收镍和铝。

关键词：废催化剂；浸出；沉积；结晶；镍；铝

(Edited by Xiang-qun LI)