Total Recycling of All Constituent Metals from a Spent Auto-Catalyst

Ha Bich Trinh[1,2], Sookyung Kim[1,2], Rajiv Ranjan Srivastava[3], *Jae-chun Lee[1,2]

[1]Resources Recycling, Korea University of Science and Technology (UST), Daejeon, 34113, Republic of Korea
[2]Mineral Resources Research Division, Korea Institute of Geoscience and Mineral Resources (KIGAM), Daejeon 34132, Republic of Korea
[3]Research & Product Development, TAE-HYUNG Recycling, Gimcheon-si, Gyeongsangbuk-do 39066, Republic of Korea
Keywords: Spent Auto-Catalyst, Total Recycling, PGMs, Cordierite, Cerium
Presenting author email: jclee@kigam.re.kr

1. Introduction

Typical recycling of spent auto-catalyst by pyro-metallurgical routes or hydro-metallurgical routes simply emphasizes on effective PGMs extraction without any attempt to dissolve cordierite substrate or recovery of cerium oxide. The major reason is the property of these materials which are resistant to the acidic and oxidizing environment. This provides the motivation to investigate an alternative approach that could reclaim not only PGMs but also other materials from spent auto-catalyst including the substrate components (Al and Mg) and cerium oxide, which satisfies all requirements of high recycling efficiency, worthy of economic benefits and friendly environmental processing.

2. Experimental

2.1. Materials

The spent auto-catalyst of the honeycomb structure used in the study (Fig. 1) was ground and sieved to collect the sample of −0.212 mm size particles . Fire assay method was used for analyzing the PGMs contents, while Na_2O_2 fusion followed by wet-dissolution method was adopted for the analysis of substrate and additive metals. Thus analyzed metal contents in the sample used in this study are given in Table 1.

Figure 1. Images of spent auto-catalyst with honeycomb structure used in this study.

2.2. Methods

Two processes have been studied: (i) pre-reduction of PGMs followed by HCl leaching and (ii) NaOH roasting followed by H_2SO_4 leaching, and then cementation of PGMs and HCl leaching of the cemented products; H_2SO_4 leaching of the residue, and precipitation of Ce with oxalate and then calcination.

Table 1: Chemical compositions of spent auto-catalyst sample.

Metals	Al	Mg	Si	Ce	Pt	Pd	Rh
Composition (%)	23.3	4.26	12.3	10.8	1290 ppm	766 ppm	149 ppm

3. Results and discussion

(i) The hydrometallurgical process using pre-reduction (15 vol.% HCOOH, 10% pulp density, 60 °C, and 60 min) followed by HCl leaching (2.0 M HCl, 1.5 M $NaClO_3$, 90 °C, 120 min) has yielded about 95% PGMs. (ii) The pyro/hydro-metallurgical process (NaOH roasting at ≥ 600 °C with a mole ratio of cordierite to NaOH = 1:17, 60 min, in which the pyro-treatment has transformed the refractory cordierite to soluble $NaAlO_2$ and Na_2MgSiO_4; leaching with 1.0 M H_2SO_4 at 90 °C in 60 min) has yielded more than 95% Al and Mg. The co-leached PGMs (6% Pt, > 23% Pd & Rh) were cemented using 0.5 g/L Al^0 at 90 °C. The PGMs and CeO_2 in the obtained concentrate were separated by leaching PGMs (> 97%) in 6.0 M HCl with 2.0 M $NaClO_3$, leaving CeO_2 in the residue. Baking the residue in H_2SO_4 at 250 °C transformed CeO_2 to acid soluble $Ce(SO_4)_2$. Calcination of $Ce_2(C_2O_4)_3 \cdot 10H_2O$, precipitated from the sulfate solution has produced pure CeO_2. The kinetics detailing the possible reaction mechanism of the select process has been investigated.

4. Conclusions

Although the pre-reduction followed by the acid leaching has shown to recover most PGMs, the pyro/hydro-metallurgical process has proven to be potentially a more promising process, since the total recycling of all valuable metals was possible from the spent auto-catalyst (Fig. 2)

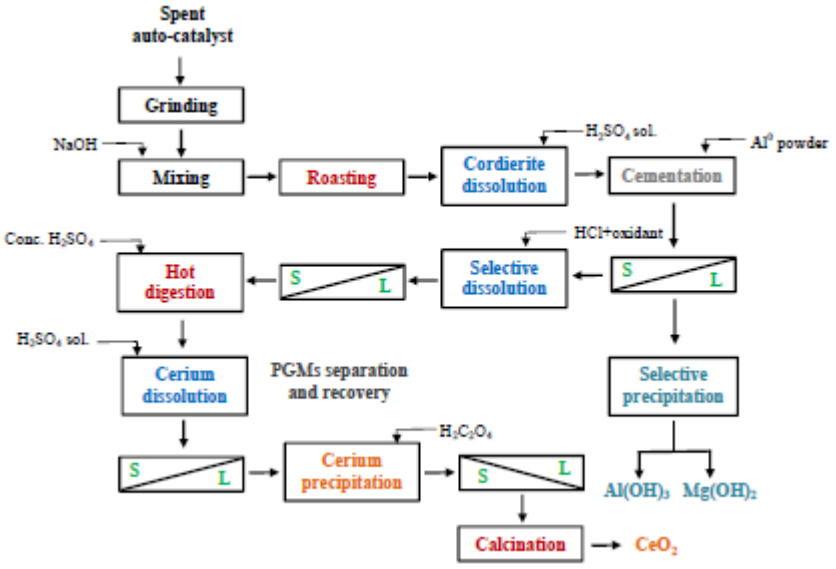

Figure 2. Flow-sheet of total recycling process of All Constituent Metals from a Spent Auto-Catalyst.