



Selective dissolution of copper from copper-chromium spent catalyst by baking–leaching process



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ABSTRACT

The selective leaching of copper from the spent Cu-Cr catalyst was carried out with H₂SO₄. The effect of different parameters such as acid concentration, pulp density, temperature and particle size on leaching was investigated. The maximum extraction of copper and chromium was 67.25 and 2.3%, respectively at particle size 45–53 μm, pulp density 2.5%, temperature 90 °C, time 180 min. Therefore, baking followed by leaching approach was adopted for dissolution of spent copper-chromium catalyst using H₂SO₄ to enhance the metal leaching efficiency. At the optimum baking–leaching condition i.e. baking time 2 h, baking temperature 300 °C, baking acid concentration 0.5 M, leaching temperature 35 °C, time 60 min, [H₂SO₄] 4%, P.D. 2.5%, the extraction of copper and chromium was 99.9% and 1.2%, respectively, ensuring the selective dissolution of copper. The XRD and Fe-SEM-Edax characterization analysis of typical samples (original, baked mass and typical residue) were compared and reported. The XRD and Fe-SEM-Edax analysis of the baked mass indicated the complete sulfation of copper and chromium by H₂SO₄ yielding CuSO₄ (H₂O) and (Cr)₂(SO₄)₃, respectively in solid phase. The absence of XRD peaks corresponding to CuSO₄·H₂O in the final typical leach residue (obtained at optimum baking–leaching condition) confirmed the complete dissolution of copper from Cu-Cr catalyst.

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1. Introduction

The rapid industrialization due to the growing demand of metals such as Cu, Co, Cr, and Ni in day to day life leads to the depletion of the primary sources. Therefore, reserve amount of primary sources bearing above metal values decreases [1,2]. The secondary resources such as spent catalysts, e-wastes, spent magnets, and spent batteries [1,3–6] which contain a considerable amount of various metals (Cu, Ni, Cd, Co, Li, Cr, Au, PGM group metals, rare and rare earth metals), are becoming the potential sources for above metals. The spent catalysts are of various kinds depending on their nature, composition and applications [2,6]. The catalyst namely copper-chromium composite oxides is widely used as a catalyst for hydrogenation, dehydrogenation, and alkylation processes [7–10]. These catalysts basically act as active

catalysts for complete oxidation of carbon monoxide and hydrocarbons to carbon dioxide [11,12]. The mixed type of copper-chromium oxide phases have good use in the simultaneous removal of nitric oxide and carbon monoxide from exhaust gases [11]. However, these catalysts gradually lose their activity after certain cycles of use due to coking, poisoning by metals, sulfur, or halides or loss of surface area due to sintering at high temperatures. These are then discarded and termed as spent catalysts [13]. These spent catalysts are hazardous in nature and their direct disposal to environment is highly restricted [14–16]. So much attention is being paid towards development of processes for extraction of metal values to minimize landfill space and to prevent pollution in land disposal [1,15,17,18]. The copper-chromium spent catalyst has more economic importance since it contains 42% copper and 36% chromium [19] and a suitable technology is urged to recover and recycle the metal values from this catalyst.

A considerable amount of work has been done by number of researchers on processing of spent catalyst [2,17–22] which involved either pyrometallurgical or hydrometallurgical process or combination of both the processes. The hydrometallurgical

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leaching method has been extensively used for dissolution of metal values from several sources [1,2,18,21]. But in some cases, direct leaching may not be very effective for complete dissolution of metals due to the nature of the source materials. So some pretreatment steps such as surface activation of material [19,22,23], phase transformation prior to leaching are required to enhance the metal dissolution rate. The baking is one of such processes where the phase transformation takes place at the desired temperature and the baked mass gets easily dissolved in the leaching stage. The metal(s) exist in metal oxide, metal sulphide and matte are transformed to corresponding sulfate in the solid phase which can be easily leached out on to the liquid phase. Sulfuric acid baking–leaching process is a widely accepted methodology in hydrometallurgy and based on this process a number of successful technologies have been developed. But reported research investigations in this domain are limited [17,18,20]. In baking process, consumption of acid and time are also minimized and this enables to ease the leach operation and favors the process economy as well.

The studies related to the recovery of metals from the spent Cu–Cr catalyst are limited [15,19]. In a leaching study of Cu–Cr spent catalyst with H₂SO₄ [19] maximum ~56% of copper and <2% of chromium were extracted and to improve the copper leaching efficiency mechanical surface activation of native material was carried out. By using above surface activation method the overall copper recovery was improved up to 80%. For quantitative recovery of chromium after extraction of copper, one additional second stage leaching (alkaline salt roasting leaching) was incorporated.

The proposed baking–leaching process for the extraction of copper from the spent Cu–Cr catalyst is an innovative approach and no work has been reported relating to this investigation. Therefore, the present research work is aimed to describe the selective dissolution of copper from the spent Cu–Cr catalyst in H₂SO₄ medium. The direct leaching followed by the baking–leaching was carried out as the function of key leaching variables. The phase transformation during the baking and leaching of the solid phase was characterized and reported.

2. Experimental

2.1. Materials and reagents

The spent Cu–Cr catalyst used in this investigation was crushed, ground and sieved to obtain different particle sizes. The particle size in the range of 75–105 μm was used in most of the experimental study. The elemental composition of the ground sample is given in Table 1. The chemicals and reagents used for the experimental work were of analytical reagent grade (Merck, India).

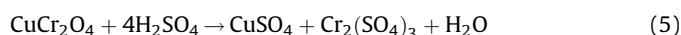
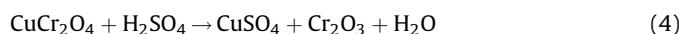
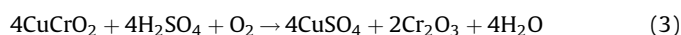
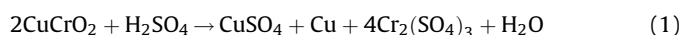
2.2. Baking of the materials

The baking experiments were carried out by thoroughly mixing the fixed amount of definite size fraction (75–105 μm) of the spent catalyst and desired volume of conc. H₂SO₄ in a porcelain crucible. The mixture was then introduced to the pre-heated furnace (at the desired temperature). After the stipulated reaction time the crucible was taken out and kept for cooling inside desiccator. The activated materials were ground and leaching studies were

carried out using predetermined amounts of H₂SO₄ for different time and temperature.

2.3. Leaching method

The leaching experiments were carried out in a three necked 500 ml glass round bottom flask which was placed on a heating mantle. The solution was kept under agitation using an externally connected agitator. The lixiviant was heated with stirring at 600 rpm in most of the studies unless otherwise specified. After attainment of desired temperature, the spent catalyst sample/baked spent catalyst of the appropriate particle size range was charged into the flask containing 250 ml of the H₂SO₄ solution. Periodically samples were collected for analyses. The possible chemical reaction associated during leaching and baking–leaching can be described as given in the following equations ((1)–(5)).



2.4. Analytical method and equipment

The slurry samples of 5 ml were withdrawn at regular time intervals, filtered, diluted and analyzed for metal content by an AAnalyst-200, Perkin-Elmer, (USA) Atomic Absorption Spectrophotometer (AAS). At the end of the leaching tests, the slurry was filtered using filter paper (Whatmann-42) followed by in situ washing of the residues with de-ionized water. The washed residues were dried overnight in an oven at 100 °C, weighed and analyzed for their metal content after digestion. The extent of leaching was calculated on the basis of both solution and solid assays. Some of the leaching tests were repeated in order to ascertain the reproducibility of the experimental results. The typical samples were characterized by XRD and FE-SEM-EDAX study (JOEL JSM-6510 model) analysis. The XRD patterns of the sample were obtained using a Phillips Powder Diffractometer (Model PAN ANALYTICAL PW 1830) in the range of 5–40° (2θ) at a scanning rate of 2°/min with molybdenum target.

3. Results and discussions

The effect of different parameters such as (i) pulp density, (ii) agitation speed, (iii) acid strength, (iv) temperature and (v) particle size range for leaching of copper was investigated where one of the above parameters was changed in the desired range while other parameters were kept constant in most of the leaching studies (unless otherwise specified). The experimental conditions and the results of the maximum leaching of copper and chromium for direct leaching of metals from spent Cu–Cr catalyst was summarized and presented in Table 2.

3.1. Direct sulfuric acid leaching of copper (function of time, agitation speed, H₂SO₄ concentration, particle size, temperature and pulp density)

The extent of copper and chromium dissolution was investigated within the time period of 300 min. Other leaching parameters for the study were: 0.75 M H₂SO₄, particle size 75–105 μm, agitation speed 600 rpm, temperature 35 °C, pulp density

Table 1
Elemental assay of the Cu–Cr spent catalyst.

Element	Cu	Cr	Mn	Ni	Co	Zn
%	42.3	36.2	2.45	0.78	0.085	0.13

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