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# Leaching kinetics of cobalt from the scraps of spent aerospace magnetic materials

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## ABSTRACT

Based on physicochemical properties of the scraps of spent aerospace magnetic materials, a roasting – magnetic separation followed by sulfuric acid leaching process was proposed to extract cobalt. Roasting was performed at 500 °C to remove organic impurity. Non-magnetic impurities were reduced by magnetic separation and then the raw material was sieved into desired particle sizes. Acid leaching was carried out to extract cobalt from the scraps and experimental parameters included agitation speed, particle size, initial concentration of sulfuric acid and temperature. Agitation speed higher than 300 r/min had a relatively small impact on the cobalt extraction. As the particle size reduced, the content of cobalt in the raw material decreases and the extraction of cobalt by acid leaching increased at first and decreased afterwards. Raising the initial concentration of sulfuric acid and temperature contributed to improve the cobalt extraction and the influence of temperature was more remarkable. SEM image revealed that the spent aerospace magnetic materials mainly existed in the sliced strip flake with a loose surface and porous structure. Under the experimental condition, the leaching rate of cobalt from the scraps in sulfuric acid solution could be expressed as  $\ln(-\ln(1 - \alpha)) = nk + nlt$ . The apparent activation energy was found to be 38.33 kJ/mol and it was mainly controlled by the surface chemical reaction.

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## 1. Introduction

Cobalt is a scarce and important strategic resource, widely used in aerospace, military, ship, automobile industries and missile fields because of its good thermal stability, excellent corrosion resistance, high wear resistance and strong magnetic (Xu et al., 2010; Zhang et al., 2012). During the manufacturing of cobalt materials, a large amount of scraps is generated in the cutting and molding process (Onoda and Kurioka, 2015; Xu et al., 2010). If a huge amount of scraps is disposed into landfill directly without any effective treatment, it can result in a serious threat to human health, living and ecological systems due to their hazardous constituents, such as toxic heavy metals (Leysens et al., 2017; Rosskopfová et al., 2011). Recycling of valuable metal from scraps is beneficial for environmental and social-economic sustainability.

### 1.1. Waste cobalt-bearing materials management and strategies

Waste cobalt-bearing materials are produced from hard metal industry, construction industry, e-waste recycling industry, pigment production and paint industry, etc. Most of these materials

have been described as toxic solid waste for the environment and human being (Leysens et al., 2017). For this reason, many methods have been used to estimate the concentration of heavy metals in soil and water (Rachwał et al., 2017; Xu et al., 2015). Rosskopfová et al. (2011) developed an efficient method to determine heavy metal in the low level solid, which is very meaningful for the environmental monitoring. The management of waste toxic metal is a challenging issue that every country has to handle. Many countries have implemented regulations and shared information at different conferences (in Montreal in 2002 and 2007, Sydney in 2003, Paris in 2004) to reduce the quantity of solid waste sending to landfills, by either prevention or redirecting it towards more valuable and environmentally friendly applications (Dhir et al., 2018; Gu et al., 2017). In the past twenty years, China has already established and promulgated a series of regulations and strategies on the recycling of waste electrical and electronic products (Zeng et al., 2017). These regulations and strategies will stimulate industrial plants to recycle of cobalt from waste cobalt-bearing materials.

### 1.2. Recycling of waste cobalt-bearing materials

Recycling of waste cobalt-bearing materials, including spent superalloys (Redden and Greaves, 1992), spent magnetic materials (Wang et al., 2015), spent catalysts (Joo et al., 2016), spent lithium

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ion batteries (Zheng et al., 2017) and hard metal scraps (Lee et al., 2011) has been paid a lot of attention in the last decades. Several processes have been proposed to recycle cobalt from these resources such as physical treatments, pyrometallurgy, hydrometallurgy and electrometallurgy (Jha et al., 2013; Li et al., 2016). Pyrometallurgical process is quite attractive to a large industrial scale due to its high efficiency and productivity. However, pyrometallurgical process increases the energy consumption and environmental contamination (Pagnanelli et al., 2016). Hydrometallurgical process was the main recycling way for the secondary resources because it represents a good selectivity, high recovery rate, low energy consumption and minimal air emission (Joulié et al., 2014; Meng et al., 2018). It is usually involved leaching followed by precipitation, solvent extraction or electrochemical processes (Chen et al., 2015; Freitas et al., 2010). Sulfuric acid is commonly used as leaching agent for cobalt-bearing materials. Ma et al. (2013) studied the cobalt-bearing waste with sulfuric acid and the cobalt leaching efficiency over 99%. Zhu et al. (2012) used sulfuric acid and  $H_2O_2$  as reagents and achieved 96.3% leaching of cobalt from spent lithium ion batteries. Additional studies of recycling cobalt with sulfuric acid can consult related literature (Bertuol et al., 2016). The recovery of cobalt from waste cobalt-bearing materials in sulfuric acid usually achieved satisfying results.

### 1.3. Kinetics of cobalt extraction from secondary resources

Although there are many efforts to the leaching process of cobalt from secondary resources, few detailed kinetic results have been reported. Jha et al. (2013) have comprehensively discussed the leaching behavior of cobalt from waste lithium ion batteries of mobile phone in the media of  $H_2SO_4$  with  $H_2O_2$ . The results revealed that the leaching kinetics of cobalt was fitted well to the model 'ash diffusion control dense constant sizes spherical particles' with an associated activation energy of 59.81 kJ/mol. Meshram et al. (2015b) observed that kinetic data for the dissolution of cobalt from spent Ni-metal hydride batteries showed the best fit to the chemical control shrinking core model and the activation energy was calculated to be 6.8 kJ/mol. Nayl et al. (2017) investigated the reactions of mixed spent Li-ion mobile batteries for cobalt in  $H_2SO_4$  with  $H_2O_2$  media and the control step was confirmed to be surface chemical reaction and the apparent activation energy was 41.4 kJ/mol. Li et al. (2018) showed that leaching cobalt from spent lithium ion batteries in citric acid was also controlled by surface chemical reaction and the apparent activation energy was 86.57 kJ/mol.

The shrinking core model (SCM) of spherical particles was used for describing the leaching data in most of above studies. When this model was unsuccessful in describing the leaching kinetics, empirical model and Avrami equation were applied, particularly for some special shape materials (Shah and Lahti, 1973), such as strip materials, agglomerate materials with different sizes, porous materials, etc. Meshram et al. (2015a, 2015c) studied the kinetics of acid leaching cobalt from waste lithium ion batteries. They found that the leaching data did not follow the SCM, but leaching kinetics followed a logarithmic rate law and noticed that reductant  $NaHSO_3$  could intensify the kinetics of this process with the apparent activation energy changed from 7.4 to 26.8 kJ/mol for cobalt. This logarithmic rate law model was also reported with an activation energy of 7.3 kJ/mol for the recovery of cobalt from spent lithium ion batteries using mild phosphoric acid (Chen et al., 2017). Zhang et al. (2015) researched cobalt leaching process in TCA with  $H_2O_2$  media. They affirmed that the Avrami equation could be well used to describe the leaching process of cathode scraps with a series of different sizes and the apparent activation

energy was determined to be 44.79 kJ/mol. In another research, Avrami equation was also successfully applied to describe the leaching kinetics of cobalt by acidic sodium thiosulphate solution (Li et al., 2011).

So far, almost all of the kinetic studies on cobalt are related to spent lithium-ion batteries, little attention has been paid to the scraps of spent magnetic materials which are poorly understood because of its special shape. Consequently, we intend to explore the leaching kinetics of cobalt from the scraps of spent aerospace magnetic materials. It is a cobalt-base magnetic material in the shape of sliced strip flake, which contains more than 30% cobalt. After a cutting and milling process, it has large oil stains and quartz sands. Based on the physicochemical properties of the spent aerospace magnetic materials, a roasting-magnetic separation followed by sulfuric acid leaching process was proposed to extract cobalt. The influences of various parameters including temperature, particle size, concentration of sulfuric acid on the cobalt leaching were investigated systematically to establish a reasonable kinetic model and determine the corresponding apparent activation energy for cobalt extraction from the scraps.

## 2. Experimental

### 2.1. Materials and pretreatment

Cobalt-based magnetic scraps were collected from a machining workshop of aerospace magnetic materials in China, which mixed with a mass of quartz sands and oil stains. The materials were first placed in a muffle furnace at 500 °C for 2 h to eliminate its surface oil stains and then cooled to room temperature. Magnetic separation was carried out to remove quartz sands and other nonmagnetic impurities. Fig. 1 shows the main pre-treatment procedures for the cobalt-based magnetic scraps. Afterwards, the scraps were sieved to yield a series of particle sizes (+150  $\mu m$ , -150 + 106  $\mu m$ , -106 + 75  $\mu m$ , -75 + 58  $\mu m$ , -58  $\mu m$ ) and the samples with different particle size were completely dissolved in the aqua regia solution ( $HNO_3$ :  $HCl$  = 1:3, v/v) to determine the contents of elements (Co, Ni, Fe) by atomic absorption spectrometry (AAS, TAS-986, Beijing Purkinje General Instrument Co., Ltd.) after required dilution with deionized water. The chemical composition of the spent aerospace magnetic materials is listed in Table 1.

The obtained results in Table 1 indicate that the cobalt content in the scraps vary drastically with different particles. Decreasing the particle size from -150 + 106  $\mu m$  to -75 + 58  $\mu m$ , a significant increase in the content of cobalt from 32.8% to 54.4% is obtained, which may be partly due to the fact of inclusions adhered to the scraps increasing the particle size. In the aqua regia decomposition process of the scrap samples, the solutions of different particle sizes present different colors, as shown in Fig. 2. As the particle size decreased from +150  $\mu m$  to -58  $\mu m$ , the color translates from yellow green into deep green. Different particle sizes resulting in different metal contents were also found in the waste printed circuit boards, which were lower copper content in the smaller particle size because there was a higher difficulty in milling metals than polymers and ceramics (Huang et al., 2014).

The X-ray diffraction (XRD, Rigaku Ru-200) pattern of the scraps using  $Cu K\alpha$  radiation source is shown in Fig. 3. The major phases identified were CoFe,  $Fe_7Ni_3$  and  $AlNi_3$ . Morphology of the spent aerospace magnetic materials was investigated using a scanning electron microscopy (SEM, JSM-7800F, JEOL). Fig. 4 manifests that the scrap sample mainly existed in the sliced strip flake with a loose and porous structure surface.

All chemicals used were of analytical grade reagent and all aqueous solutions were prepared using deionized water.

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