

## Selective leaching of cobalt and iron from cobalt white alloy in sulfuric acid solution with catalyst



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### ABSTRACT

In order to improve the leaching ratios and filtration rate during the dissolution of cobalt white alloy, selective leaching of cobalt and iron from cobalt white alloy under controlling potential in sulfuric solution with catalyst is studied. The effects of factors on the variation of leaching ratios, and potential and residue phase change are studied in detail. The results show that appropriate  $H_2SO_4$  concentration was helpful to improve the separation efficiencies of the metals. The leaching ratios and solution potential increased with the increase of oxidant dosage. When hydrofluoric acid was added as catalyst, the filtration rate would be accelerated with increasing its concentration. Temperature had played an important role in the oxidation capacity of hydrogen peroxide. At the optimal conditions, the leaching ratios of iron, cobalt and copper could reach to 95.00%, 95.00% and 13.50% respectively. Meanwhile, the solution potential was 280 mV and the filtration rate was 9.45 m/h. The results of XRD revealed that the peak intensity of Fe/Co–Si weakened gradually and that of Cu strengthened due to the replacement reaction. In addition, the new phases of  $SiO_2$  and  $Cu_2O$  appeared.

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### 1. Introduction

Cobalt, a steel gray metal with high melting point, possesses many unique properties, and it is mainly used in producing alloy, magnet, pigment and rechargeable battery electrodes (Wang, 2006; Peek et al., 2009). According to the data of U.S. geological survey, the total cobalt productions of the world and China in 2011 are 109,000 t and 45,000 t respectively. Congo is the global leading producer of cobalt ore, nearly 65,400 t of cobalt are exported from Congo and Zambia in 2011. And China is the dominant destination of their cobalt exportation (Shedd, 2011).

Normally, the main raw material exported from Congo is Cu–Co oxide ores. Because of the execution of new policy in Congo, the ores must be further processed before exportation (Ferron, 2008; Shedd, 2011). In view of the level of industrial development, the Cu–Co oxide ores are commonly smelted in an arc furnace and produced in the form of Cu–Co alloy (Yan, 2006; Shedd, 2011; Hu, 2012; Li, 2012), which is also called cobalt white alloy due to the section color (Bai, 2007). So cobalt white alloy has become the dominant raw material instead of ores.

The conventional process of extracting cobalt is designed to treat Cu–Co oxide ores, so the effective dissolution is the key link in the whole process while cobalt white alloy is used as raw material (Bai, 2007; Chong et al., 2013). Many methods have been reported for

dissolving cobalt white alloy, including acid oxidative dissolution, electrochemical dissolution, pretreatment, pressure oxidation leaching and bacterial leaching (Hu et al., 2012), etc. Acid oxidative dissolution is most commonly used to dissolve cobalt white alloy in different acid solutions ( $H_2SO_4$ , HCl or  $HNO_3$ ) by adding oxidants. Compressed air (Cai et al., 1996), sodium chlorate, chlorine (Wang et al., 1997) and hydrogen peroxide can be used as oxidants. It is an effectively way to dissolve cobalt white alloy in hydrochloric acid by adding hydrogen peroxide as oxidant (Xia et al., 2011). However, the leach solution cannot match the demand of the conventional process.

Electrochemical dissolution can be viewed as an economic method. Burzynska et al. studied electrochemical dissolution of Cu–Co alloy in the different system including  $NH_3-(NH_4)_2SO_4$  (Burzynska et al., 2004a),  $NH_3-NH_4Cl$  (Burzynska et al., 2008),  $H_2SO_4$  (Burzynska et al., 2004b; Feng et al., 2014) and  $Na_2SO_4$  (Hong et al., 2010). The metals can be dissolved into the electrolyte or remained into anode slime in different system. The anodic passivation will be improved by adding chlorine ion during electrochemical dissolution in sulfuric acid solution (Kim et al., 2006; Feng et al., 2014).

Due to the erosion-resisting structure of cobalt white alloy, pretreatment method is adopted to destroy its structure before  $H_2SO_4$  leaching. These two pretreatment methods of high temperature smelting with limestone (Peng et al., 2007) and low temperature roasting with NaOH (Xu et al., 2012) can improve the leaching ratios of cobalt in the subsequent  $H_2SO_4$  leaching. Pressure leaching is a clean and efficient method to strengthen leaching. When cobalt white alloy is treated by pressure leaching in sulfuric acid (Ruiz et al., 2007; Wang et al., 2006),

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cobalt and copper are dissolved, while iron is oxidized and precipitated into residue.

It is generally believed that cobalt white alloy is hard to be dissolved (Chong et al., 2013). Xue et al. leach cobalt white alloy in  $H_2SO_4$  solution by adding  $NaClO_3$  as oxidant and  $NH_4HF_2$  as additive (Xue and Zheng, 2011). The leaching ratio of copper and cobalt is above 95.00% and iron is precipitated into the leaching residue. The high leaching ratios and rapid filtration rate can be obtained because  $NH_4HF_2$  can react with silica to form  $(NH_4)_2SiF_6$  deposit. But there are still some problems such as high consumption of ammonium bifluoride and low separation efficiency.

In order to improve the leaching ratios and filtration rate during the dissolution of cobalt white alloy, this present paper focuses on the selective leaching of cobalt and iron from cobalt white alloy under controlled potential in sulfuric solution with adding hydrofluoric acid (HF) as catalyst. The effects of parameters on the variation of leaching ratios, potential and residue phase change are studied in detail. The study could provide a reference to the selective dissolution of similar alloy.

## 2. Experimental

### 2.1. Material

The raw material is cobalt white alloy imported from Congo of Africa. The alloy particle was dried at 110 °C, then ground and sieved, which ensured all of them passing the 200 mesh sieve. The chemical composition of cobalt white alloy is given in Table 1.

All the used reagents of sulfuric acid (98 wt.%), hydrofluoric acid (40 wt.%) and hydrogen peroxide (30 wt.%) are of analytical grade.

The XRD pattern and SEM image of cobalt white alloy are shown in Figs. 1 and 2 respectively.

It can be seen from Fig. 1 that there are two major phases, one is the elemental copper phase (Cu), and another is the complex alloy phase (Fe/Co–Si). The result was consistent with the findings of Bai (Bai, 2007). Fig. 2 shows that the shapes of ground cobalt white alloy are irregular.

### 2.2. Selective leaching under controlling potential

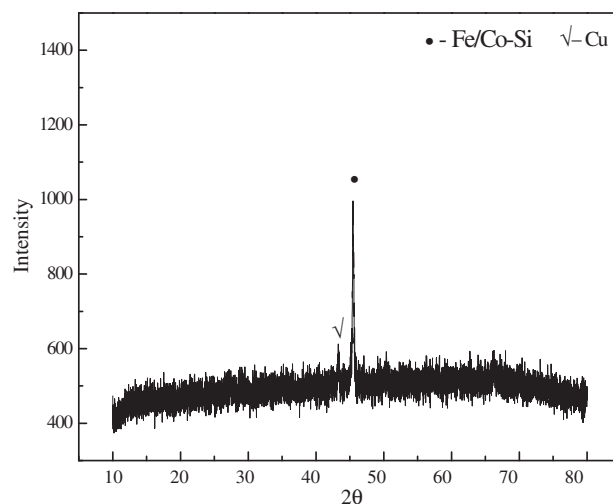
According to the standard electrode potential of metals, the method of selective dissolution under controlling potential in sulfuric acid solution with adding HF is proposed to treat cobalt white alloy. With controlling a certain potential during the selective dissolution, cobalt and iron will be dissolved into the solution, and copper was not oxidized and remained into residue because of its positive electrode potential.

All leaching experiments were conducted in a flask of 500 ml (polytetrafluoroethylene) by equipping a mechanical stirrer and a condenser. Firstly, cobalt white alloy powder of 50 g was added into the sulfuric acid solution containing hydrofluoric acid, and then a special potentiometer was equipped and used to monitor the solution potential. The reaction temperature was controlled by a constant temperature water bath. Secondly, hydrogen peroxide was pumped into the slurry by a constant-flow pump with the velocity of 1.0 ml/min, and the solution potential was controlled by adjusting the volume of  $H_2O_2$ . At last, the resultant slurry was vacuum-filtered immediately and the filtrate volume was measured. The leach residue was washed using hot water and then dried in air oven at 110 °C before weighing.

**Table 1**

Chemical composition of cobalt white alloy.

Components	Co	Cu	Fe	Si	O
wt.%	34.83	16.80	30.11	11.70	6.56



**Fig. 1.** XRD pattern of cobalt white alloy.

The leaching ratios of metals mean the proportion of the dissolved quantity to the total quantity in the raw material, and it can be calculated as Eq. (1).

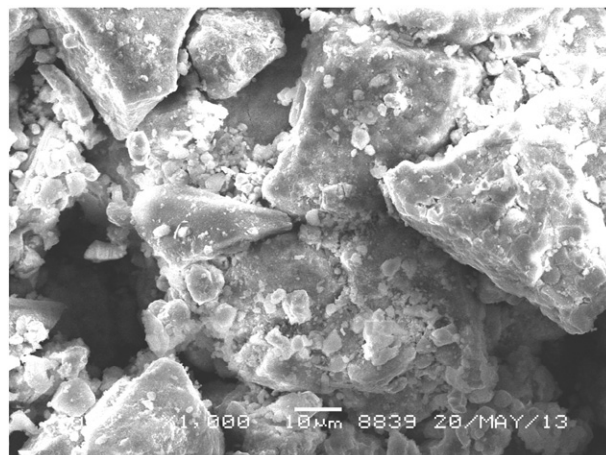
$$R_{Me} = \left( 1 - \frac{m_{residue} \times C_{Me}}{M_{material} \times C_{Me}} \right) \times 100\% \quad (1)$$

Where,  $R_{Me}$  is the leaching ratio (%),  $M_{Me}$  and  $m_{Me}$  are the weights of raw material and residue respectively (g),  $C_{Me}$  and  $c_{Me}$  are the contents of metals in them respectively (wt.%).

### 2.3. Analysis and characterization

The contents of metals in raw material and residue were determined by ICP-AES (IRIS Interprid III XRS). The phases of them were identified by XRD (TTRAX-3, 50KV, 300 mA, 10°/min), and microstructures were characterized by SEM and EDS (JSM-6360LV, 20 kV).

The solution potential was monitored by a special potentiometer in which platinum electrode is used as a working electrode, and calomel electrode acts as a reference electrode, and MT320-SpH instrument (Mettler Toledo Inc.) is used as indicator (Shu and Chen, 1990). The solution potential ( $E_T$ ) is a mixed potential that relates to the potentials from differing constituents, and it is the sum of displayed potential



**Fig. 2.** SEM image of cobalt white alloy.

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