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A simple and effective process for recycling zinc-rich paint residue

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ABSTRACT

Continuous growth of the shipping industry and infrastructure has consumed large amounts of zinc-rich paint (ZP) for the protection of steel structures against corrosion. Consequently, a growing amount of waste zinc-rich paint residue (ZPR) is being generated from anticorrosion spraying. ZPR is classified as hazardous waste in most industrialized countries, but it contains considerable amounts of organic compounds with high calorific value and zinc species that can potentially be recycled. Most of the ZPR generated is not properly treated, and this study presents a simple and efficient process for recycling ZPR. The zinc in ZPR was recovered via a hydrometallurgical route through oxidative alkaline leaching and electrowinning. The results show that the leaching ratio of zinc was greater than 98% at 95 °C, NaOH concentration of 250 g/L, liquid/solid ratio of 10:1, air flow rate of 0.6 L/min, and leaching time of 1.5 h. The appropriate minimum concentration of zinc for electrowinning was determined to be 10 g/L. Adding 50 mg/L of gelatin to the electrolyte significantly refined the grain and the optimum current density was determined to be 200 A/m². Fern shaped cathode zinc powders with a purity of 99.8% were obtained. A high current efficiency (92.7%) was also obtained with energy consumption of 2330.3 kWh per ton of zinc produced. The composition and thermal analysis of the leaching residue suggest that co-processing in cement kiln may be suitable for disposing the leaching residue of ZPR. The experimental results show that the proposed process is promising for ZPR recycling.

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1. Introduction

Zn has been widely used in metal corrosion protection since as early as 1840, when the French chemist Stanislas Sorel developed a hot-dip galvanizing process to protect steel plates. In the early part of last century, zinc-rich paint (ZP) was developed (Finlay and Morley-Smith, 1954). Nowadays ZP is commonly used for corrosion protection of steel structures, such as ships, containers, offshore platforms, bridges, and buildings (Kowalczyk and Szychaj, 2014). In the last two decades, continuous production of structural steelwork for the shipping industry and infrastructure has consumed a great deal of ZP (Xie et al., 2004). Simultaneously, a growing amount of waste zinc paint residue (ZPR) has been generated from anticorrosion spraying. Taking the anticorrosion spraying of container as an example, waste paint spray is generally absorbed by circulating water containing flocculant and is condensed into the paint residue. The production of one thousand TEU (twenty-foot equivalent unit) generally generates 4 tons of ZPR (Zhao and Pan, 2015). China's container production in 2015 was approximately 2.8 million TEU; as the leading

container producer, China accounts for approximately 95% of the total container produced. Hence, a conservative estimate of the annual yield of ZPR in container production alone in China is about 10,000 tons. ZPR is classified as hazardous waste in most industrialized countries and the management of ZPR has become a challenge, especially in China.

Previous studies have focused on the treatment of waste paint residue (PR) generated from the automotive and appliance industries by regeneration, landfill, incineration, pyrolysis, and co-processing in cement kiln (McCarty and Louis, 2011; Avci et al., 2017). However, the literature related to the treatment of ZPR is scarce. Although some research on the regeneration of ZPR was tried, the performance of the generated paint declined, because the entrainment of dirt and impurities, oxidation of zinc, and aging of organic matter were typically unavoidable in the storage and removal of ZPR (Qian and Wu, 2008). In contrast to PR, ZPR contains considerable amounts of zinc (both metallic zinc and zinc oxide) which represents a potential source of zinc if efficient recycling processes can be developed (Salihoglu and Salihoglu, 2016). In addition, the main organic component in ZPR is resin, which has high calorific value. Hence, both the extraction of zinc and the recovery of thermal energy should be studied for the disposal of ZPR.

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Considerable efforts have been devoted to the pyrometallurgical and hydrometallurgical extraction of zinc from zinc-containing wastes, such as zinc leaching residue (Turan, et al., 2004; Behnajady and Moghaddam, 2017; Rüşen and Topçu, 2018), electric arc furnace steel dust (Zhao and Stanforth, 2000; Menad et al., 2003; Havlik et al., 2005; Dutra et al., 2006; Oustadakis, et al., 2010; Chairaksa-Fujimoto et al., 2016), hot galvanizing slag (Dvořák and Jandová, 2005; Ren et al., 2010; Lum, et al., 2014), and spent batteries (De Michelis et al., 2007; Buzatu et al., 2013; Nogueira and Margarido, 2015; Deep et al., 2016; Sadeghi et al., 2017). Hydrometallurgical acid leaching and alkaline leaching are favored because they are economical and environmentally friendly. The leaching mechanism mainly recovers zinc in the form of zinc oxide, which is an amphoteric oxide and can react with both acids and bases. Precipitation, crystallization, and electrowinning are subsequently used to extract zinc from the solution. However, acid leaching is not applicable for ZPR recovery, as a considerable proportion of zinc in ZPR exists in metallic form which would lead to intense reactions with acid, accompanied by the discharge of large amounts of hydrogen gas. The generation of flammable hydrogen is very unfavorable for safe industrial production. In addition, impurities such as calcium and magnesium in the ZPR would also increase acid consumption.

Herein, a novel oxidative alkaline leaching-electrowinning process is proposed to recover zinc from ZPR. The influence of various factors on the leaching and electrowinning were investigated in detail. In addition, thermal analysis of the leaching residue rich in organic substances was performed.

2. Materials and methods

2.1. Materials

The ZPR used in this study was provided by a local spraying factory and had been dried beforehand. The ZPR sample was ground to a particle size of 100 mesh, analyzed by inductively coupled plasma atomic emission spectrometry (ICP-AES), and leached. The chemical composition of the ZPR is listed in Table 1, and it is composed primarily of zinc and carbon with zinc accounting for the largest proportion. The concentrations of metals impurities, such as calcium and magnesium, are also substantial.

The X-ray diffraction (XRD) pattern of the ZPR was measured with a Rigaku Ultima IV diffractometer (Cu, K_{α}) and is shown in Fig. 1. Analysis of the XRD pattern indicates that zinc and zincite (ZnO) were the major phases, with calcite (CaCO_3) and dolomite ($\text{CaMg}(\text{CO}_3)_2$) as minor phases. Fig. 1 also shows that the main zinc-bearing phases are zinc and zincite. It should be noted that amorphous organic compounds in ZPR were not detected by XRD.

2.2. Methods

The leaching batch tests were performed in a flask under mechanical stirring (RW20 digital, IKA, Germany) at 500 rpm in a water bath (DF-101S, Bangxi Instrument Technology Co. Ltd., China). The mass of ZPR used for the leaching batch tests was 40 g and analytically pure grade sodium hydroxide was used as the leaching reagent. The reactor was attached to an air pump

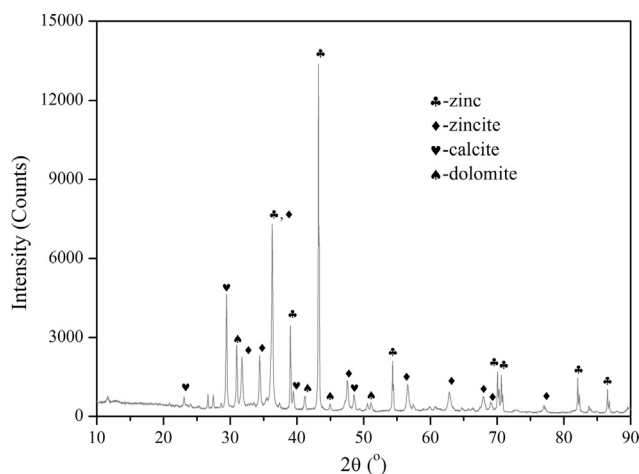


Fig. 1. XRD pattern of the ZPR.

(ACO-5501, HAILEA) equipped with a flowmeter for oxidative alkaline leaching. After the leaching, the slurry was filtered to separate the leaching liquor from the residue and the obtained filter cake was subsequently washed with deionized water.

Zinc electrowinning was conducted at room temperature ($(25 \pm 2)^\circ\text{C}$) in a cubic Plexiglas vessel ($70\text{ mm} \times 70\text{ mm} \times 70\text{ mm}$) with a stainless-steel cathode ($60\text{ mm} \times 60\text{ mm} \times 0.5\text{ mm}$) and two stainless-steel anodes ($60\text{ mm} \times 60\text{ mm} \times 0.5\text{ mm}$). The electrode gap was maintained at 20 mm and the electrodes were connected to a direct current regulated power supply (IT6332A, ITECH). Electrolyte samples were periodically collected to determine their zinc content and to calculate the current efficiency and energy consumption.

The zinc content in the leaching residues, liquors, electrolyte, and cathode were analyzed by ICP-AES.

The metal extraction ratio was calculated as follows:

$$E = (1 - MR/M_0) \times 100\% \quad (1)$$

where E is the metal extraction ratio (%), M_0 and M are the mass fractions of zinc in the ZPR before and after leaching (%), respectively, and R is the ratio of the mass of the ZPR before and after leaching (%).

The current efficiency was calculated as follows:

$$\eta = ((C_0 - C) \times V) / (I \times T \times q) \times 100\% \quad (2)$$

where η is the current efficiency (%), C_0 and C are the zinc concentrations in the solution before and after electrowinning (g/L), respectively, V is the volume of the electrolyte solution (L), I is the current (A), T is the electrowinning time (h), and q is the electrochemical equivalent of zinc (1.22 g/(Ah)).

The energy consumption was calculated as follows:

$$W = U \times 1000 / (q \times \eta) \quad (3)$$

where W is the energy consumption (kWh/t) and the U is the cell voltage (V).

Table 1
Chemical composition of the ZPR.

Component	Zn	Ca	Mg	SiO_2	Ti	Fe	Al	C^a
Content (wt%)	45.82	8.40	1.98	1.77	0.66	0.54	0.31	16.29

Note:

^a Carbon was analyzed using a carbon-sulfur analyzer (EMIA-920V2, HORIBA).

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