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A novel method of preparing highly dispersed spherical lead nanoparticles from solders of waste printed circuit boards

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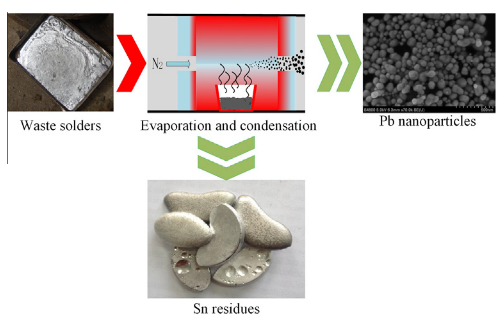
HIGHLIGHTS

- Vacuum separation and dynamic inert gas condensation were applied in this research.
- More than 98 wt% of Pb were separated from waste solders.
- Spherical Pb nanoparticles with high purity were successfully prepared.
- The formation mechanism of Pb nanoparticles was studied.

GRAPHICAL ABSTRACT



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ABSTRACT

In this study, highly dispersed spherical lead (Pb) nanoparticles were prepared from Pb contained solders of waste printed circuit boards (WPCBs) by combining vacuum evaporation and forced flow inert gas condensation. With a relatively high vapor pressure, Pb could be easily separated from waste Pb/Sn solders by vacuum evaporation, while Sn still remained in the residues. Simultaneously, Pb nanoparticles were prepared by the dynamic inert gas condensation. Agglomeration and size inhomogeneity phenomena were eliminated to the maximum extent by sharp quenching using forced flow inert gas and optimizing operation parameters like dynamic nitrogen gas pressure, heating and condensation temperature and condensation distance away from the heater. Highly dispersed spherical Pb nanoparticles were prepared under the optimized conditions of 1223 K heating temperature, 1000 Pa dynamic nitrogen gas pressure, 413 K condensation temperature and 60 cm condensation distance away from the heater. The separation efficiency of Pb from waste solders could reach to higher than 98.2 wt% with the product purity of more than 98 wt% and the size distribution ranging from 20 to 100 nm. This work provides the theoretic foundation for recycling Pb with high added values from waste Pb/Sn solders or other Pb contained solid wastes.

1. Introduction

The increasing volume of waste electrical and electronic equipment (e-waste) is drawing more and more attentions as they are not just sources of contaminants, but also of secondary resources [1]. It is estimated that 20–50 million tons of e-waste are generated around the world and expected to increase by at least 3–5% per annum [2,3]. The typical scrap of these e-waste contains about 40 wt% of different metals which are copper:~20 wt%, iron:~8 wt%, tin:~4 wt%, nickel:~2 wt%, lead:~2 wt%, aluminum:~2 wt%, zinc:~1 wt%, silver:~0.2 wt%, gold:~0.1 wt% and palladium:~0.005 wt% [4]. As for such a high

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content of different metal resources, the e-waste is also called "Urban ore" [5].

Pb is usually found in some components of various kinds of obsolete electrical and electronic equipment. It is reported that the lead contained in printed circuit boards accounts for about 4 wt% by weight. The proportion of lead oxide is more than 20 wt% in waste cathode ray tube, while this proportion reaches to 65 wt% in lead acid batteries [6–8]. When dismantling and recycling e-waste, much attention has been attracted to precious metals like Au, Ag and Pd or relatively expensive metal like Cu and yet Pb is always ignored [9,10]. However, Pb is a kind of toxic heavy metal with great potential hazards to the environment and human beings. When recycling metals from e-waste, Pb either enters into the electrolytic slime during hydro-metallurgy processes or goes into the dust by pyro-metallurgy methods. If treated improperly, these Pb contained pollutants may be emitted into the atmosphere along with dust or enter into the aquatic and terrestrial ecosystems by means of waste solutions and slags. It was reported that the water, soil and atmosphere had been heavily polluted by Pb or other heavy metals in e-waste processing sites like Guiyu and Taizhou in south China [11,12]. It finally leads to the elevation of blood lead levels, threatening the health of local residents [13].

In order to alleviate the environmental pollution risk of Pb and simultaneously recycle the metal resources, many methods including pyrometallurgy [14], hydrometallurgy [15], biometallurgy [16] and supercritical water oxidation [17] processes had been developed in recent years and are still in use nowadays. But the products of these processes are usually the bulk Pb metal with impurities and relatively low economic values.

Vacuum metallurgy separation (VMS) method is considered to be the state of the art method which can produce high purity products as operated under high vacuum conditions. It is an efficient method for both metal purification and nanoparticles preparation when coupled with inert gas condensation. Zhan and Xu [6] used the VMS method to separate Pb from WPCBs and provided the separation criteria of high vapor pressure elements from mixed metallic particles. Xing and Zhang [18] prepared lead agglomerates with a purity of 95 wt% from waste CRT funnel glasses by combining vacuum carbon–thermal reduction and inert-gas consolidation procedures. However, the application of the lead agglomerates may be limited for the agglomeration phenomenon and low purity.

Pb nanoparticles have great potentials to be utilized as lubricating oil additives [19], catalysts [20], doping materials in sensor and wavelength filters [21,22], superconducting materials [23] and X-ray shields [24]. In this work, vacuum evaporation and forced flow inert gas condensation methods were combined together to separate Pb from Pb/Sn solders of WPCBs and simultaneously prepared highly dispersed spherical Pb nanoparticles. The key factors influencing the morphology and size of Pb nanoparticles were studied and optimized. The morphologies of the prepared Pb nanoparticles were characterized and the formation mechanism was studied in detail. This research presents an alternative method for recycling Pb with high added values from waste solders and other Pb contained wastes.

2. Materials and methods

2.1. Principle of Pb nanoparticles preparation

As shown in Fig. 1, there are two main stages involved in the preparation process, namely evaporation and condensation. Firstly, Pb will evaporate and escape into the gaseous phase. After that, Pb vapors will be carried into the relatively cool condensation chamber by the inert quenching gas and transfer their active energies to the cold nitrogen gas via fierce collisions during this migration process [25]. When the Pb vapors become supersaturated in the condensation chamber, homogeneous nuclei will be generated. Then, large numbers of extremely small nuclei will collide with each other to aggregate or coalesce by Brownian motion. This process in the condensation chamber is the growth stage of nanoparticles formation.

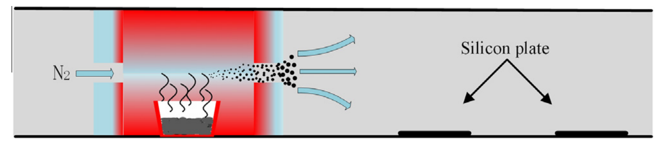

Fig. 1. The schematic illustration of Pb nanoparticles preparation with forced flow inert gas.

2.2. Materials and apparatus

In this study, waste solders obtained from waste printed circuit boards were used as researching objects. The atomic absorption spectrometer analysis indicated that the waste solders were mainly consisted of Pb: 46 wt%, Sn: 52.82 wt%, other: 1.18 wt%. The waste solders were treated by vacuum evaporation and inert gas condensation to prepare Pb nanoparticles.

In order to separate Pb from waste Pb/Sn solders and simultaneously prepare Pb nanoparticles, a self-designed horizontal vacuum quartz tube furnace system was used. The quartz tube in the furnace is 1500 mm long with inner diameter of 95 mm and thickness of 5 mm. The lengths of the heating and condensation chambers are 30 cm and 90 cm respectively. The corundum crucible in the heating chamber is 40 mm in height and 35 mm in diameter. A schematic illustration of the apparatus can be found in our previous work [26]. The furnace was heated by resistance wires and could reach to the maximum temperature of 1273 K. Two plugs (diameter of 95 mm, thickness of 30 mm) made of Al_2O_3 were placed at each side of the heating chamber to avoid heat diffusion to the condensation chamber. There is a hole (diameter of 10 mm) in the middle of each plug so that nitrogen gas could pass through. A few location-adjustable silicon plates were positioned in the condensation chamber to collect Pb nanoparticles. Different dynamic pressures were kept by a nitrogen gas supplier and a mechanical pump working together.

2.3. Experimental procedure

Under all operation conditions, 20 g of waste solders loaded with a corundum crucible were placed in the heating chamber of the furnace. The system was sealed by vacuum flanges and evacuated to 1 Pa by the mechanical pump to avoid oxidation. After that, pure nitrogen gas (N_2, 99.99%) was flowed into the system and kept a dynamic vacuum level of 100, 1000 and 10,000 Pa respectively. When the dynamic nitrogen gas pressure was steady, the furnace was heated to the preset temperature (1123, 1173, 1223 and 1273 K) and maintained for a specific time. As the tube of the furnace was made of quartz, the heating temperature higher than 1273 K might cause damage to the tube and shorten the lifespan of the system. Therefore, the maximum temperature in this study was limited at 1273 K. A flowchart of our whole experimental procedure is displayed in Fig. 2.

2.4. Analysis

After the furnace cooled down, the products collected on the silicon plate were scraped down and preserved in nitrogen atmosphere to avoid oxidation. The morphology of the prepared