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Preparing lead oxide nanoparticles from waste electric and electronic equipment by high temperature oxidation-evaporation and condensation

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ABSTRACT

In this work, two kinds of lead oxides nanoparticles were prepared from Pb-Sn solders of waste electric and electronic equipment by high temperature oxidation-evaporation and condensation using air as oxidizing agent and carrier gas. Lead oxides of hexagonal nanosheets with the thickness of 10–20 nm, length of 150–200 nm and width of 100–150 nm were prepared at 573 K condensation temperature. While, uniform lead oxides of nanorods with dimensions of 10–15 nm in diameter and 50 nm in length were collected at 373 K. The effects of condensation conditions on the morphologies of lead oxides nanoparticles were investigated as well as their growth mechanisms. Further, the as-prepared lead oxides nanosheets and nanorods were applied for constructing Li-ion batteries to test the electro-chemical performance. The galvanostatic cycling tests of Li-ion batteries made by the lead oxides nanosheets showed an initial specific capacity of 917.9 mAh/g and could maintain at 202.2 mAh/g after 100 cycles. The lead oxides nanorods had an initial specific capacity of 1869.6 mAh/g and decreased to 190.2 mAh/g after 100 cycles. This study provides a novel and environmentally friendly way for the resource utilization of Pb from solders and other Pb contained wastes.

1. Introduction

Waste electric and electronic equipments (WEEE) are receiving considerable concerns not only for the enormous amounts and fast growing rate but also for the high content of valuable materials and threatens to our living environment [1–3]. Due to the high contents of metals contained, WEEE is also known as "Urban ore" [4]. However, when exploiting this valuable "Urban ore", most of the recycling processes and techniques are focused on valuable metals like gold, silver, palladium and relatively expensive element like copper [5–7]. The relatively cheap elements like lead, cadmium, chromium are always ignored. Lack of enough attentions and processes for the recovery of Pb from WEEE, serious environmental problems may occur. In fact, the air, soil and water in some WEEE dismantling sites like Guiyu and Taizhou in south China have been seriously contaminated by Pb and other various toxic heavy metals [3,8], which finally resulted in the elevation of blood lead levels of local residents [9]. Therefore, how to recycle Pb or other toxic heavy metals properly will determine the environmental friendliness and integrity of comprehensive recovery of WEEE.

In the past few decades, nanoparticles preparation had drawn great attentions. Until now, it is still a research hotspot for their specific properties and wide applications in nano-devices and functional materials [10–13]. Lead oxide nanoparticles have potential applications in many fields such as lead-acid or Li-ion battery manufacturing [14,15], catalysts [16], pigments and glasses industry [17], dielectric glass-ceramics [18] and so on. A variety of techniques and methods are studied for PbO nanoparticles preparation, and different morphologies are obtained up to now. Zeng prepared hexagonal shape PbO nanosheets by a solution phase synthesis method in the presence of gold nanoparticles [19]. Jia successfully synthesized rod shape PbO single crystal using hydrothermal method [20]. Masood obtained PbO nanocrystals via thermal decomposition of lead oxalate, but the final products seem to be agglomerates without uniform morphologies [21]. To date, the preparation of uniform and highly dispersed lead oxides nanosheets or nanorods by high temperature oxidation-evaporation and condensation method has rarely been reported, especially from waste Pb contained solders.

Pb/Sn alloying solders are important connection materials in most WEEE. These Pb contained waste solders will impose heavy risks on our living environment and health if treated improperly. Herein, we propose a simple but effective procedure to prepare both lead oxides nanorods and nanosheets from waste solders by directly oxidizing and evaporating in a low dynamic atmospheric pressure ambience. Different operation parameters influencing the morphology and size of the final products like condensation temperature and condensation distance

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were studied. The formation mechanisms of lead oxides nanoparticles in the dynamic atmospheric ambience was explored. It is hoped that this work can provide a theoretical foundation for Pb recycling with high added values from waste solders or other Pb contained waste.

2. Materials and methods

2.1. Apparatus

In this study, a self-designed vacuum furnace system was manufactured and used for all experiments as schematically displayed in Fig. 1. The system was consisted of three sections: tube furnace, water cooling jacket and vacuum pump team. The tube furnace had three heating zone which could be respectively heated to different temperatures with the maximum temperature of 1273 K. A quartz tube ($\Phi 10 \times 150$ cm) was placed in the middle of the furnace which connected with an inlet valve and a water cooling jacket through vacuum flanges. A flexible thermo couple was equipped to detect temperatures of different locations in the furnace. Many semicircle stainless steel plates were fixed in the inside of the cooling jacket alternatively with the aim of preventing nanoparticles from entering into the pump oil and polluting the pump team. The crucibles in experiments were provided by Shanghai Shuocun machine electricity hardware limited company. The furnace was designed by our team and manufactured by Shanghai Shengli test instrument limited company.

2.2. Methods

In a typical experiment, 20 g of solders obtained from waste printed circuit boards with the composition of Pb: 46 wt%, Sn: 52.82 wt% and others: 1.18 wt% were loaded with a corundum crucible. Then it was placed in the first heating zone, while the second and third zone were used as the condensation chamber. To avoid the heat diffusion from the first heating zone, two alumina plugs with variant widths were placed at each side of the first zone with a hole (diameter of 1 cm) in the middle of each plug. When the system was sealed with vacuum flanges, the vacuum pump team was started. Simultaneously, the air was pumped into the quartz tube through the inlet valve and adjusted to maintain at a steady dynamic pressure of a desired value. After that, the first zone was heated to the preset temperature and maintained for a desired residence time. When the temperature rose gradually, Pb and Sn in the crucible will be oxidized and the PbO will be evaporated and separated from SnO_2 for lower boiling point. The evaporated PbO vapor above the crucible will be quenched by the cold air which constantly flowed into the furnace. Then these PbO nuclei would be carried into the condensation chamber through the hole in the plug between heating and condensation chambers. A silicon plate was positioned in the condensation chamber to collect the prepared PbO nanoparticles at different distances away from the first zone.

2.3. Analysis

When the furnace cooled down, the collected nanoparticles on the plate were preserved in a dry environment for further analysis. The morphology of the as-prepared products was observed by Scanning Electron Microscope (SEM, S-4800, HITACHI, Japan) and Transmission Electron Microscopy (TEM, HT7700, HITACHI, Japan). The phase was characterized by X-Ray Diffraction (XRD-6100, SHIMADZU, Japan) with Cu Kα radiation, operated at 40 kV and 30 mA at a rate of 2° per minute over an angle of $20° < 2\theta < 80°$.

The as-prepared nanoparticles were electrochemically tested as electrodes in Li-ion batteries. Electrochemical tests were performed by using 2025 type coin cells that were assembled in an Ar-filled glove box (IL-2GB, Innovative Technology). The galvanostatic cycling tests were performed by charging/discharging the assembled cells in the potential range of 0.01–3.0 V at a constant current density of 100 mA/g on a Land CT2001A system. Cyclic voltammetry (CV) measurements were performed on a CHI 760D electrochemical workstation in the potential range of 0.0–3.0 V at a scan rate of 0.1 mV/s.

3. Results and discussion

3.1. Oxidation and evaporation processes of waste Pb-Sn solders

According to pre-experiments, Pb-Sn solders can be oxidized and evaporated at 1223 K heating temperature under 1000 Pa air pressure. Therefore, in this study, all the experiments were processed under this condition.

During the temperature rising period, waste solders in the crucible firstly melt and were gradually oxidized from the surface to the core of the solder melt. As shown in Fig. 2, yellow powder covered the melted waste Pb-Sn solders. Then, when the heating temperature rose to a certain value, lead oxides would evaporate as a result of the high vapor pressure. It was found that a small portion of yellow powder started to evaporate and could be collected in the condensation district once the heating temperature attained to 1223 K.

When the residence time increased to 30 min at 1223 K, the waste solders were all oxidized. More yellow powder was generated and covered on the surface of the Pb-Sn melt as shown in Fig. 2b. Increasing to 60 min, only a small portion of yellow powder remained in the crucible and most of the residues were grey powder as shown in Fig. 2c. Further increasing to 100 min, only grey powder left in the crucible (Fig. 2e). No

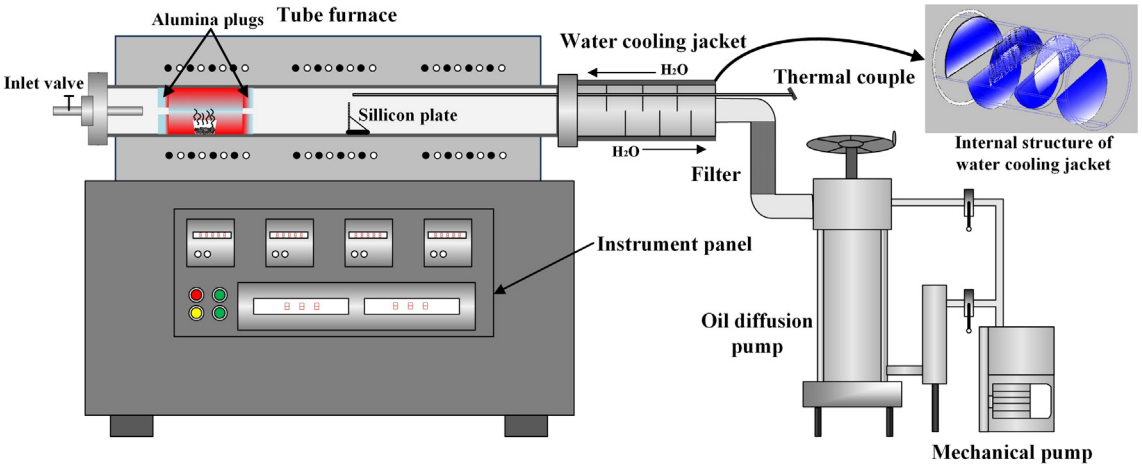

Fig. 1. Schematic representation of the experimental system.