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Preparation of CuO powder for electroplating using lead frame etching wastes

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ABSTRACT

A novel method for manufacture of copper(II) oxide for copper electroplating solution is proposed in this paper. The copper(II) oxide was produced through two chemical reaction steps without sintering process after refinement of waste lead frame etching solution. The experimental major parameters were the amount of additives for first and second step, sodium carbonate and sodium hydroxide, respectively, to evaluate reaction characteristics. Also, the liquidity (angle of repose), solubility to sulfuric acid, chloric ion concentration and thickness of dimple thickness of plating hole were verified for the physical properties of copper(II) oxide as electroplating material.

The reaction molar ratio of sodium carbonate was low, and Cu_2CO_3 was generated more than $Cu(OH)_2$. The optimum reaction mole ratio of sodium carbonate to copper chloride was revealed as 1.5. The optimum usage of sodium hydroxide for manufacture of copper(II) oxide using basic copper carbonate, produced at first reaction step, was 150 g. In these conditions, the average particle size of copper(II) oxide, the dissolution time for sulfuric acid, and the angle of repose were 21.49 μm, 62 s, and 35.5°, respectively. The yield of copper(II) oxide was 98.0 wt.%, for this optimum usage. Also, the via-filling hole thickness was 13.5 μm, which satisfies general via-filling hole thickness range, less than 15 μm.

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Introduction

Lead frame is a copper structure which serves as electric cable as well as supporter of semiconductor chip for the electric circuit. Generally, the lead frame is manufactured by stamping or etching method. For etching method, Na_2ClO_3 is popular etching agent to corrode copper alloy [1,2]. The annual amount of waste lead frame etching solution including copper chloride during etching process is about 600 tons. Moreover the waste contains high concentrations of heavy metals such as iron, nickel, zinc, calcium and magnesium [3,4]. For those reason copper recovery technique from the etching waste is required these days for economic and environmental issue. This paper, therefore, presents a method for manufacturing copper(II) oxide for electroplating by recovering copper. The heavy metal was removed with combination of reduction–oxidation method and ion exchange method as pretreatment process [5,6].

Recently, demands of copper(II) oxide for via-filling electroplating using on high electric capacity circuit board increase with high smartphone penetration rate. The copper(II) oxide reduces surface tension of plating solution so that it ensures seamless control of plating micro patterns and holes. Also it soothes plating thickness distribution. Reaction of acidic copper solution ($CuCl_2$, $CuSO_4$) and salt hydroxide (NaOH, KOH) or basic carbonate salt (Na_2CO_3, K_2CO_3, $(NH_4)_2CO_3$, $NaHCO_3$) yields copper hydroxide ($Cu(OH)_2$) or basic copper carbonate ($CuCO_3 \cdot Cu(OH)_2$).Generally, copper(II) oxide is manufactured by sintering those copper hydroxide or basic copper carbonate at the temperature range of 250–800 °C [7–10]. However, the copper hydroxide, the intermediate, has very low fluidity due to its low solubility to sulfuric acid and small particle size, less than 1 μm. Moreover, the basic copper, intermediate product, has good fluidity with a particle size of 15 μm or more, but has a disadvantage in high sintering temperature of as high as 400–800 °C and copper(I) oxide is produced as a by-product. This paper suggests a method to produce basic copper carbonate as intermediate product as well as to produce copper(II) oxide for electroplating without sintering process. The basic lead carbonate, which is the first step reaction product, was prepared by reacting sodium carbonate (Na_2CO_3)

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with the refined lead frame etching waste solution. The basic copper carbonate was reacted with sodium hydroxide to produce copper(II) oxide. The amount of sodium carbonate at the first step and the amount of sodium hydroxide were controlled as reaction variables to watch characteristics of reaction. Angle of repose and apparent specific gravity were also measured to evaluate copper(II) oxide for electroplating. The solubility to sulfuric acid, chlorine ion concentration, and via-filling hole thickness were evaluated to maintain constant copper concentration.

Experimental

Pretreatment of lead frame etching wastes

The initial heavy metal contents of semiconductor lead frame etching wastes were 2450, 297.3 and 174.3 ppm for Fe^3, Ni^{2+} and Zn^{2+}, respectively. The etching waste solution was refined by combination of reduction–oxidation method and ion exchange method [5]. The reducing agent for reduction–oxidation method was hydrazine. The copper(I) oxide was oxidized and refined with the oxidant of sodium persulfate. The final heavy metal contents of refined lead frame etching waste were 4.3, 2.4 and 0.78 ppm, for Fe^{3+}, Ni^{2+} and Zn^{2+}, respectively [5].

Manufacture of copper(II) oxide by chemical reaction steps

This paper presents the two steps of copper(II) oxide manufacturing processes [11]. The basic copper carbonate ($m\mathrm{CuCO_3}\cdot n\mathrm{Cu(OH)_2}$) was produced by reacting copper chloride (II) in the etching waste and sodium carbonate ($\mathrm{Na_2CO_3}$, 99 wt.%, OCI) in first reaction as shown in Eqs. (1) and (2). The basic copper carbonate, product of the first step, was reacted with sodium hydroxide (NaOH, 99 wt.%, OCI) so that the copper(II) oxide

$$\mathrm{CuCl_2} + \mathrm{Na_2CO_3} \rightarrow \mathrm{CuCO_3} + 2\mathrm{NaCl} \quad (1)$$

$$(m+n)\mathrm{CuCO_3} + n\mathrm{H_2O} \rightarrow m\mathrm{CuCO_3}\cdot n\mathrm{Cu(OH)_2} + n\mathrm{CO_2} \quad (2)$$

$$m\mathrm{CuCO_3}\cdot n\mathrm{Cu(OH)_2} + 2m\mathrm{NaOH} \rightarrow (m+n)\mathrm{CuO} + m\mathrm{Na_2CO_3} + (m+n)\mathrm{H_2O} \quad (3)$$

The reaction ratio of sodium carbonate to copper chloride was maintained at the range from 1.0 to 2.0 at 80 °C of reaction temperature to produce basic copper carbonate, first reaction step product. The copper oxide was produced by reacting 300 g of basic copper carbonate to sodium hydroxide for 1 h at the reaction temperature of 80 °C.

Physical properties of copper(II) oxide

Dissolution time, angle of repose particle size and distribution and apparent specific gravity were evaluated to check basic copper carbonate and copper(II) oxide. The angles of repose were measured with a measuring set, model BT-200D, product of K-ONE. Also, the particle size and distribution were measured with a particle size analyzer, Bettersize model BT-2000, product of K-ONE. The apparent specific gravities for each experimental conditions were measured with a gravimeter, product of Kuramochi scientific. The morphology of the particle was observed using a scanning electron microscope (SEM, COXEM, CX-20).

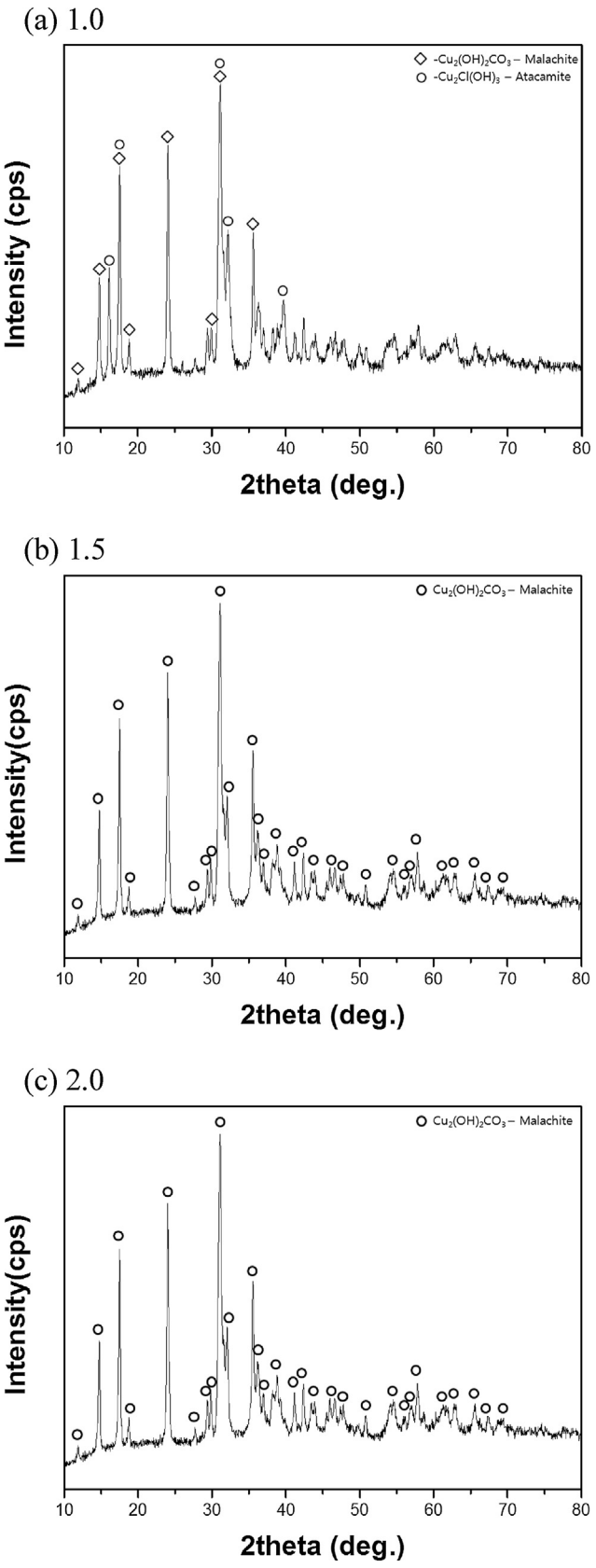

Fig. 1. XRD patterns of basic copper carbonate with $\mathrm{Na_2CO_3/CuCl_2}$ mole ratio.

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