

Electrochemically deposited p–n homojunction cuprous oxide solar cells

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ABSTRACT

The electrical properties of both p- and n-type cuprous oxide (Cu_2O) films electrochemically deposited from two electrolyte solutions were examined by current–voltage measurements. The resistivity of p-type Cu_2O varied from 3.2×10^5 to $2.0 \times 10^8 \Omega\text{cm}$, while that of n-type Cu_2O from 2.5×10^7 to $8.0 \times 10^8 \Omega\text{cm}$, depending on deposition conditions such as solution pH, deposition potential and temperature. With optimized deposition conditions for minimum resistivity, p–n homojunction Cu_2O solar cells were fabricated by a two-step deposition process. The p–n homojunction Cu_2O solar cells showed a conversion efficiency of 0.1% under AM1 illumination. The low efficiency is attributed to the high resistivity of p- and n-type Cu_2O , which require doping to reduce.

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1. Introduction

The high cost of silicon solar cells forces the development of new photovoltaic devices utilizing cheap and non-toxic materials prepared by energy-efficient processes [1]. Cuprous oxide (Cu_2O) has attracted attention for solar cell applications as the p-type active layer due to its direct band gap around 2 eV and high absorption coefficient, combined with material abundance, non-toxicity and low-cost fabrication [2,3]. However, our poor understanding about intrinsic point defects, difficulty in doping and absence of n-type Cu_2O have impeded the development of Cu_2O -based solar cells. Over the past three decades, Cu_2O -based solar cells have been fabricated with metal/ Cu_2O Schottky junctions [2] and p–n heterojunctions such as n-CdO/p- Cu_2O [4], n-ZnO/p- Cu_2O [3,5] and n-ITO/p- Cu_2O [6]. The highest conversion efficiency with Cu_2O as the active layer is around 2% with a p–n heterojunction structure [6], while the theoretical efficiency for Cu_2O solar cells is about 19% [7]. It is generally accepted that a p–n homojunction of Cu_2O is the best way to increase the efficiency of Cu_2O -based solar cells [8]. Our previous study [9] found that solution pH can control the conduction type of electrochemically deposited Cu_2O . Cu_2O electrochemically deposited at solution pH below 8.0 in an aqueous solution containing 0.4 M copper sulfate and 3 M lactic acid was n-type, while Cu_2O deposited at solution pH above 9.0 was p-type. n-Type behavior of Cu_2O electrochemically deposited at low solution pH in a different electrolyte solution was also reported by others [10]. With both p- and n-type Cu_2O , a p–n

homojunction of Cu_2O was demonstrated by a two-step deposition process [9]. However, the small forward current (~ 0.1 mA/cm² at 2 V) in the p–n junction indicated highly resistive p-type, n-type or both-type Cu_2O . Hence, it is important to optimize the deposition conditions for both p- and n-type Cu_2O to achieve best efficiency in Cu_2O p–n homojunction solar cells.

In this study, we investigated the dependence of resistivity of p- and n-type Cu_2O on several deposition conditions such as solution pH, deposition potential and temperature. With the optimized deposition conditions, p–n homojunction Cu_2O solar cells were fabricated by an inexpensive two-step electrochemical deposition process.

2. Experimental

Two different electrolyte solutions were used for p- and n-type Cu_2O electrochemical deposition. For p-type Cu_2O , the aqueous solution contained 0.4 M copper sulfate and either 1.8 or 3 M sodium lactate. The solution for n-type Cu_2O contained 0.01 M copper acetate and 0.1 M sodium acetate. The deposition of p- and n-type Cu_2O was potentiostatically carried out using a Princeton Applied Research Versastat II potentiostat without stirring. A traditional three-electrode electrochemical cell was used, where the counter electrode was a Pt foil, the reference electrode was Ag/AgCl/saturated NaCl and the working electrode was a 200-nm Cu film thermally evaporated on glass substrate. Prior to deposition, the Cu/glass substrate was sonicated in acetone, etched in 1 M nitric acid for a minute and rinsed in deionized water.

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In order to examine the dependence of resistivity of p-type Cu_2O on deposition conditions, the solution pH was varied from 9.0 to 13.0 by adding different amounts of NaOH. The deposition potential was controlled from -0.3 to -0.6 V vs. the Ag/AgCl/saturated NaCl reference electrode. The deposition temperature was adjusted from 40 to 80 °C. In case of n-type Cu_2O , the solution pH was controlled from 5.2 to 6.4 with different amounts of acetic acid. The deposition potential was fixed at -0.1 V and the temperature was 60 °C.

Four-point probe and Hall measurements are the standard and reliable methods to characterize electrical properties of thin semiconductor films. However, it is difficult to characterize the electrical properties of electrochemically deposited semiconductor films with these methods, because of the highly conductive substrate required for electrochemical deposition. Even though a two-layer model is available for Hall measurements, it is difficult to characterize highly resistive semiconductor films due to the small Hall voltage [11]. In this study we adopted a simple current–voltage (I – V) method to characterize the electrical properties of both p- and n-type Cu_2O . Circular Cu electrodes with an area of $2.2 \times 10^{-3} \text{ cm}^2$ were deposited on the top surface of both p- and n-type Cu_2O by thermal evaporation. A voltage was applied between substrate and top electrode, and the current was measured with an Agilent 4155C semiconductor parameter analyzer at room temperature. From the slope of the I – V relation and the thickness of the Cu_2O film, the resistivity was determined. An Alpha Step IQ surface profiler was used to measure the thickness of Cu_2O films.

For solar cell fabrication, glass substrates were cut to approximately $1.2 \times 3.7 \text{ cm}^2$ and were coated with 75-nm Cr using thermal evaporation for good adhesion with Au. 100-nm Au was then thermally evaporated on the Cr/glass substrate. To make a p–n homojunction, p- and n-type Cu_2O films were sequentially deposited in two different electrolyte solutions. Photovoltaic devices were completed by RF sputtering of 100-nm ITO on n-type Cu_2O , followed by DC sputtering of Al top contact. The finished devices were $1 \times 1 \text{ mm}^2$ in size. The performance of the devices was obtained with a home-made solar simulator. The light source was a 300-W GE lamp calibrated at AM1 by matching the short-circuit current of a standard silicon solar cell. The I – V data was obtained with a Keithley 2420 SourceMeter.

3. Results and discussion

3.1. Resistivity of Cu_2O

Before resistivity measurements, photocurrent characterization was performed on all the samples at 0 V vs. Ag/AgCl/saturated NaCl to verify their conduction type. All the samples deposited in an aqueous solution containing 0.4 M copper sulfate and 3 or 1.8 M sodium lactate at solution pH above 9.0 showed cathodic photocurrents, representing p-type conductivity. Samples deposited in an aqueous solution containing 0.01 M copper acetate and 0.1 M sodium acetate at solution pH below 6.4 exhibited anodic photocurrents, indicating n-type conductivity.

When I – V measurements were performed to determine the resistivity of both p- and n-type Cu_2O , none displayed a linear I – V behavior due to the Schottky barrier between Cu and Cu_2O [8]. The resistivity of Cu_2O was thus determined by linearization of the forward current between 1.7 and 2 V and showed similar order of magnitude to reported values by a different method [12], indicating the validity of the measurement. These resistivities were enough to evaluate the dependence of resistivity of electrochemically deposited Cu_2O on different deposition conditions.

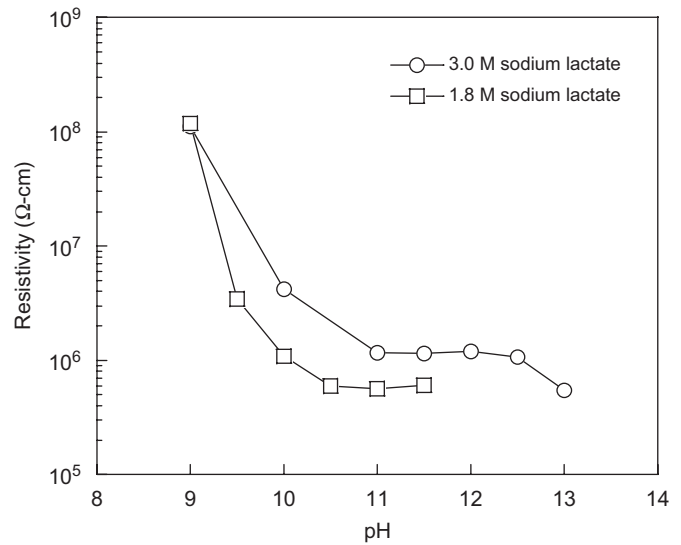


Fig. 1. Resistivity of p-type Cu_2O as a function of solution pH. The deposition was performed at -0.4 V and 60 °C with different sodium lactate concentrations.

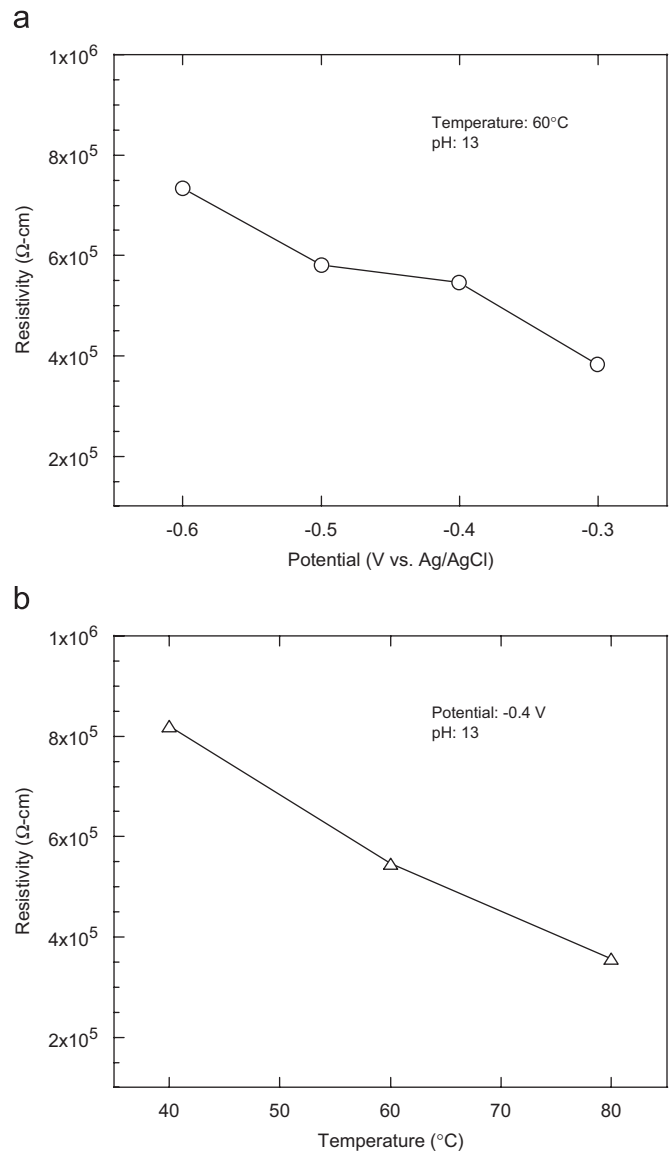


Fig. 2. Resistivity of p-type Cu_2O as a function of (a) deposition potential and (b) deposition temperature.

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