Effect of Annealing Atmosphere on Interface State Density in p-CuO/n-Si Heterojunction Devices: A Capacitance–Voltage Study

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ABSTRACT

Thin film p-CuO/n-Si heterojunction devices were fabricated by depositing CuO thin films on n-type silicon substrates using the Spray Pyrolysis Deposition (SPD) technique, followed by post-annealing at 450 °C under air and nitrogen atmospheres. The interface properties of the heterojunctions were investigated using frequency-dependent capacitance-voltage (C–V) measurements carried out over the frequency range of 100 Hz to 1 MHz. The interface state density was estimated from the difference between the quasi-low-frequency and high-frequency capacitances measured at 10 kHz and 100 kHz, respectively. The results indicate that the interface states strongly influence the electrical characteristics of the CuO/Si junction and that the annealing atmosphere plays a significant role in controlling the density of interface states. The study demonstrates that C–V characterization is an effective tool for evaluating the interfacial properties of p-CuO/n-Si heterojunctions and provides valuable insight into the defect states present at the CuO/Si interface.

KEYWORDS

CuO thin films; p-CuO/n-Si heterojunction; Interface states; C–V characteristics; Annealing effects

INTRODUCTION

Copper oxide (CuO) is a promising p-type semiconductor material for photovoltaic and optoelectronic applications owing to its narrow band gap, high absorption coefficient, low cost, and non-toxic nature. In particular, p-CuO/n-Si heterojunction solar cells have attracted considerable interest because of their simple device architecture and compatibility with silicon technology. However, the photovoltaic performance of these heterojunction devices is often limited by the presence of interface states at the CuO/Si junction, which act as trapping and recombination centres for charge carriers and thereby reduce the photocurrent and overall device efficiency [1, 2].

The quality of the interface between the CuO thin film and the n-type silicon substrate plays a crucial role in determining the electrical and photovoltaic characteristics of the heterojunction. Interface states may originate from thin-film growth processes, oxygen vacancies in CuO, lattice mismatch, and post-deposition thermal treatments. These defect states influence carrier transport across the junction and affect important device parameters such as series resistance, open-circuit voltage, fill factor, and conversion efficiency in case of a solar cell. Therefore, the evaluation of interface state density is essential for understanding the recombination mechanisms and improving the performance of CuO/Si heterojunction devices [3].

Capacitance-voltage (C–V) characterization is a widely employed, non-destructive technique for investigating the electrical properties of semiconductor junctions and estimating the density of interface states [4-6]. The frequency dependence of the capacitance response provides valuable information regarding the ability of interface states to exchange charge with the semiconductor. At low frequencies, interface states are able to follow the applied alternating signal and contribute to the measured capacitance, whereas at high frequencies their response is suppressed and the measured capacitance primarily corresponds to the depletion capacitance of the junction. Consequently, the interface state density can be estimated from the difference between the quasi-low-frequency and high-frequency capacitances [7].

In the present work, p-CuO/n-Si heterojunction devices were fabricated by spray pyrolysis deposition of CuO thin films on n-type silicon substrates, followed by annealing under air and nitrogen atmospheres. Device fabrication was completed by depositing 120 nm thick Ag front contacts and a 120 nm thick Al back contact using RF sputtering. Frequency-dependent C–V measurements were carried out using an impedance analyser over the frequency range from 100 Hz to 1 MHz and within a bias range of −5 V to +5 V. The interface state density was evaluated from capacitance measurements obtained at 10 kHz and 100 kHz, and the influence of annealing atmosphere on the interface characteristics of the p-CuO/n-Si heterojunction was investigated.

MATERIALS AND EXPERIMENTAL METHODS

Copper oxide (CuO) thin films were deposited using the spray pyrolysis deposition (SPD) technique. A 0.1 M precursor solution was prepared by dissolving the required amount of copper acetate monohydrate, [$(CH_3COO)_2Cu\cdot H_2O$, Sigma-Aldrich], in double-distilled water. The solution was magnetically stirred at room temperature for approximately 10 min to obtain a homogeneous precursor solution.

N-type Si substrates of 1cmx1cm dimensions were used for the fabrication of p-CuO/n-Si heterojunction devices. Prior to deposition, the substrates were cleaned using the standard RCA-1 and RCA-2 cleaning procedures to remove organic and metallic contaminants. The CuO films were deposited on the cleaned Si substrates maintained at a temperature of 350 °C. Compressed air was employed as the carrier gas with a pressure of 4.2 kg cm^{-2}, and the distance between the spray nozzle and the substrate was fixed at 30 cm. The deposition was carried out for 16 min. To maintain the substrate temperature and ensure uniform film growth, the deposition was performed intermittently using a spray cycle of 5 s followed by a pause interval of 60 s.

The as-deposited CuO films were subjected to post-deposition annealing at 450 °C under air and nitrogen atmospheres in order to investigate the influence of annealing environment on the interface characteristics of the heterojunction. Subsequently, p-CuO/n-Si heterojunction devices were fabricated by depositing metallic contacts using radio-frequency (RF) sputtering. Four circular Ag contacts of diameter 2 mm and thickness 120 nm were deposited on the CuO-coated surface to serve as the front electrodes. A 120 nm thick Al layer was deposited over the entire backside of the Si substrate to establish an ohmic contact. The resulting device structure consisted of Ag/p-CuO/n-Si/Al.

INTERFACE STATE DENSITY ESTIMATION BY CAPACITANCE–VOLTAGE MEASUREMENTS

In p-CuO/n-Si heterojunction devices, the quality of the interface between the CuO thin film and the n-type silicon substrate plays a crucial role in determining device performance. Defects originating from thin-film growth processes, oxygen vacancies in Cuo, and post-annealing treatments lead to the formation of interface states at the CuO/Si junction. These interface states act as trapping and recombination centres for charge carriers, thereby influencing charge transport across the junction and affecting important device parameters [8].

At quasi-low frequencies, interface states are able to respond to the applied AC signal and contribute to the measured capacitance, whereas at higher frequencies their response is suppressed and the measured capacitance primarily represents the depletion capacitance of the junction. Accordingly, the interface state density (N_{is}) in the depletion region was calculated using the relation [9],

$$N_{is} = \frac{C_{LF} - C_{HF}}{qA} \qquad (1)$$

where C_{LF} and C_{HF} are the quasi-low-frequency and high-frequency capacitances, respectively, q is the electronic charge, and A is the effective area (= 3.14×10^{-2} cm^2) of the Ag dot contact.

RESULT AND ANALYSIS

Spray-deposited CuO thin films grown on n-type silicon substrates and subsequently annealed under air and nitrogen atmospheres were used for the present study. Frequency-dependent capacitance-voltage (C–V) measurements were performed using an impedance analyser over the frequency range of 100 Hz to 1 MHz to investigate the interfacial properties and estimate the interface state density of the p-CuO/n-Si heterojunctions. Figure 1 shows the capacitance–voltage (C–V) characteristics of the air annealed (C16-A450) and nitrogen annealed (C16-N450) p-CuO/n-Si heterojunctions measured at frequencies ranging from 100 Hz to 1 MHz.

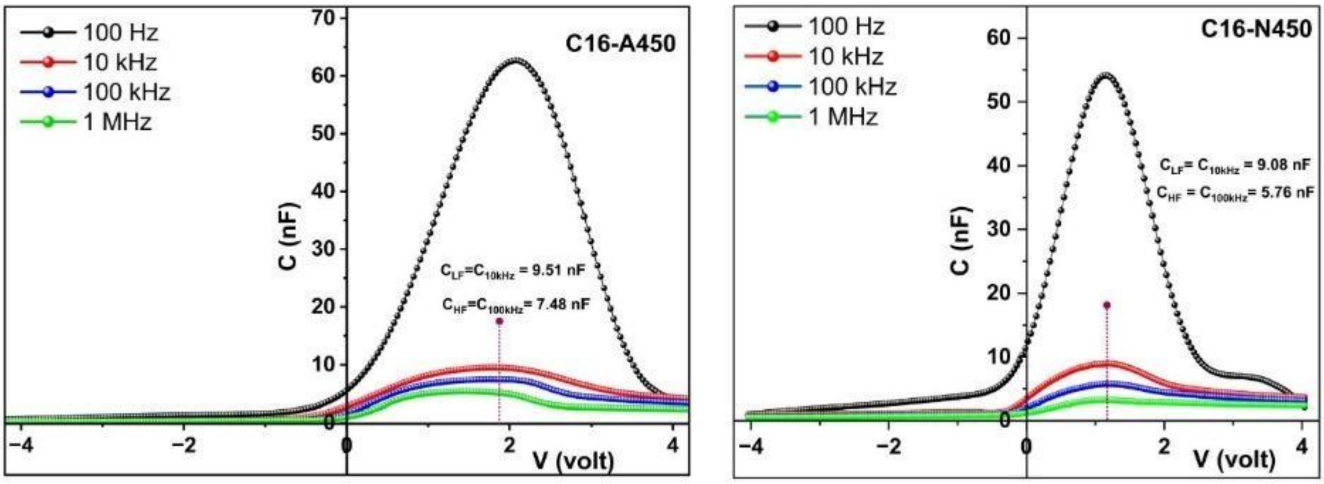

Figure 1. Capacitance-voltage characteristics of p-CuO/n-Si heterojunction devices measured at 100 Hz, 10 kHz, 100 kHz, and 1 MHz.

Table 1. Interface state density values of air- and nitrogen-annealed p-CuO/n-Si heterojunction devices obtained from C–V measurements.

Sample	C_{LF} nF	C_{HF} nF	A cm^2	$N_{is} = \frac{C_{LF} - C_{HF}}{qA}$ cm^{-2}
C16-A450	9.51	7.48	3.14×10^{-2}	4.04×10^{11}
C16-N450	9.08	5.76	3.14×10^{-2}	6.61×10^{11}

Table 1 presents the quasi-low-frequency capacitance (C_{LF}), high-frequency capacitance (C_{HF}), and the corresponding interface state density (N_{is}) values of the air-annealed (C16-A450) and nitrogen-annealed (C16-N450) p-CuO/n-Si heterojunction devices. The interface state density was calculated using Eq. (1).

For the air-annealed sample (C16-A450), the quasi-low-frequency and high-frequency capacitances were found to be 9.51 nF and 7.48 nF, respectively, yielding an interface state density of 4.04×10^{11} cm^{-2}. In contrast, the nitrogen-annealed sample (C16-N450) exhibited capacitance values of 9.08 nF and 5.76 nF, resulting in a higher interface state density of 6.61×10^{11} cm^{-2}.

The larger difference between the low-frequency and high-frequency capacitances observed for the nitrogen-annealed sample indicates a greater contribution from interface states [10]. At low frequencies, these states are able to exchange charge with the semiconductor and contribute to the measured capacitance, whereas their response is suppressed at high frequencies. Consequently, a larger difference between C_{LF} and C_{HF} corresponds to a higher density of interface states [11].

The lower interface state density obtained for the air-annealed sample suggests that annealing in air improves the quality of the CuO/Si interface by reducing electrically active defects and recombination centres [1, 12]. On the other hand, the higher interface state density observed in the nitrogen-annealed sample may be attributed to the formation of oxygen vacancies and defect states under oxygen-deficient conditions. These interface states act as carrier trapping and recombination centres, thereby impeding charge transport across the junction and contributing to the relatively poor photovoltaic performance of the heterojunction devices. Therefore, the results demonstrate that the annealing atmosphere plays a crucial role in controlling the interface quality and defect density in p-CuO/n-Si heterojunctions [12, 13].

CONCLUSION

In this work, the interface state density of spray-deposited p-CuO/n-Si heterojunction devices annealed under air and nitrogen atmospheres was investigated using frequency-dependent capacitance-voltage measurements. The interface state density was estimated from the difference between the quasi-low-frequency and high-frequency capacitances. The air-annealed sample exhibited a lower interface state density of 4.04×10^{11} cm^{-2} compared to 6.61×10^{11} cm^{-2} for the nitrogen-annealed sample, indicating superior interface quality. The higher interface state density observed in the nitrogen-annealed device is attributed to the increased concentration of electrically active defects and oxygen vacancy-related states, which act as carrier trapping and recombination centres. The results demonstrate that the annealing atmosphere significantly influences the interfacial properties of p-CuO/n-Si heterojunctions and that capacitance-voltage characterization provides an effective means for evaluating interface states. The lower interface state density obtained for the air-annealed device suggests that annealing in air improves the CuO/Si interface quality and is more favourable for enhancing the electrical and photovoltaic performance of CuO-based heterojunction devices.

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