

Investigation of the Electrical Properties of Cu-doped CoOx/n-Si Structures Fabricated by the Sol-Gel Method

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Sol-jel yöntemiyle üretilen Cu katkılı CoOx/n-Si yapıların elektriksel özelliklerinin araştırılması

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Abstract

The sol-gel spin coating technique was employed for the deposition of thin films comprising CoOx, Cu-doped CoOx, and CuOx onto n-Si substrates. Subsequently, an exhaustive examination of the electrical properties of the resultant heterojunction structures was conducted. The outcomes unequivocally indicate that the incorporation of Cu through doping exerts a pronounced influence on the electrical attributes of the CoOx/n-Si diode. Notably, all diodes exhibit rectifying behavior, a discernible feature in their dark current-voltage (I-V) characteristics. The I-V data was further utilized to ascertain pivotal junction parameters, encompassing series resistance (Rs), rectification ratio (RR), ideality factor (n), and barrier height (ΦB). The values of the ideality factor for CoOx/n-Si, Cu doped CoOx/n-Si and CuOx/n-Si are obtained to be 3.19, 1.99 and 2.19 eV, respectively. Furthermore, the capacitance-voltage (C-V) characteristics of diodes were performed within the frequency range of 10 kHz to 1 MHz. These findings underscore that judicious manipulation of the copper doping concentration can serve as an effective means to modulate the electrical properties of CoOx/n-Si diodes.

Anahtar Kelimeler: Cu doped Cobalt Oxide; Heterojunction structures; Sol gel method; Current-Voltage.

Öz

CoOx, Cu katkılı CoOx ve CuOx içeren ince filmlerin n-Si substratlar üzerine biriktirilmesi için sol-jel döndürerek kaplama tekniği kullanıldı. Daha sonra, elde edilen heteroeklem yapılarının elektriksel özelliklerinin kapsamlı bir incelemesi yapıldı. Sonuçlar açıkça Cu'nun katkılama yoluyla dahil edilmesinin CoOx/n-Si diyotun elektriksel özellikleri üzerinde belirgin bir etki yarattığını göstermektedir. Özellikle, tüm diyotlar, karanlık akım-voltaj (I-V) özelliklerinde fark edilebilir bir özellik olan doğrultucu davranış sergiledi. I-V verileri diyotların seri direnci (Rs), düzeltme oranını (RR), idealite faktörünü (n) ve bariyer yüksekliğini (ΦB) kapsayan önemli bağlantı parametrelerini belirlemek için kullanıldı. CoOx/n-Si, Cu katkılı CoOx/n-Si ve CuOx/n-Si için idealite faktörü değerleri sırasıyla 3,19, 1,99 ve 2,19 eV olarak elde edilmiştir. Ayrıca diyotların kapasitans-gerilim (C-V) özelliklerinin ölçümleri 10 kHz ila 1 MHz frekans aralığında gerçekleştirildi. Bu bulgular, bakır doping konsantrasyonunun uygun şekilde seçilmesinin, CoOx/n-Si diyotların elektriksel özelliklerini iyileştirmek için etkili bir yol olduğunu göstermektedir.

Keywords: Cu katkılı Kobalt Oksit; Heteroeklem yapılar; Sol jel metodu, Akım-Voltaj.

1. Introduction

Semiconductor metal oxide thin films have appeared as a highly sophisticated technology for optoelectronic devices based on p-n junctions (Dhanabalan et al. 2017; Tawfik et al. 2019; Rakotonarivo et al. 2021). In recent times, there has been considerable focus on copper oxide nanostructures owing to their remarkable characteristics, including a tunable bandgap, chemical stability, and customizable electrical properties (Bayansal et al. 2014; Asl and Rozati 2020). Over the past few decades, copper oxide (CuOx) has found extensive application in various electronic and optoelectronic devices. These include gas sensors (Nam et al. 2006), smart windows (Lei et al. 2021), storage energy devices (Qiu et al. 2015), super capacitors (Liu et al. 2017), and photovoltaic devices (Shalan et al. 2016).

Cobalt oxide offers several key benefits, such as its cost-effectiveness in production, thermal stability, non-toxic nature, and substantial optical absorption (Maduraiveeran et al. 2019). A thorough investigation into the structural, electrical, and optical characteristics of CoOx thin films has shown a strong relationship with the techniques used to prepare the films. The CoOx thin films are a popular choice among researchers due to the wide variety of conventional deposition methods that are available for them. The following deposition methods have been generally employed for CoOx thin films: sputtering (Estrada et al. 1993), electrodeposition (Casella 2002), thermal oxidation (Salavati-Niasari et al. 2009), spray pyrolysis (Shinde et al. 2006), and spin coating (Valanarasu et al. 2014). One of the deposition techniques used for CoOx thin films is the sol-gel spin coating

technique. It has multiple benefits for thin film deposition, such as producing films with precise control over thickness and uniform deposit, which makes it appropriate for a range of applications. This method works at relatively low temperatures and is suitable for a variety of substrates, including materials that are sensitive to heat. Moreover, sol-gel spin coating requires less complex equipment, is less expensive, and is scalable for large-scale production. Taking everything into account, this method provides researchers with a versatile, reasonably priced, and controllable means of depositing thin films that exhibit remarkable compatibility and homogeneity (Kamaruddin et al. 2011). Moreover, the physical characteristics of the films are greatly affected by the doping process, which incorporates different chemical constituents. By precisely controlling the doping level, it becomes possible to obtain films with enhanced properties that are highly relevant for diverse applications. Consequently, there exists a substantial interest in investigating the relationship between the physical and electrical properties of the films and the concentration of the doping elements. Extensive research has been conducted to enhance the performance of CoOx thin films, leading to investigations into the incorporation of various transition metals, including N, Mn, and Cu, as promoter elements (Zhang et al. 2012; Li et al. 2018; Ke et al. 2020). These studies aim to develop semiconductors with improved properties, thereby advancing the capabilities and functionalities of thin film-based devices. Among these metals, the copper doping is expected to improve the electrical, catalytic activity, optical, magnetical and stability properties of CoOx by modifying its electronic structure (Zhang et al. 2012; Berenguer et al. 2017; Behzad et al. 2018). This study entailed the utilization of a cost-effective sol-gel methodology for coating undoped and Cu-doped CoOx thin films onto n-Si substrates, with the objective of fabricating heterojunction structures. Subsequent to the

film deposition, I-V and C-V measurements were conducted on the resulting diodes under dark conditions and at ambient temperature. The electrical characteristics of the diodes were examined with respect to the impact of Cu doping. Essential electrical parameters of the diodes were evaluated using established methodologies, including the Thermionic Emission Theory, the Norde method, and C-V measurements.

2. Materials and Methods

In this study, copper acetate monohydrate and cobalt acetate tetrahydrate were preferred as precursor materials for $Cu_xCo_{1-x}O$ ($x=0, 0.25$ and 1) thin film solution, respectively. 2-Methoxyethanol was used as the solvent and monoethanolamine (MEA) was used as the stabilizer. For the starting solution, 0.1M copper acetate and cobalt acetate were dissolved in 2-Methoxyethanol and sufficient amount of MEA was added. Copper acetate monohydrate and cobalt acetate tetrahydrate solutions were prepared. For the copper-doped cobalt oxide film, the Cu/Co solution ratio by volume was mixed at 1/3. The Cu-doped CoOx thin film was coded as $Cu_{0.25}Co_{0.75}O$. It was mixed with a magnetic stirrer at 2000 rpm for 120 min at 60 °C. All thin films were coated onto n-type Si substrates utilizing the sol-gel spin coating technique. These commercially available substrates are polished on one side, have (111) crystal orientation, phosphorus doping, resistivity of 1-20 Ωcm , and donor concentration of 10^{15}cm^{-3} . After each Coating, it was dried for 10 minutes in a preheated 300°C oven to remove organic residues and let the solvent evaporate. The coating and drying process was repeated 5 times. Then, the prepared thin films were annealed in a tube furnace in air at 450°C for 1 hour. Finally, the top contacts of the $Cu_xCo_{1-x}O/n\text{-Si}$ structures were formed by evaporating Ag metal under a vacuum of 4×10^{-5} torr by using a shadow mask.

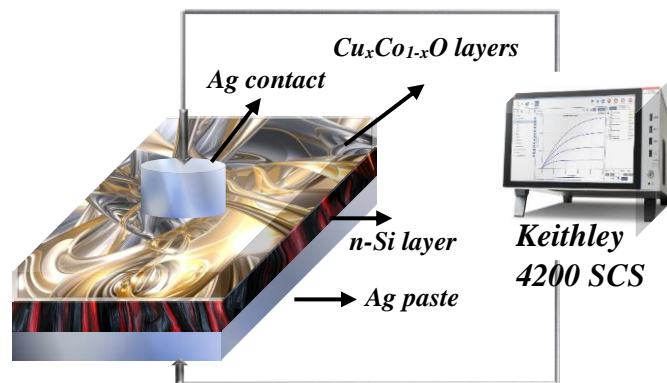


Figure 1. Schematic diagram of $Cu_xCo_{1-x}O/n\text{-Si}$ structures with electrical characterization system

Thus, Ag/Cu_xCo_{1-x}O/n-Si heterojunction diodes were obtained. The schematic diagram of the heterojunction structures formed is shown in Figure 1.

3. Results and Discussions

In order to examine the heterojunction application of Cu_xCo_{1-x}O thin films, the electrical properties of Ag/Cu_xCo_{1-x}O/n-Si diodes were investigated by taking conventional current-voltage measurements. Figure 2 illustrates the Log I-V measurements of the produced heterojunction diodes, showcasing both the reverse and forward bias behavior.

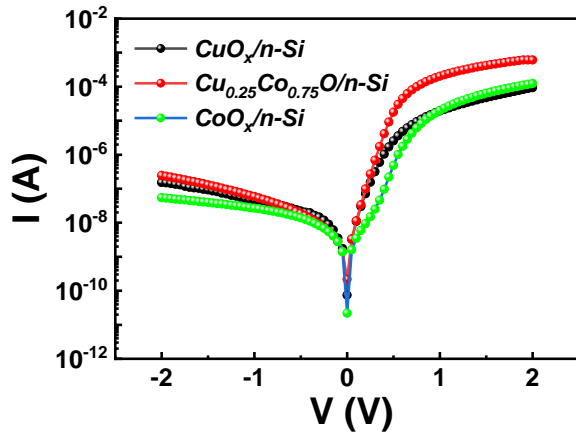


Figure 2. The I-V characteristics of Cu_xCo_{1-x}O/n-Si structures under dark condition

These measurements were conducted in a dark environment at room temperature for the voltage range of -2 and +2 V. It is important to measure the electrical properties of heterojunction diodes under dark conditions in order to eliminate the photovoltaic effect, perform the characterization of dark current and eliminate external variables. This form of electrical measurement helps to obtain reliable data about the properties of diodes and to develop efficient and stable devices. The obtained I-V characteristics were used to calculate various electrical parameters of the heterojunction, including the barrier height (Φ_B), which is representation the energy difference between the metal/semiconductor interface, reverse saturation current (I_0), which is a measure of the leakage current in the reverse biased diode, rectification ratio, which is a measure of the diode's ability to conduct current in one direction (forward bias) compared to the opposite direction (reverse bias), and ideality factor (n), which indicates how closely the diode adheres to the ideal diode equation. It takes into account the deviations from ideality due to factors like recombination, series resistance, and trapping effects (Pür and Tataroğlu 2012; Namkoong et al. 2013). The equations commonly used to

calculate the electrical parameters for diodes are as follows (Sze and Ng 2006);

$$I = I_0 \left[\exp \left(\frac{q(V - IR_s)}{nkT} \right) - 1 \right] \quad (1)$$

In the equation, I_0 represents the reverse saturation current, while n denotes the ideality factor. The symbols q , k , T , and R_s correspond to the elementary charge, Boltzmann constant, temperature of measurement, and series resistance, respectively. The n value, a dimensionless parameter indicating the deviation from ideal behavior, equals 1 in the case of ideal diodes.

Table 1. The determined electrical characteristics of the diodes

Diodes	CoO _x /n-Si	Cu _{0.25} Co _{0.75} O _x /n-Si	CuO _x /n-Si
RR (dark, ±2V)	2.92×10 ³	1.19×10 ³	1.29×10 ³
n (I-V)	3.19	1.99	2.19
Φ_B (eV) (I-V)	0.78	0.77	0.78
Φ_B (eV) Norde	0.81	0.84	0.81
Φ_B (eV) C-V	1.69	1.89	0.79
Vbi (eV) C-V	1.49	1.69	0.60
R_s (kΩ) (Norde)	20.6	2.6	133.3

The value of n can be determined using the following equation;

$$n = \frac{q}{kT} \left[\frac{dV}{d \ln I} \right] \quad (2)$$

The ideality factor (n) values of the diodes were ascertained by analyzing the linear region slope of the $\ln(I)$ versus V graph under forward bias conditions. These values are subsequently presented alongside other pertinent electrical parameters, including barrier height (Φ_B) and rectification ratio (RR), in Table 1. Upon introducing Cu doping, a discernible reduction in the ideality factor (n) is noted, suggesting an enhancement in the overall quality of the diodes. In the literature, it has been reported that tunneling, generation-recombination and leakage current mechanism are dominant in current-conduction mechanisms, respectively, when n values are $1 < n < 2$, $n=2$ and $n > 2$ for diodes (Ay and Tolunay 2007; Sharma et al. 2018). In our case, the ideality factor was observed as 1.99 for Cu_{0.25}Co_{0.75}O/n-Si and it was observed that the copper doping brought the diode closer to the ideal. The series resistance and barrier height of Cu_xCo_{1-x}O/n-Si diodes can also be defined by Norde's method (Norde 1979). Following equation gives the definition of Norde's function;

$$F(V) = \frac{V_0}{\gamma} - \frac{kT}{q} \ln\left(\frac{I(V)}{A^*AT^2}\right) \quad (3)$$

The parameter γ is an integer value exceeding the ideality factor associated with diodes, and it possesses no physical dimensions. The current derived from the I-V characteristic is known as the $I(V)$. The graph of $F(V)$ vs. voltage for the diodes is shown in Figure 3.

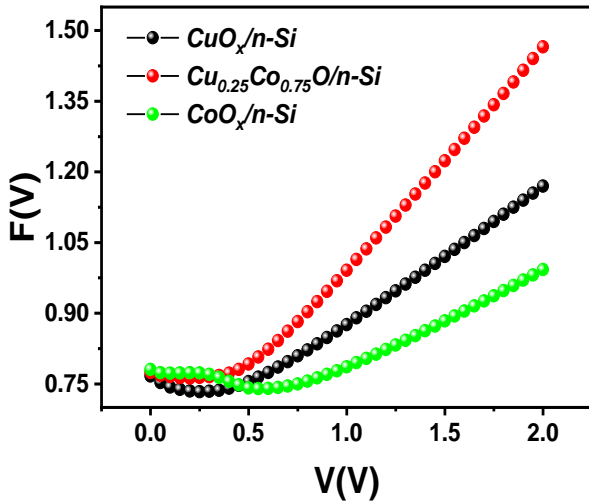


Figure 3. The $F(V)$ - V of $\text{Cu}_x\text{Co}_{1-x}\text{O}/\text{n-Si}$ structures under dark condition

Due to the existence of a minimum point in the $F(V)$ curve, the expressions for barrier height and series resistance are as follows:

$$\phi_B = F(V_0) + \frac{V_0}{\gamma} - \frac{kT}{q} \quad (4)$$

$$R_s = \frac{kT(\gamma-n)}{qI_0} \quad (5)$$

where V_0 is the corresponding voltage and $F(V_0)$ is the minimal point of $F(V)$. The calculated Φ_B and R_s parameters were tabulated in Table 1. A significant decrease in R_s was observed with the doping of copper to CoO_x . The series resistance in a heterojunction structure is the sum of the resistances of the different layers and interfaces that make up the device. The series resistance of the diode is influenced by several factors, including the SiO_2 of n-Si at the junction, the resistance of the contact wires, the resistance of the semiconductor layers, and the quality of the interface between oxide semiconductor and n-Si (Cifci et al. 2018; Buyuk and Ilican 2020). Consequently, the improvement of any of these parameters or the total diode's ohmic loss, along with doping, are linked to the decrease in R_s .

In order to comprehend the conduction mechanisms, we examine the I-V characteristic of the manufactured diode, which is sketched on a log-log scale as depicted in Figure 4.

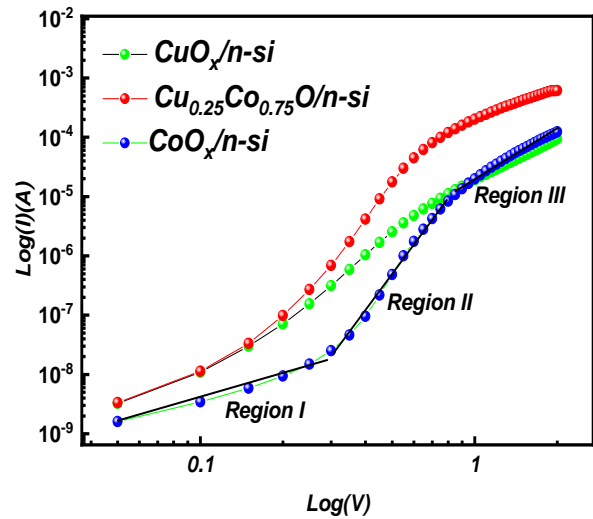


Figure 4. The $\text{Log}(I)$ - $\text{Log}(V)$ of $\text{Cu}_x\text{Co}_{1-x}\text{O}/\text{n-Si}$ structures under dark condition

The logarithmic graph reveals a power law pattern in the current ($I \propto V^m$), with various exponent values (m). The charge transfer mechanism is determined by the constant exponent value m . Figure 4 prominently exhibits three well-defined linear voltage domains, delineated by transitional segments, within the double logarithmic representation of the forward bias current-voltage (I-V) characteristics. These segments are characterized by distinct slopes, aligning with different conduction mechanisms. The existence of deep trap states at the p-n junction interface is typically the cause of these various current transport processes in diodes (Ahmed et al. 2019). Figure 5 illustrates the C-V graph of diodes in the frequency range of 10 kHz-1 MHz. The C-V graph clearly shows that the applied voltage and frequency have a substantial impact on the diodes' capacitance value. The presence of interface states leads to a decline in the capacitance value of the diodes as the frequency increases within a specific positive voltage range in the forward bias region (Khusayfan 2016; Cavdar et al. 2016). The interface states of the diodes exhibit an inability to accurately follow the AC signal at elevated applied frequencies. As a result, they do not significantly influence the diode's capacitance, resulting in a reduced capacitance value at higher frequencies (Caglar et al. 2016; Aydin et al. 2016). The depletion region in the diodes has a capacitive effect.

For this purpose, capacitance voltage measurements are made and C-V graphs are drawn. Then, electrical characterization is made using these graphs. The barrier height and carrier concentration of the rectifier contact are calculated with the capacity measured depending on the reverse supply voltage.

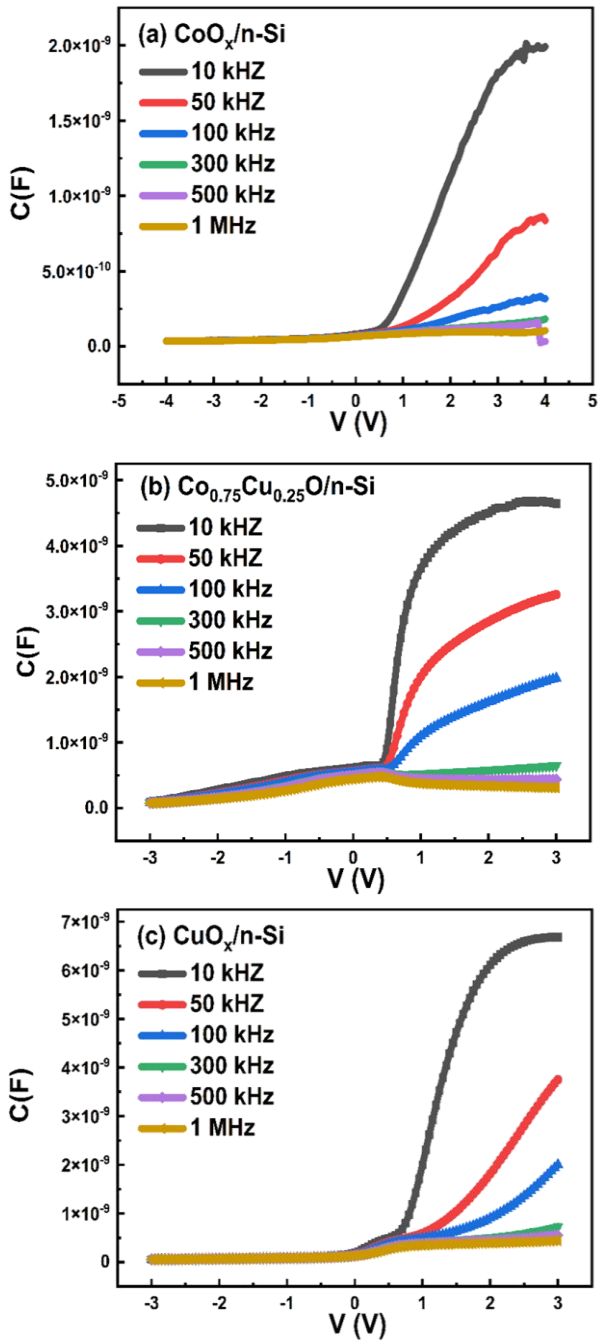


Figure 5. The C-V characteristics of Cu_xCo_{1-x}O/n-Si structures under dark condition

The C⁻²-V graphs of the diodes were plotted by using C-V data and shown in Figure 6. The extrapolation of C⁻² - V curves along the voltage axis serves to ascertain the diffusion potential (V_d). Meanwhile, the gradient of this curve (refer to Table 1) imparts information regarding the doping concentrations on both sides of the structural configuration. The capacitance of the junction region of a heterojunction diode is expressed by the following formula [25];

$$C^2 = \left[\frac{q\epsilon_{S1}\epsilon_{S2}N_A N_D}{2(\epsilon_{S1}N_A + \epsilon_{S2}N_D)} \right] \frac{1}{(V_{bi} - V)} \quad (6)$$

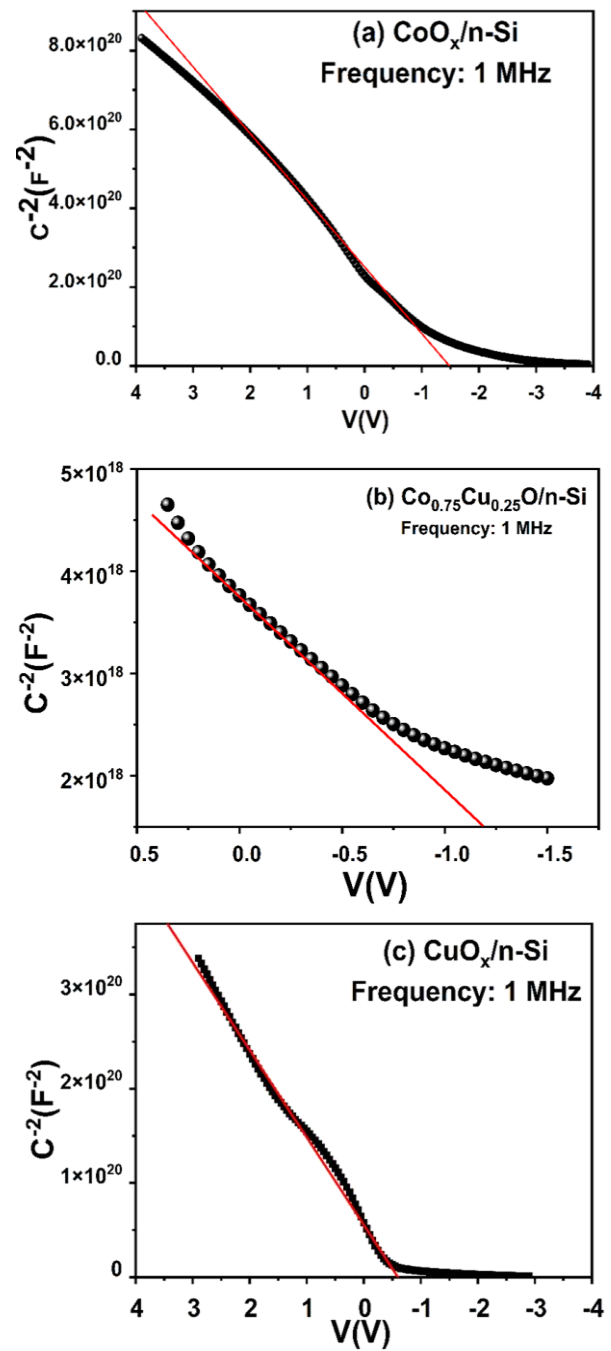


Figure 6. The 1/C²-V characteristics of Cu_xCo_{1-x}O/n-Si structures at 1 Mhz

In Equation (6), q refers to the electron charge, ϵ_{S1} and ϵ_{S2} are the dielectric constants, N_A and N_D are the acceptor and donor concentrations of the semiconductors, respectively, and V_{bi} is the built in potential. The Equation 6 can be expressed more simply;

$$\frac{1}{C^2} = \left[\frac{2(V_{bi} - V)}{A^2 q \epsilon_{S1} N_A} \right] \quad (6)$$

In this expression, A is the effective diode area. The electrical parameters derived from the C⁻²-V graph are presented in Table 1. Upon scrutinizing the Φ_B (C-V)

values derived from the C-V data, it becomes evident that they surpass the Φ_B value ascertained from the current-voltage (I-V) data. This discrepancy in barrier heights arises from the intrinsic distinctions between the C-V and I-V methodologies. Furthermore, the dissonance between Φ_B (C-V) and Φ_B (I-V) may be rationalized by the influence of excess capacitance (Mönch 1994; Karataş et al. 2016).

4. Conclusions

The CoOx, Cu-doped CoOx, and CuOx thin films were coated by using the sol gel spin coating method on n-Si substrates. We examined the electrical characteristics of heterojunction structures obtained from these thin films. The findings demonstrate that Cu doping has a major impact on the CoOx/n-Si diode's electrical characteristics. The dark I-V characteristics of all diodes indicate rectification behavior. I-V data was utilized to determine the critical junction parameters, including R_s , RR , n , and Φ_B . The ideality factor values for the configurations of CoOx/n-Si, Cu-doped CoOx/n-Si, and CuOx/n-Si were determined to be 3.19, 1.99, and 2.19 eV, correspondingly. The lowest series resistance was obtained from the $\text{Cu}_{0.25}\text{Co}_{0.75}\text{Ox/n-Si}$ diode with 2.6 k Ω . Additionally, measurements of the diodes' C-V properties were made between the frequencies of 10 kHz and 1 MHz. The findings show that copper doping has an affirmative effect to regulate the electrical characteristics of CoOx/n-Si diodes.

Declaration of Ethical Standards

This study is obtained from Yusuf YILDIZ's MS.c. Thesis titled "Electrical characterization of cobalt doped copper oxide thin film based structures" dated April 6, 2022. The supervisor of the thesis is Assoc. Prof. Dr. Şerif RÜZGAR.

The authors declare that they comply with all ethical standards.

Credit Authorship Contribution Statement

Author-1: Methodology, conceptualization, investigation.

Author-2: Formal analysis, Data curation, Writing – original draft, supervision

Declaration of Competing Interest

The authors have no conflicts of interest to declare regarding the content of this article.

Data Availability Statement

All data generated or analyzed during this study are included in this published article.

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