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Environmentally Benign ZnS/RGO Nanocomposite Buffer Layers for High-Performance Cu₂O/CdTe Thin-Film Solar Cells

Ahmed Mostafa^{1,*}, Samar Al-Shehri², Azzah A. Alshehri³ and A. Ashour⁴

¹Department of Physics, Faculty of Science, Suez University, Suez, Egypt

²Department of Physics, College of Science, University of Bisha, Bisha, Saudi Arabia

³Department of Chemistry, College of Science, University of Bisha, Bisha, Saudi Arabia

⁴Department of Physics, Faculty of Science, Islamic University of Madinah, Madinah, Saudi Arabia

*Corresponding Author: Ahmed Mostafa. Email: am1662184@gmail.com or amostafa0004@gmail.com

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ABSTRACT: ABSTRACT: Interface engineering is an effective strategy for improving the efficiency and operational stability of thin-film photovoltaic devices. In this work, zinc sulfide/reduced graphene oxide (ZnS/RGO) nanocomposites (NCs) were synthesized using environmentally benign methods and incorporated as buffer layers into Glass/ITO/Cu₂O/CdTe/ZnS–RGO/Al solar cells. X-ray diffraction (XRD) measurements confirmed the formation of phase-pure hexagonal ZnS, with crystallite sizes ranging from 18 to 26 nm. The introduction of RGO resulted in lattice microstrain values of $(3.1\text{--}6.4) \times 10^{-4}$, suggesting effective interfacial coupling within the composite structure. X-ray photoelectron spectroscopy (XPS) provided further evidence for the successful integration of ZnS and RGO, with the characteristic Zn 2p and S 2p core-level signals corresponding to Zn²⁺–S bonding, together with a distinct C 1s contribution from RGO and an O 1s component associated with residual oxygen-containing surface functionalities. These spectroscopic features support the presence of strong interactions at the ZnS/RGO interface. Optical characterization showed enhanced absorption throughout the 300–900 nm wavelength range and a progressive reduction in the optical band gap as the RGO content increased. This behavior was accompanied by substantial photoluminescence quenching, indicating more efficient interfacial charge transfer and reduced radiative recombination. Incorporation of the ZnS/RGO buffer layer substantially enhanced the photovoltaic response of the devices, yielding a short-circuit current density of approximately 29 mA cm^{−2}, an open-circuit voltage of approximately 0.82 V, and a fill factor of approximately 72%. Consequently, a maximum power conversion efficiency of approximately 17.1% was achieved. Electrochemical impedance measurements revealed a low series resistance of 2.5–4.0 Ω cm², together with a pronounced increase in recombination resistance to nearly 3.5×10^3 Ω cm², demonstrating effective inhibition of interfacial charge recombination. In addition, temperature-dependent electrical measurements performed between 280 and 320 K revealed improved thermal stability and a lower degree of performance deterioration for the devices incorporating the ZnS/RGO buffer layer. Overall, the results demonstrate the potential of environmentally benign ZnS/RGO NCs as efficient and sustainable buffer-layer materials for advanced thin-film photovoltaic applications.

KEYWORDS: ZnS/RGO nanocomposites; buffer layer engineering; thin-film solar cells; interface recombination; impedance spectroscopy; external quantum efficiency; environmentally benign synthesis

1 Introduction

Thin-film photovoltaic technologies have emerged as leading candidates for next-generation solar energy conversion owing to their reduced material consumption, low fabrication cost, and compatibility

with large-area and flexible substrates [1–4]. Among these technologies, CdTe-based solar cells have achieved considerable attention due to their near-ideal direct band gap (~ 1.45 eV), high optical absorption coefficient ($>10^5$ cm $^{-1}$), and well-established industrial scalability [5–8]. Despite these advantages, further improvements in device performance remain constrained by interfacial losses, particularly at the junctions involving the buffer layer and absorber [8,9]. These interfacial limitations lead to increased recombination, non-ideal band alignment, and resistive barriers, which collectively degrade charge collection efficiency, open-circuit voltage, and fill factor.

The buffer layer plays a pivotal role in determining the overall performance of thin-film solar cells by mediating charge transport across heterointerfaces, optimizing band alignment, and suppressing defect-assisted recombination [10–13]. In this context, ZnS has been extensively investigated as a buffer material due to its wide band gap, low optical absorption in the visible region, favorable conduction band offset with CdTe, and good chemical stability. However, pristine ZnS suffers from inherently low electrical conductivity and limited carrier mobility, which can impede efficient electron transport and lead to charge accumulation at the interface [13,14]. These drawbacks necessitate the development of modified ZnS-based buffer layers with enhanced electrical conductivity and improved interfacial properties.

Recent advances in nanostructured materials and interface engineering have highlighted the potential of incorporating conductive carbon-based materials, particularly RGO, into semiconductor matrices to improve device performance [14–20]. RGO possesses exceptional electrical conductivity, high carrier mobility, large specific surface area, and tunable surface chemistry, making it an effective candidate for facilitating charge transport and improving interfacial contact. When integrated with semiconductor nanostructures, RGO can act as an efficient electron transport pathway, reducing recombination losses by promoting rapid charge separation and extraction [16,18]. Furthermore, graphene-based materials have been widely reported to enhance the optoelectronic properties of photovoltaic devices through improved carrier dynamics, interfacial engineering, and enhanced electrical percolation networks. In addition, the adoption of environmentally benign synthesis routes for ZnS and RGO aligns with current trends toward sustainable and green energy materials [20].

Although previous studies have explored ZnS- and graphene-based buffer layers, a systematic understanding of the concentration-dependent role of RGO in modulating interfacial charge transport, recombination dynamics, and overall device performance remains insufficient [6–10]. In particular, the correlation between material composition, carrier lifetime, and photovoltaic parameters has not been fully established. Addressing this gap is essential for the rational design and optimization of high-performance buffer layers in thin-film solar cells [21].

In this work, ZnS/RGO NCs with controlled RGO content were synthesized using environmentally benign methods and employed as buffer layers in Glass/ITO/Cu $_2$ O/CdTe/ZnS–RGO/Al solar cell architectures. A comprehensive investigation of the structural, optical, and electrical properties was conducted to elucidate the impact of RGO incorporation on device performance. Special emphasis was placed on impedance spectroscopy, external quantum efficiency analysis, and temperature-dependent electrical measurements to probe interfacial charge transport mechanisms and recombination processes. The results demonstrate that optimized incorporation of RGO significantly enhances charge transport, suppresses recombination losses, and improves overall photovoltaic performance and operational stability. These findings provide valuable insights into the design of advanced hybrid buffer layers and highlight the potential of ZnS/RGO NCs for next-generation thin-film solar cell applications.

2 Materials and Experimental Systems

2.1 Materials

Zinc acetate dihydrate, $\text{Zn}(\text{CH}_3\text{COO})_2 \cdot 2\text{H}_2\text{O}$, $\geq 99.0\%$, Sigma-Aldrich, and thiourea, $\text{CH}_4\text{N}_2\text{S}$, $\geq 99.0\%$, Sigma-Aldrich, were used as precursors for the synthesis of ZnS nanoparticles. A naturally derived organic chelating agent, analytical grade, Sigma-Aldrich, was employed in the sol–gel process. Graphite rods obtained from spent zinc–carbon batteries were used as the carbon source for the preparation of reduced graphene oxide (RGO). Sulfuric acid, H_2SO_4 , 95–98%, Merck, served as the electrolyte during the electrochemical exfoliation process. Deionized water, resistivity $\geq 18.2 \text{ M}\Omega\text{-cm}$, was used in all solution preparation and washing steps. Copper precursor material for the synthesis of Cu_2O nanoparticles, $\geq 99.0\%$, Sigma-Aldrich, was used without further purification. All chemicals were of analytical grade and were used as received.

2.1.1 Environmentally Benign Synthesis of ZnS Nanoparticles (Sol–Gel Route)

An alternative environmentally benign synthesis of ZnS nanoparticles was carried out using a sol–gel route assisted by eco-friendly organic complexing agents, as illustrated in Fig. 1. In this method, a zinc precursor was dissolved in deionized water and subsequently coordinated with a naturally derived chelating agent to ensure homogeneous distribution of zinc ions at the molecular level. A sulfur source was then gradually introduced under mild stirring, resulting in the formation of a stable sol that evolved into a gel through controlled hydrolysis and condensation processes. The obtained gel was aged and dried at low temperature, followed by gentle thermal treatment to yield ZnS nanoparticles without the use of toxic solvents or harsh chemicals. This environmentally benign sol–gel approach allows precise control over particle size, crystallinity, and surface characteristics while maintaining low energy consumption, thereby producing ZnS nanoparticles well suited for sustainable photovoltaic and optoelectronic applications [11].

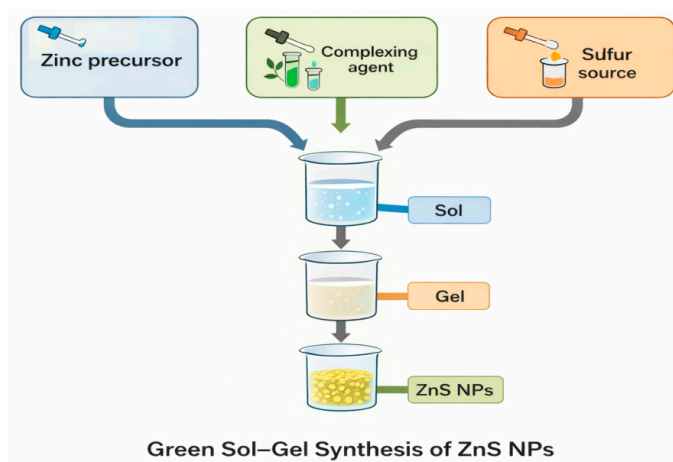


Figure 1: Environmentally benign synthesis of ZnS NPs using sol-gel method [12].

2.1.2 Environmentally Benign Synthesis of RGO Nanosheets Using an Electrochemical Method

RGO was synthesized using a novel, simple, and efficient electrochemical exfoliation method, as illustrated in Fig. 2. The electrochemical cell consisted of two graphite electrodes obtained from waste carbon sources, such as spent batteries, thereby promoting resource reutilization and sustainability. In this method, no chemical reducing agent is used; instead, the reduction of RGO occurs electrochemically via electron transfer under an applied DC voltage, providing a clean and environmentally friendly alternative to

conventional chemical reduction routes. An acidic electrolyte, typically sulfuric acid (H_2SO_4) with varying concentrations, was employed, and a DC voltage in the range of 10–12 V was applied to induce interlayer exfoliation of the graphite. The electrochemical process resulted in the gradual formation of exfoliated graphene-based material suspended in the electrolyte. After completion of the reaction, the resulting mixture was allowed to stand for a sufficient period to enable the precipitate to settle at the bottom of the cell. The collected material was then thoroughly washed multiple times with distilled water to remove residual electrolyte and impurities, followed by drying in an oven at 50–60°C to obtain fine RGO powder suitable for subsequent nanocomposite fabrication.

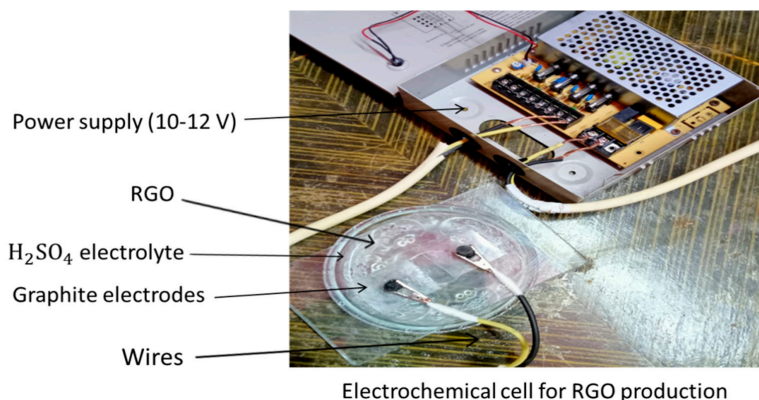


Figure 2: An electrochemical cell for RGO production using novel method.

2.1.3 Environmentally Benign Fabrication of ZnS/RGO NCs

ZnS/RGO NCs with systematically controlled RGO contents were synthesized using a sustainable, solution-based fabrication route as indicated in Fig. 3 to elucidate the influence of RGO incorporation on the structural and optoelectronic characteristics of the nanocomposites [12]. GO was first uniformly dispersed in deionized water via ultrasonication to form a stable colloidal suspension. Subsequently, environmentally benign-synthesized ZnS NPs were introduced under continuous stirring, followed by the addition of an eco-friendly reducing agent that enabled the *in situ* reduction of GO to RGO while promoting strong interfacial coupling between ZnS NPs and the RGO sheets. By precisely regulating the RGO loading, a series of nanocomposites—ZnS/00%RGO, ZnS/05%RGO, ZnS/10%RGO, ZnS/15%RGO, and ZnS/20%RGO—were successfully obtained. This green fabrication strategy ensures uniform dispersion of ZnS within the conductive RGO matrix, effectively suppresses nanoparticle agglomeration, and facilitates efficient charge transport. The controlled variation of RGO content provides a reliable pathway for optimizing nanocomposite performance, highlighting the suitability of ZnS/RGO NCs as eco-friendly buffer layers for high-efficiency photovoltaic devices [13].

2.1.4 Environmentally Benign Synthesis of Cu_2O Nanoparticles (Sol–Gel Route)

Cu_2O nanoparticles were synthesized via an eco-friendly sol–gel route designed to avoid toxic reagents and harsh processing conditions. In this method, an aqueous solution of a copper precursor was prepared and complexed with a naturally derived chelating agent to ensure homogeneous distribution of Cu^+ ions at the molecular level. Controlled hydrolysis and condensation reactions were initiated under mild stirring, leading to the formation of a stable sol that gradually evolved into a gel [15]. The resulting gel was aged and subsequently dried at low temperature, followed by gentle thermal treatment under a controlled atmosphere to obtain phase-pure Cu_2O nanoparticles. This green sol–gel approach enables effective control

over particle size, crystallinity, and surface characteristics while maintaining low energy consumption and environmental impact, making the synthesized Cu_2O nanoparticles suitable for hole transport layers in sustainable photovoltaic applications.

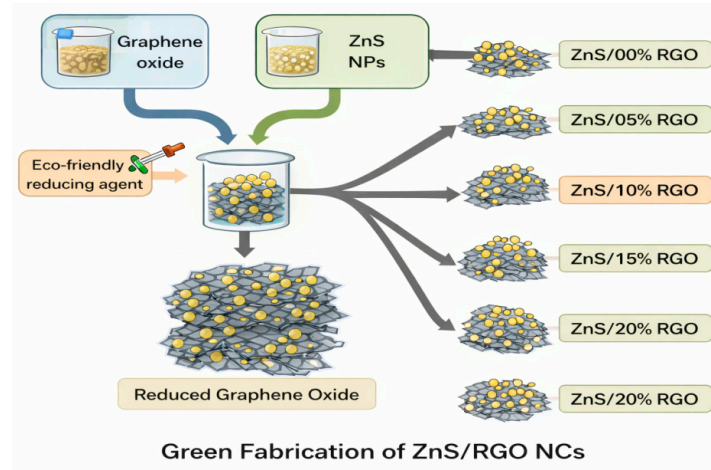


Figure 3: Environmentally benign synthesis of ZnS/RGO NCs [14].

2.2 Fabrication of ZnS/RGO-Based Thin-Film Solar Cell Device

The ZnS/RGO-based thin-film solar cell was fabricated following the device architecture illustrated in Fig. 4, consisting of Glass/ITO/ Cu_2O /CdTe/ZnS:RGO/Al. All functional thin films were sequentially deposited under controlled conditions to ensure high film quality, uniform thickness, and optimized interfacial properties [16].

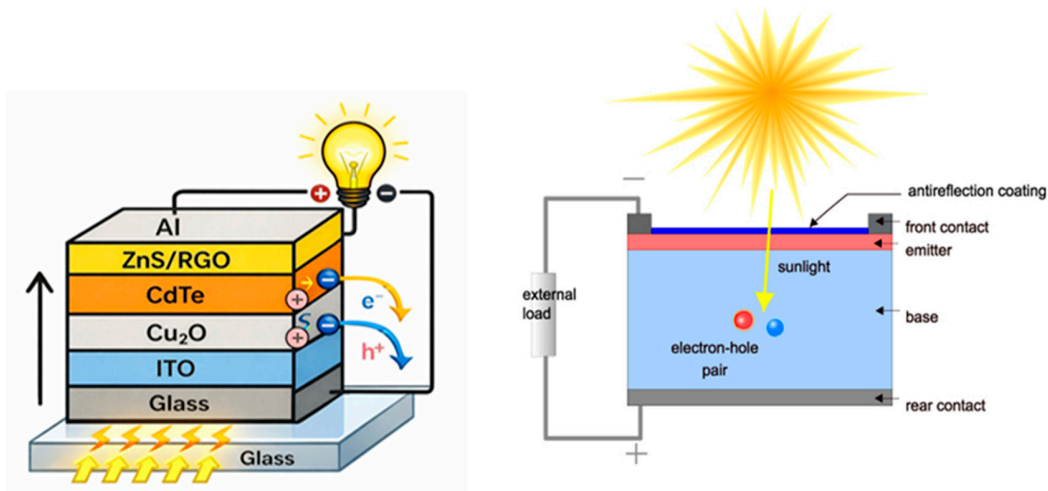


Figure 4: Schematic illustration of the ZnS/RGO-based thin-film solar cell architecture (Glass/ITO/ Cu_2O /CdTe/ZnS:RGO/Al) and the corresponding charge-carrier transport mechanism under illumination.

2.2.1 Preparation of ITO-Coated Glass Substrates

ITO-coated glass substrates which possess a typical electrical resistivity of $\sim 10^{-4} \Omega \cdot \text{cm}$, were employed as transparent conducting electrodes and light-incident windows. Prior to deposition, the substrates were ultrasonically cleaned in acetone, ethanol, and deionized water for 15 min each to remove surface

contaminants. The cleaned substrates were dried under nitrogen flow and treated with UV–ozone to improve surface wettability and interfacial adhesion [17].

2.2.2 Deposition of Cu_2O Hole Transport Layer

The p-type Cu_2O hole transport layer was deposited onto the prepared ITO substrates using a thermal evaporation technique. Film deposition was carried out in a Denton Vacuum DV-502A coating unit under a high vacuum of approximately 10^{-7} mbar. The deposition rate was maintained at $20 \text{ \AA s}^{-1}$, while the substrate temperature was held at 150°C to promote film uniformity and crystallinity. Film thickness was precisely controlled to ~ 100 nm using an FTM-6 quartz crystal thickness monitor [18].

2.2.3 Fabrication of CdTe Absorber Layer

The CdTe absorber layer was subsequently deposited onto the Cu_2O -coated substrates using the same thermal evaporation system under identical high-vacuum conditions. The substrate temperature was maintained at 150°C , and the deposition rate was fixed at $20 \text{ \AA s}^{-1}$. The thickness of the CdTe thin film was controlled at approximately 100 nm using the FTM-6 thickness monitor. Post-deposition thermal treatment was applied to enhance grain growth, reduce defect density, and improve photoactive properties [19].

2.2.4 Deposition of ZnS/RGO NCs Buffer Layer

ZnS/RGO NC buffer layers with different RGO loadings (ZnS/00%RGO, ZnS/05%RGO, ZnS/10%RGO, ZnS/15%RGO, and ZnS/20%RGO) were deposited onto the CdTe absorber using thermal evaporation. Deposition was conducted in the Denton Vacuum DV-502A system under a vacuum level of 10^{-7} mbar, with a deposition rate of $20 \text{ \AA s}^{-1}$ and a substrate temperature of 150°C . The thickness of the ZnS/RGO layer was maintained at approximately 100 nm and monitored *in situ* using the FTM-6 thickness controller. This buffer layer improves energy-band alignment, suppresses interfacial recombination, and facilitates efficient electron transport toward the top electrode [20].

2.2.5 Deposition of Aluminum Top Electrode

An aluminum (Al) metal electrode was deposited onto the ZnS/RGO layer by thermal evaporation under high vacuum using the Denton Vacuum DV-502A coating unit. The deposition rate and thickness were carefully controlled to ensure uniform coverage and low contact resistance. Aluminum serves as the electron-collecting electrode, completing the device structure.

2.2.6 Device Architecture and Operating Mechanism

The finalized solar cell device adopts the configuration Glass/ITO/ Cu_2O /CdTe/ZnS–RGO/Al, as illustrated in Fig. 4. Upon illumination through the glass/ITO side, photogenerated electron–hole pairs are generated within the CdTe absorber. Holes are transported toward the ITO electrode via the Cu_2O layer, while electrons are efficiently extracted through the ZnS/RGO buffer layer to the Al electrode. The incorporation of the ZnS/RGO NCs enhances interfacial charge separation, minimizes recombination losses, and improves overall device performance.

2.3 Experimental Systems

2.3.1 XRD Characterization and Data Analysis

XRD measurements were conducted using a JEOL 60PA-JSDX diffractometer at the Central Laboratory of El-Minia University. The instrument was operated at an accelerating voltage of 42 kV and a tube

current of 36 mA, employing Cu K α radiation with a wavelength of $\lambda = 0.145184$ nm. Diffraction data were collected over a 2θ range of 30° – 90° at a scanning rate of 1° min^{-1} with a time constant of 1 s. This scanning configuration enabled reliable phase identification and assessment of preferred crystallographic orientation. The recorded diffraction patterns were further analyzed to extract microstructural parameters using Rietveld refinement implemented in the MAUD software package, supported by additional analytical tools for comprehensive structural evaluation [21].

2.3.2 Microscopic Characterization of ZnS Nanoparticles and ZnS/RGO NCs

The morphology and size distribution of the synthesized nanostructures were examined using a high-resolution scanning electron microscope (SEM, JEOL JSM-IT200) operated at an accelerating voltage of 25 kV. Detailed analysis of particle dimensions, crystallinity, and interfacial features of the ZnS nanoparticles and ZnS/RGO NCs was further carried out by high-resolution transmission electron microscopy (HRTEM, JEOL 3010, Japan) at an accelerating voltage of 200 kV. In addition, the surface morphology, dispersion uniformity, and elemental composition of the nanostructures were investigated using a ZEISS EVO-MA10 SEM equipped with an energy-dispersive X-ray spectroscopy (EDX) detector. This combined microscopic approach enabled comprehensive structural and compositional evaluation of the synthesized materials [22].

2.3.3 FTIR Instrument

FTIR spectra were recorded in transmission mode using a Spectrum One spectrometer (PerkinElmer, USA) over the wavenumber range of 4000 – 800 cm^{-1} with a spectral resolution of 4 cm^{-1} . For each sample, 20 scans were averaged to enhance the signal-to-noise ratio. Prior to measurement, the powdered samples were thoroughly mixed with potassium bromide (KBr) at a mass ratio of 2:300 (sample:KBr) and pressed into transparent pellets, while a pure KBr pellet was used as the background reference. FTIR analysis was employed to identify characteristic functional groups and to investigate the chemical bonding environment within the synthesized materials, thereby confirming the successful formation of the nanocomposite structures and the interactions between their constituent components [8,23].

2.3.4 Raman Spectroscopy Measurements

Raman spectra were acquired using an inVia Raman spectrometer (Renishaw, Gloucestershire, UK) equipped with a thermoelectrically cooled charge-coupled device (CCD) detector maintained at -80°C . A He–Ne laser with an excitation wavelength of 532 nm was used as the light source, while the laser power was carefully limited to 1 mW to avoid thermal effects on the samples. Each spectrum was collected with an integration time of 100 s, ensuring a high signal-to-noise ratio and reliable spectral analysis [24].

2.3.5 TGA System

TGA was employed to examine the thermal decomposition behavior of the samples, while differential thermal analysis (DTA) was used to identify the corresponding thermal transitions. The measurements were carried out using a Q50 thermogravimetric analyzer (TA Instruments, USA) to evaluate the thermal stability of ZnS nanoparticles and ZnS/RGO NCs. Approximately 10 mg of each sample was heated from 35 to 1000°C at a constant heating rate of $10^\circ\text{C min}^{-1}$ under a nitrogen atmosphere to ensure inert conditions throughout the analysis [13]. This approach enabled reliable assessment of weight-loss profiles and thermal stability characteristics of the investigated materials.

2.3.6 DRS System

Diffuse reflectance spectra of the ZnS/RGO NCs were recorded using a Shimadzu UV-2600 UV-Vis spectrophotometer at the National Research Center, Cairo. The measurements were performed over the ultraviolet-visible spectral range to evaluate the optical response of the samples. The obtained reflectance data were converted into absorbance using the Kubelka-Munk function, $F(R)$, according to the relation [13]:

$$F(R) = \frac{(1 - R)}{2R} \quad (1)$$

where (R) represents the diffuse reflectance of a semi-infinite sample. This transformation enables reliable estimation of the optical band gap (E_g) for both bulk and nanostructured ZnS/RGO composites through appropriate optical analyses.

2.3.7 Electrical Characterization of ZnS/RGO-Based Thin-Film Solar Cells

The current density-voltage (J - V) characteristics of the Glass/ITO/Cu₂O/CdTe/ZnS-RGO/Al solar cells were evaluated using a Keithley 2400 SourceMeter (Tektronix, USA) under simulated AM 1.5G illumination. A Newport Oriel Sol3A Class AAA solar simulator (Newport Corporation, USA), calibrated with a silicon reference cell, was employed to deliver an incident power density of 100 mW cm⁻². All J - V measurements were performed at room temperature. External quantum efficiency (EQE) spectra were acquired using a QE-R quantum efficiency measurement system (Enlitech, Taiwan) equipped with a xenon lamp, monochromator, optical chopper, and lock-in amplifier. The incident photon flux was calibrated using a certified silicon photodiode, and measurements were conducted over the wavelength range of 300–900 nm under zero-bias conditions [24].

Photoelectrochemical impedance spectroscopy (PEIS) measurements were carried out using a Gamry Reference 3000 potentiostat/galvanostat (Gamry Instruments, USA). Impedance spectra were recorded over a frequency range of 1 Hz to 1 MHz with an AC perturbation amplitude of 10 mV. Bias-dependent PEIS measurements were performed under dark conditions at applied DC voltages between 0.0 and 0.8 V, while temperature-dependent measurements were conducted at 280, 300, and 320 K using a temperature-controlled sample stage. All electrical characterization experiments were conducted at the National Research Centre (NRC), Cairo, Egypt.

2.3.8 Manuscript Revision and AI-Assisted Language Editing

During revision of this manuscript, OpenAI GPT-5.6 Luna was used as an AI-assisted language- and consistency-editing tool to improve grammatical clarity, streamline repetitive wording, and audit the consistency of in-text citations and the reference list. The authors independently reviewed and verified all scientific interpretations, experimental details, numerical values, figure descriptions, citations, and references, and retain full responsibility for the accuracy and integrity of the manuscript.

3 Results and Discussions

3.1 Structural Analysis

XRD was employed to comprehensively investigate the structural properties, phase purity, and crystallographic features of pristine ZnS, ZnS/RGO NCs with varying RGO contents, and the Cu₂O layer used in the solar cell device. This section discusses the structural evolution of ZnS upon RGO incorpo-

ration, detailed crystallographic contributions through peak deconvolution, and quantitative evaluation of crystallite size, lattice microstrain, and defect density using W–H analysis. In addition, the crystalline phase and orientation of the Cu_2O layer are examined to confirm its structural integrity and suitability for photovoltaic applications.

3.1.1 Structural Evolution of ZnS/RGO NCs with Varying RGO Content

The XRD patterns of pristine ZnS and ZnS/RGO NCs with different RGO loadings (0–20 wt%) are presented in Fig. 5. All samples exhibit distinct diffraction peaks located at approximately 28.5° , 33.1° , 47.5° , and 56.3° , which correspond to the (100), (002), (101), and (110) crystallographic planes of hexagonal wurtzite ZnS, respectively, confirming the successful formation of crystalline ZnS. An additional weak reflection observed at higher angles is indexed to the (112) plane, further supporting phase purity. With increasing RGO content, the characteristic ZnS diffraction peaks are retained without the emergence of secondary phases, indicating that the incorporation of RGO does not alter the crystal structure of ZnS. A slight reduction in peak intensity accompanied by marginal peak broadening is observed as the RGO loading increases, which can be attributed to the intimate interfacial interaction between ZnS nanoparticles and the RGO sheets as well as a decrease in crystallite size. The absence of a distinct diffraction peak corresponding to RGO suggests its low content and disordered nature within the nanocomposite matrix. Overall, the XRD results demonstrate the successful formation of ZnS/RGO nanocomposites with preserved ZnS crystallinity and effective RGO incorporation.

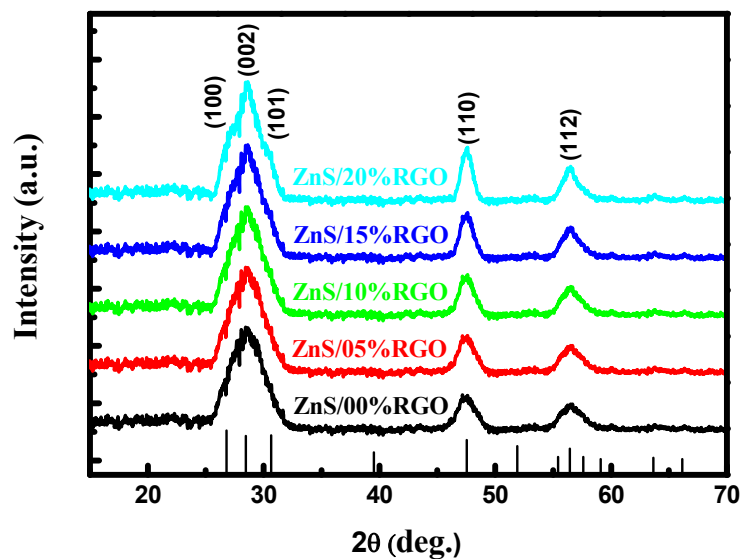


Figure 5: XRD patterns of pristine ZnS and ZnS/RGO NCs with different RGO loadings.

3.1.2 Peak Deconvolution Analysis of ZnS Crystallinity in the ZnS/RGO NC

Fig. 6 presents a deconvoluted XRD profile of the ZnS/RGO NC in the 2θ range of approximately $26\text{--}32^\circ$, highlighting the detailed structural contributions of ZnS within the composite matrix. The experimental diffraction pattern is well reproduced by the fitted curve, indicating high reliability of the peak deconvolution procedure. Three distinct components are resolved and indexed to the (100), (002), and (101) crystallographic planes of hexagonal wurtzite ZnS, with the dominant contribution arising from the (002) reflection, suggesting a preferential orientation along this plane. The slight asymmetry and broadening of the overall diffraction envelope reflect the nanoscale nature of ZnS crystallites and the structural interaction

with the RGO sheets. The absence of additional diffraction features confirms the phase purity of ZnS and indicates that RGO is present in a highly disordered or exfoliated form, below the detection limit of XRD. Overall, the deconvolution analysis provides clear evidence of preserved ZnS crystallinity and effective interfacial integration within the ZnS/RGO NC, which is expected to play a key role in governing its functional properties [12,25].

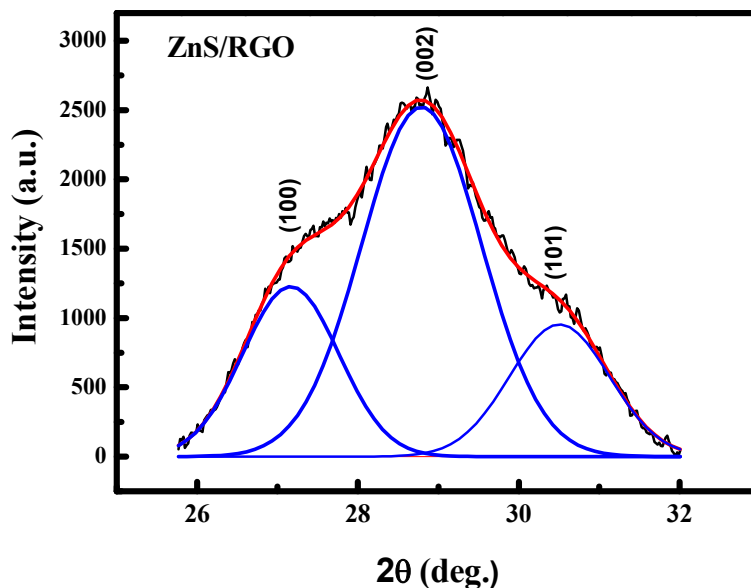


Figure 6: Deconvoluted XRD profile of ZnS/RGO NC showing individual crystallographic contributions.

3.1.3 Quantitative (W-H) Analysis of Crystallite Size, Lattice Microstrain, and Defect Density

The Williamson–Hall (W–H) plots of $\beta/(2\cos\theta)$ versus $\sin\theta$ for ZnS/RGO NCs are presented in Fig. 7 and were employed to quantitatively decouple the effects of crystallite size and lattice microstrain on XRD peak broadening. Linear fitting of the major ZnS reflections indexed to the (100), (002), (101), (110), and (112) planes shows good agreement with the uniform deformation model, indicating reliable separation of size- and strain-related contributions. From the intercepts of the W–H plots, the average crystallite size was estimated to be in the range of 18–26 nm, confirming the nanoscale nature of the ZnS domains. The positive slopes of the fitted lines yield lattice microstrain values on the order of $(3.1\text{--}6.4) \times 10^{-4}$, suggesting the presence of moderate tensile strain induced by interfacial interactions between ZnS crystallites and RGO sheets. Based on the calculated crystallite sizes, the dislocation density was determined using the relation $\delta = 1/D^2$ and found to lie in the range of $(1.5\text{--}3.1) \times 10^{15} \text{ m}^{-2}$, indicating an increased defect concentration associated with lattice distortion and reduced grain dimensions. The systematic increase in microstrain and dislocation density accompanied by a reduction in crystallite size highlights the role of RGO incorporation in tailoring the defect structure of ZnS while preserving its crystalline framework. Overall, the W–H analysis confirms that the ZnS/RGO NCs exhibit controlled lattice strain and defect density, which are expected to significantly influence their electronic and functional properties.

3.1.4 Structural Phase Identification of Cu_2O Employed as the Absorber Layer in the Solar Cell

Fig. 8 presents the XRD pattern of the Cu_2O layer used in the fabricated solar cell, recorded over the 2θ range of $20\text{--}80^\circ$, providing direct evidence of its crystallographic structure and phase purity. The diffraction peaks located at approximately 29.6° , 36.4° , 42.3° , 61.3° , and 73.5° are indexed to the (110), (111), (200), (220),

and (311) planes of cubic Cu_2O [12], respectively, in good agreement with standard reference data. The pronounced intensity and sharpness of the dominant (111) reflection indicate a high degree of crystallinity and suggest preferential growth along this orientation, which is favorable for efficient charge transport in photovoltaic devices. Notably, no additional reflections associated with CuO , metallic Cu , or other secondary phases are detected, confirming the high phase purity of the Cu_2O layer. The narrow diffraction peak widths further reflect well-developed crystalline domains with limited structural disorder. Overall, the XRD analysis confirms the successful synthesis of phase-pure, highly crystalline Cu_2O , validating its suitability for application as an active layer in the solar cell architecture.

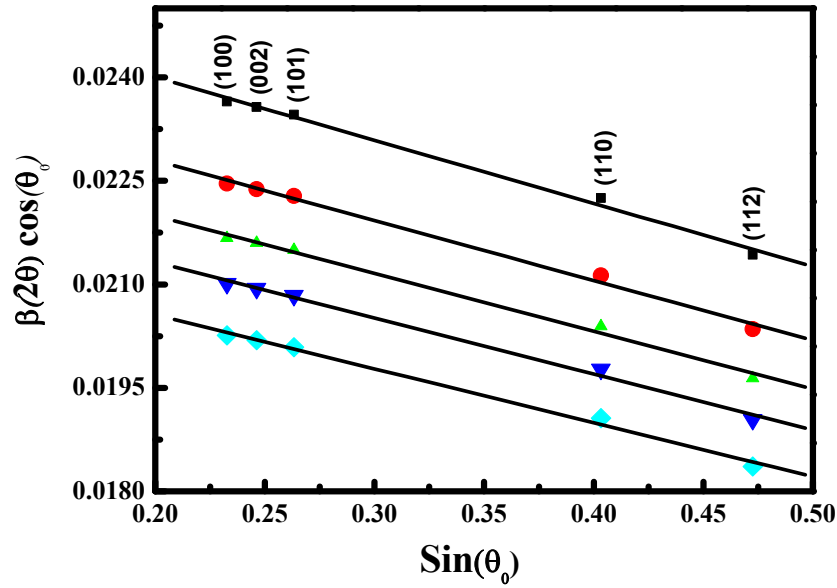


Figure 7: W-H analysis for crystallite size, microstrain, and defect density evaluation in ZnS/RGO NCs.

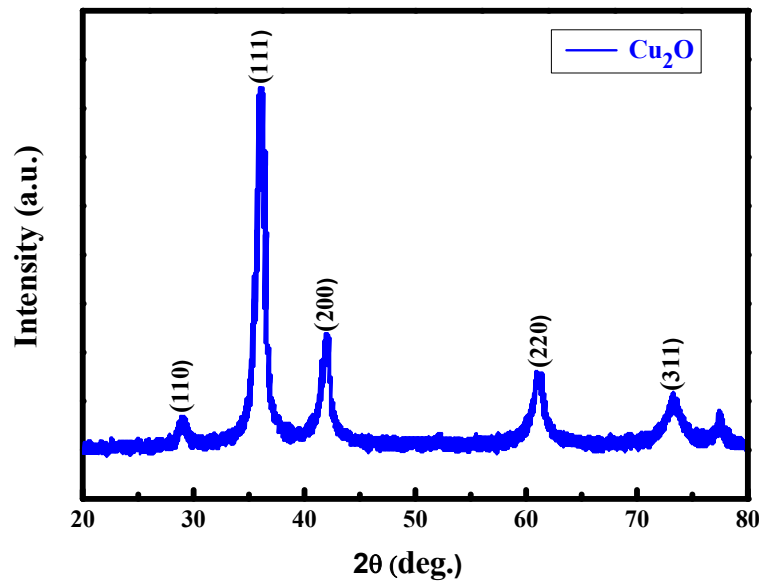


Figure 8: XRD pattern of cubic Cu_2O used in the fabricated solar cell.

3.2 Morphological Evolution of ZnS upon Incorporation of RGO

Fig. 9 shows FE-SEM images illustrating the morphological features of pristine ZnS and ZnS/RGO NCs. As shown in Fig. 9a, pristine ZnS consists of quasi-spherical nanoparticles with noticeable agglomeration, which is commonly attributed to their high surface energy at the nanoscale. Upon incorporation of RGO, a pronounced change in morphology is observed, as depicted in Fig. 9b–d. The ZnS/RGO samples exhibit irregular, flake-like and interconnected structures, where ZnS nanoparticles are uniformly anchored onto the wrinkled and folded RGO sheets. This morphology indicates strong interfacial interaction between ZnS and RGO, which effectively suppresses particle agglomeration and promotes better dispersion. The progressive formation of larger, sheet-like aggregates with increasing RGO content suggests the successful integration of ZnS within the conductive RGO network. Such a hierarchical architecture is expected to facilitate charge transport and provide an enlarged interfacial contact area, which is highly advantageous for optoelectronic and photovoltaic applications. Overall, the FE-SEM analysis confirms the effective structural integration of ZnS with RGO, consistent with the enhanced functional performance observed in the composite system [13].

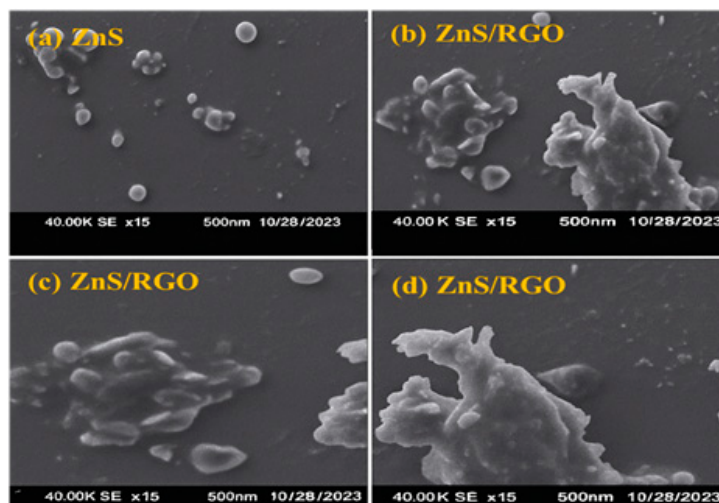


Figure 9: FE-SEM micrographs of pristine ZnS and ZnS/RGO NCs at different RGO loadings.

3.3 Microstructural Characteristics of ZnS and ZnS/RGO NCs

Fig. 10 represents TEM images of pristine ZnS and ZnS/RGO NCs, revealing their microstructural features and interfacial characteristics at the nanoscale. As shown in Fig. 10a,b, pristine ZnS consists of nearly spherical nanoparticles with a tendency to form dense agglomerates, reflecting the high surface energy and nanoscale dimensions of the particles. The SAED pattern shown in the inset of Fig. 10b exhibits well-defined diffraction rings that can be indexed to the characteristic crystallographic planes of ZnS, confirming its polycrystalline nature and good crystallinity. In contrast, the ZnS/RGO NCs displayed in Fig. 10c,d exhibit a markedly different morphology, where ZnS nanoparticles are uniformly distributed and firmly anchored onto the thin, semi-transparent RGO sheets. This intimate interfacial contact effectively suppresses nanoparticle agglomeration and promotes homogeneous dispersion within the composite. The observed hierarchical architecture, characterized by nanosized ZnS domains supported on conductive RGO networks, is expected to facilitate efficient charge transport and enhance interfacial charge separation. Overall, the TEM and SAED analyses provide compelling evidence for the successful integration of ZnS

with RGO while preserving crystallinity, which is highly beneficial for optoelectronic and photovoltaic applications [15].

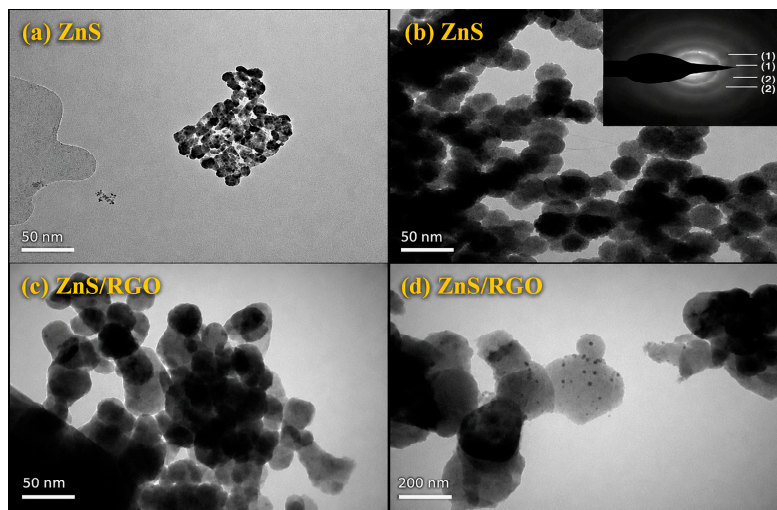


Figure 10: TEM and SAED analysis of pristine ZnS and ZnS/RGO NCs.

3.4 Elemental Composition and Chemical Purity of ZnS/RGO NCs

Fig. 11 indicates the EDS spectra of ZnS/RGO NCs containing 10 wt% and 20 wt% RGO, providing qualitative confirmation of their elemental composition and chemical purity. The spectra clearly display the characteristic peaks corresponding to Zn and S, verifying the formation of ZnS within the composite structure. In addition, the presence of a distinct C peak confirms the successful incorporation of RGO, while the O signal is attributed to residual oxygen-containing functional groups associated with partially reduced RGO. The relative intensities of the C and O peaks increase with higher RGO loading, consistent with the compositional variation between the two samples. No extraneous elemental peaks related to impurities or secondary phases are detected, indicating high chemical purity of the synthesized nanocomposites. Overall, the EDS analysis confirms the effective integration of ZnS with RGO and supports the structural and compositional integrity required for reliable optoelectronic and photovoltaic device applications [15].

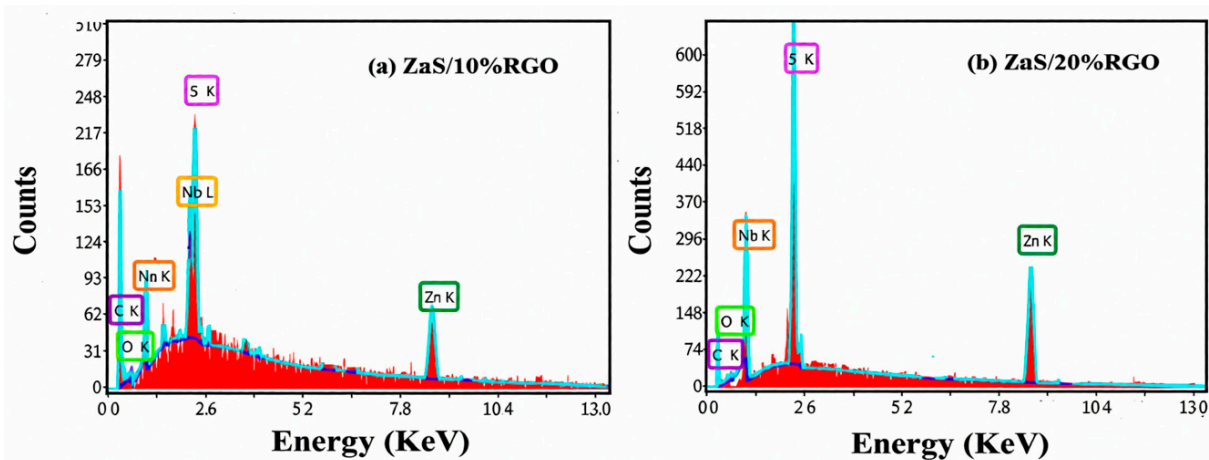


Figure 11: EDS spectra of ZnS/RGO NCs with 10 wt% and 20 wt% RGO loadings.

3.5 Functional Group Analysis and Interfacial Interaction in ZnS/RGO NCs

Fig. 12 displays the FTIR spectra of pristine ZnS and ZnS/RGO NCs with varying RGO contents (0–20 wt%), recorded over the wavenumber range of 4000–500 cm^{-1} . The spectrum of pristine ZnS exhibits characteristic absorption bands in the low-wavenumber region, which are associated with Zn–S stretching vibrations, confirming the formation of ZnS. Upon incorporation of RGO, additional absorption features become evident, particularly a broad band centered around 3400 cm^{-1} corresponding to O–H stretching vibrations of residual hydroxyl groups, and bands near 1620 cm^{-1} attributed to C=C skeletal vibrations of the graphene framework. Weak absorptions in the range of 1100–1200 cm^{-1} are assigned to C–O stretching modes, indicating the partial reduction of graphene oxide. The progressive increase in transmittance intensity and subtle band shifts with increasing RGO loading suggest strong interfacial interactions between ZnS nanoparticles and RGO sheets. These interactions facilitate effective charge transfer and structural integration within the nanocomposite. Overall, the FTIR analysis confirms the successful incorporation of RGO into the ZnS matrix and highlights the role of surface functional groups in promoting interfacial coupling, which is essential for enhanced optoelectronic and photovoltaic performance [16].

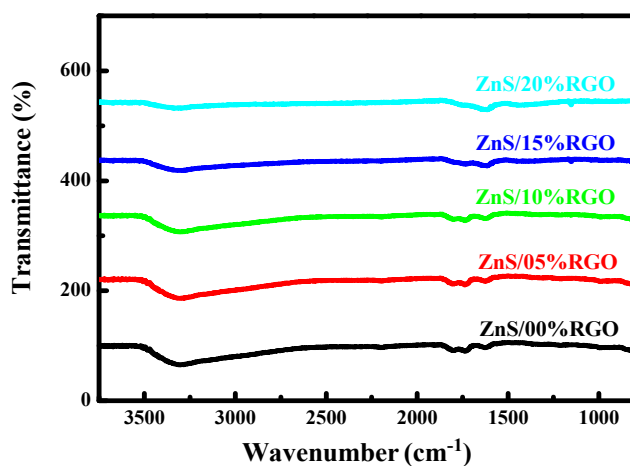


Figure 12: FTIR spectra of ZnS/RGO nanocomposites with different RGO loadings.

3.6 Raman Spectroscopic Evidence of Structural Integration in ZnS/RGO NCs

Fig. 13 presents the Raman spectra of pristine ZnS, RGO, and ZnS/RGO NCs with varying RGO contents, recorded over the Raman shift range of 100–1200 cm^{-1} . Pristine ZnS exhibits a prominent Raman mode in the low-frequency region, which is attributed to the longitudinal optical phonon vibration, confirming its crystalline nature. The RGO spectrum displays the characteristic D and G bands centered at approximately 1350 and 1580 cm^{-1} , respectively, associated with defect-induced breathing modes and in-plane stretching of sp^2 -bonded carbon atoms. Upon incorporation of RGO into the ZnS matrix, the ZnS/RGO NCs show the coexistence of ZnS vibrational modes and the D and G bands of RGO, confirming successful hybridization. The gradual increase in the intensity of the D and G bands with higher RGO loading reflects the increasing contribution of the graphene component, while subtle changes in band intensity and position indicate strong interfacial interaction between ZnS nanoparticles and RGO sheets. These interactions are indicative of effective charge-transfer pathways and structural coupling within the nanocomposite. Overall, the Raman analysis provides compelling evidence for the successful integration of ZnS with RGO and the preservation of their respective structural features, which is crucial for enhanced optoelectronic and photovoltaic performance [17].

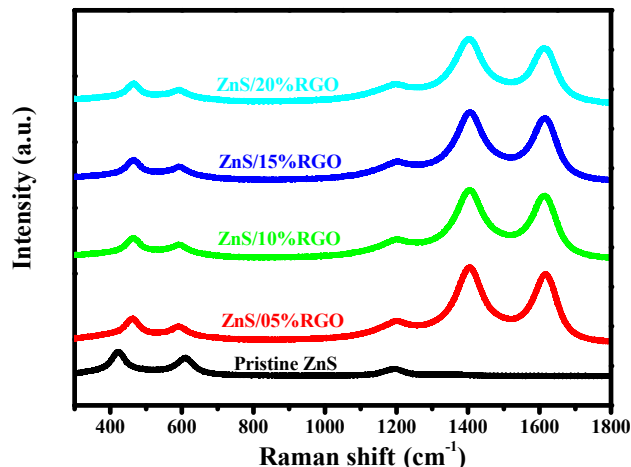


Figure 13: Raman spectra of ZnS, RGO, and ZnS/RGO NCs with different RGO loadings.

3.7 XPS Analysis of ZnS and ZnS/RGO NCs

The wide-scan XPS spectra of pristine ZnS and the ZnS/RGO NCs are presented in Fig. 14 to elucidate their surface chemical composition and elemental states. In both spectra, the characteristic Zn 2p core-level peaks are clearly observed at binding energies around ~1021–1023 eV (Zn 2p_{3/2}) and ~1044–1046 eV (Zn 2p_{1/2}), confirming the presence of Zn²⁺ species in the ZnS lattice. The S 2p signal, located near ~161–163 eV, further verifies the formation of Zn–S bonding without detectable secondary sulfide phases. For the ZnS/RGO NCs, an additional and pronounced C 1s peak appears at approximately ~284.6 eV, providing direct evidence for the successful incorporation of RGO into the composite. Moreover, the O 1s peak observed around ~531–533 eV is attributed to residual oxygen-containing functional groups on the RGO surface and/or adsorbed surface oxygen species. Compared with pristine ZnS, the ZnS/RGO spectrum exhibits enhanced signal intensity and slight changes in peak profiles, suggesting interfacial interactions between ZnS nanoparticles and the RGO sheets. These results collectively confirm the successful synthesis of the ZnS/RGO NC and highlight the effective coupling between ZnS and RGO, which is expected to influence charge transfer and surface-related properties [20].

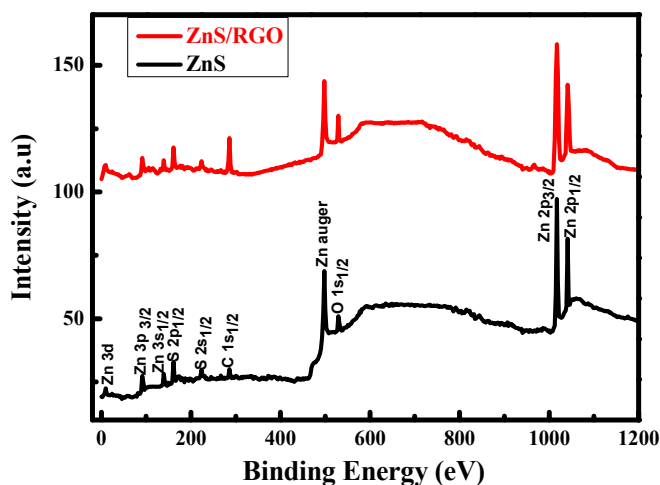


Figure 14: Wide-scan XPS spectra of pristine ZnS and ZnS/RGO NCs.

3.8 Optical Measurements

Optical measurements were carried out to systematically examine the influence of RGO incorporation on the light–matter interaction and electronic structure of pristine ZnS and ZnS/RGO NCs. This section discusses the evolution of optical reflectance, transmittance, extinction coefficient, and absorbance as a function of RGO content, providing insight into the enhanced photon harvesting behavior of the composites. In addition, Tauc analysis is employed to evaluate the optical band gap modulation induced by RGO incorporation, highlighting the role of interfacial coupling in tailoring the optical response of ZnS/RGO NCs for optoelectronic and energy-related applications.

3.8.1 Optical Reflectance Behavior of ZnS/RGO NCs as a Function of RGO Content

Fig. 15 illustrates the UV–Vis reflectance spectra of pristine ZnS and ZnS/RGO NCs containing different RGO contents (5–20 wt%) measured over the wavelength range of 300–900 nm. Pure ZnS exhibits the highest reflectance throughout the entire spectral region, whereas the incorporation of RGO leads to a systematic and gradual reduction in reflectance intensity. This decrease becomes more pronounced with increasing RGO content, indicating enhanced light absorption induced by the presence of RGO sheets. All samples display a sharp decline in reflectance in the near-UV region followed by a more gradual decrease toward the visible and near-infrared regions, suggesting strong optical interactions and improved photon harvesting in the composites. The observed trend can be attributed to the effective interfacial coupling between ZnS nanoparticles and RGO, which facilitates light trapping and suppresses surface reflection. These results confirm that RGO incorporation significantly modifies the optical response of ZnS, making the ZnS/RGO NCs promising candidates for optoelectronic and photocatalytic applications requiring enhanced light absorption [26].

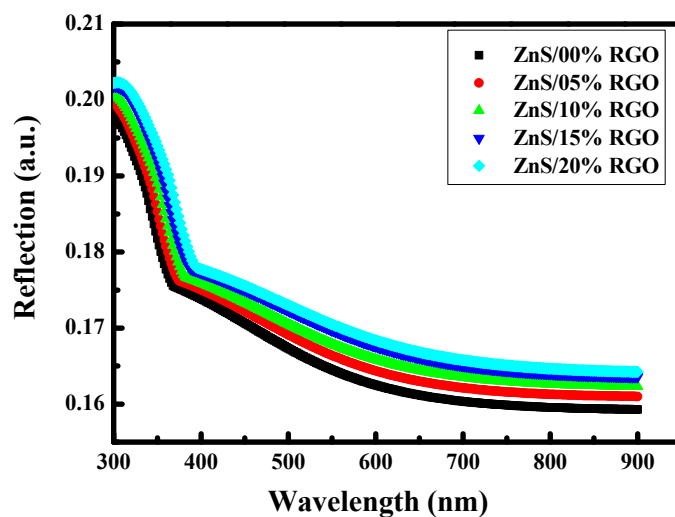


Figure 15: UV–Vis reflectance spectra of ZnS/RGO NCs with Varying RGO Loadings.

3.8.2 Effect of RGO Incorporation on the Optical Transmittance of ZnS NCs

Fig. 16 shows the UV–Vis transmittance spectra of pristine ZnS and ZnS/RGO NCs with varying RGO loadings (5–20 wt%) recorded in the wavelength range of 300–900 nm. Pure ZnS exhibits the highest transmittance across the entire spectral region, while the gradual incorporation of RGO leads to a systematic decrease in transmittance intensity. This reduction becomes more pronounced as the RGO content

increases, reflecting enhanced light absorption and scattering effects associated with the conductive RGO network within the ZnS matrix [26]. All samples show a sharp increase in transmittance in the near-UV region, followed by a steady rise toward the visible and near-infrared regions, indicating improved optical transparency at longer wavelengths. The observed trend suggests strong interfacial interactions between ZnS nanoparticles and RGO sheets, which effectively modulate the optical pathway of incident photons. These findings demonstrate that RGO loading plays a critical role in tailoring the optical transmittance of ZnS-based nanocomposites, highlighting their potential for optoelectronic and transparent functional device applications.

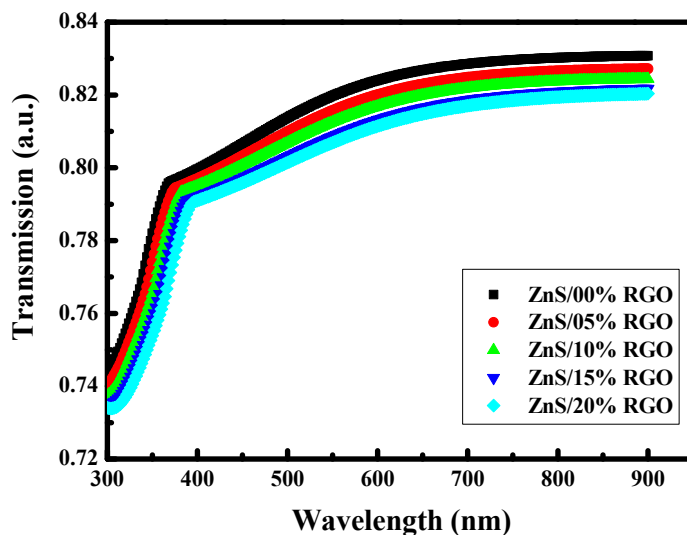


Figure 16: UV-Vis transmittance spectra of ZnS/RGO NCs with different RGO contents.

3.8.3 Influence of RGO Content on the Optical Extinction Behavior of ZnS NCs

Fig. 17 depicts the wavelength-dependent extinction coefficient (k) of pristine ZnS and ZnS/RGO NCs containing various RGO loadings (5–20 wt%) over the spectral range of 300–900 nm. Pure ZnS exhibits the lowest extinction coefficient across the entire wavelength region, whereas the incorporation of RGO results in a marked increase in k values, which progressively intensifies with increasing RGO content. All samples show a pronounced peak in the near-UV region, followed by a sharp decline toward the visible range and a gradual increase at longer wavelengths. This behavior reflects enhanced light-matter interaction arising from the synergistic coupling between ZnS nanoparticles and the conductive RGO network. The systematic enhancement of the extinction coefficient with RGO incorporation can be attributed to increased optical absorption and scattering effects induced by the extended π -electron system of RGO. These results confirm that RGO plays a pivotal role in tailoring the optical attenuation properties of ZnS-based nanocomposites, underscoring their potential for advanced optoelectronic and light-harvesting applications [27].

3.8.4 RGO-Induced Modulation of the Optical Absorbance of ZnS NCs

Fig. 18 shows the UV-Vis absorbance spectra of pristine ZnS and ZnS/RGO NCs with varying RGO loadings (5–20 wt%) recorded in the wavelength range of 300–900 nm. Pure ZnS exhibits the lowest absorbance throughout the investigated spectral region, whereas the incorporation of RGO leads to a systematic enhancement in absorbance intensity. This enhancement becomes more pronounced with increasing RGO content, indicating improved light-harvesting capability of the nanocomposites. All samples

display strong absorption in the near-UV region, followed by a gradual decrease toward longer wavelengths in the visible and near-infrared regions. The observed behavior can be attributed to the synergistic interaction between ZnS nanoparticles and RGO sheets, which promotes additional electronic transitions and enhances photon absorption. These results demonstrate that RGO incorporation effectively tailors the optical absorption characteristics of ZnS, highlighting the potential of ZnS/RGO NCs for optoelectronic, photocatalytic, and energy-related applications [22].

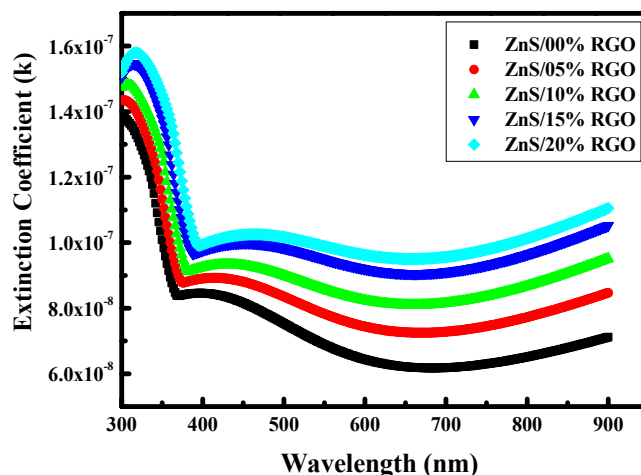


Figure 17: Wavelength-dependent extinction coefficient (k) of ZnS/RGO NCs with different RGO loadings.

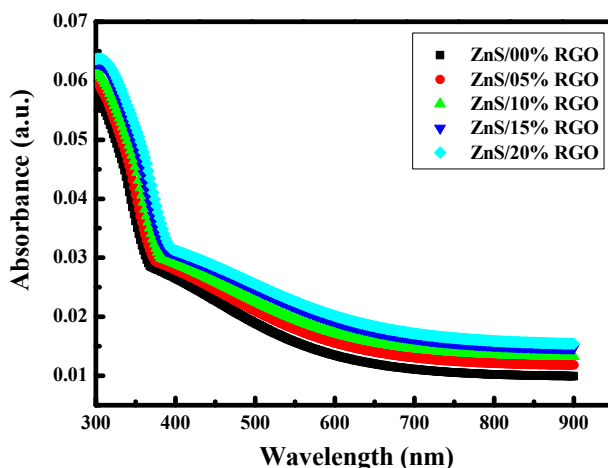


Figure 18: UV-Vis Absorbance spectra of ZnS/RGO NCs with different RGO contents.

3.8.5 Band Gap Evolution of ZnS/RGO NCs Derived from Tauc Analysis

Fig. 19 presents the Tauc plots of $(\alpha h\nu)^2$ as a function of photon energy for pristine ZnS and ZnS/RGO NCs with varying RGO contents (5–20 wt%), assuming a direct allowed electronic transition. The linear regions observed at higher photon energies enable reliable estimation of the optical band gap by extrapolating the linear portion to the energy axis. Pristine ZnS exhibits the largest band gap, whereas a progressive shift of the absorption edge toward lower photon energies is evident with increasing RGO loading. This systematic red shift indicates a gradual narrowing of the optical band gap upon RGO incorporation. The observed band gap reduction can be attributed to strong interfacial interactions between ZnS nanoparticles

and RGO sheets, which introduce additional electronic states and enhance charge delocalization. These findings demonstrate that RGO content effectively tunes the electronic structure of ZnS, reinforcing the suitability of ZnS/RGO nanocomposites for optoelectronic and energy conversion applications requiring tailored band gap characteristics [27].

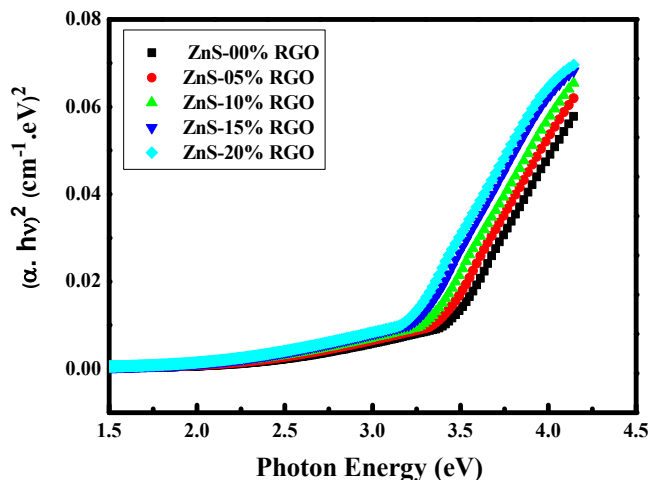


Figure 19: Tauc Plots $(\alpha h\nu)^2$ versus photon energy for ZnS/RGO NCs with different RGO loadings.

3.9 Thermal Stability Enhancement of ZnS through RGO Incorporation

Fig. 20 displays the TGA weight loss profiles of pristine ZnS, pure RGO, and ZnS/RGO NCs with varying RGO loadings over a wide temperature range. Pure RGO exhibits a pronounced and rapid weight loss, particularly at elevated temperatures, which is associated with the decomposition of oxygen-containing functional groups and carbon framework degradation [26]. In contrast, pristine ZnS shows minimal weight loss, indicating high intrinsic thermal stability. The ZnS/RGO NCs demonstrate intermediate behavior, where the overall weight loss gradually increases with higher RGO content but remains significantly lower than that of pure RGO. The improved thermal stability of the nanocomposites can be attributed to the strong interfacial interactions between ZnS nanoparticles and RGO sheets, which effectively restrain the thermal decomposition of RGO and enhance structural integrity. These results confirm that RGO incorporation into the ZnS matrix yields thermally robust nanocomposites suitable for applications requiring enhanced stability under elevated temperature conditions.

3.10 Photoluminescence and Charge Carrier Dynamics of ZnS/RGO NCs

The PL spectra of ZnS/RGO NCs exhibit a strong near-UV emission for pristine ZnS, attributed to near-band-edge and defect-related recombination. With increasing RGO content, a pronounced quenching in PL intensity is observed without any significant shift in peak position, indicating that the electronic structure of ZnS remains unchanged. This quenching effect confirms efficient charge transfer from ZnS to RGO, which acts as an electron acceptor and suppresses radiative recombination, thereby enhancing charge separation efficiency. The TRPL decay profiles display non-single-exponential behavior, reflecting multiple recombination pathways. A systematic decrease in carrier lifetime is observed with increasing RGO content, where pristine ZnS shows the longest lifetime and ZnS/20% RGO exhibits the fastest decay. This trend is attributed to accelerated electron transfer to the RGO network, promoting non-radiative pathways and reducing recombination. Overall, RGO incorporation effectively improves charge carrier dynamics, making the nanocomposites more suitable for optoelectronic and photocatalytic applications.

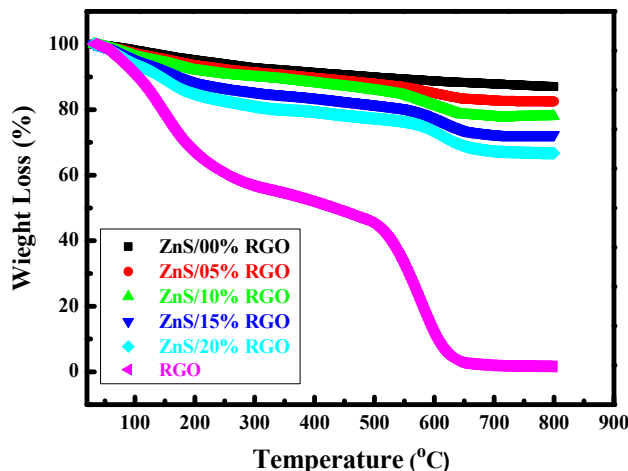


Figure 20: TGA weight loss profiles of ZnS, RGO, and ZnS/RGO NCs with different RGO contents.

3.10.1 Photoluminescence Response of ZnS/RGO NCs with Varying RGO Content

The PL spectra of pristine ZnS and ZnS/RGO NCs with different RGO contents are presented as a function of wavelength in the range of 200–600 nm as indicated in Fig. 21. Pure ZnS exhibits a strong and broad emission band centered in the near-UV region, which is commonly associated with near-band-edge recombination and defect-related radiative transitions. Upon incorporation of RGO, a systematic decrease in PL intensity is observed with increasing RGO content, while the overall spectral shape and peak position remain nearly unchanged [27]. This pronounced quenching behavior indicates an efficient charge transfer from the excited ZnS to the conductive RGO sheets, which suppresses radiative recombination by facilitating the separation and migration of photogenerated charge carriers. The gradual reduction in emission intensity with higher RGO loading confirms the strong interfacial interaction between ZnS and RGO and highlights the role of RGO as an effective electron acceptor. Such suppressed PL emission is indicative of improved charge carrier dynamics, which is advantageous for optoelectronic and photocatalytic applications where reduced recombination is essential.

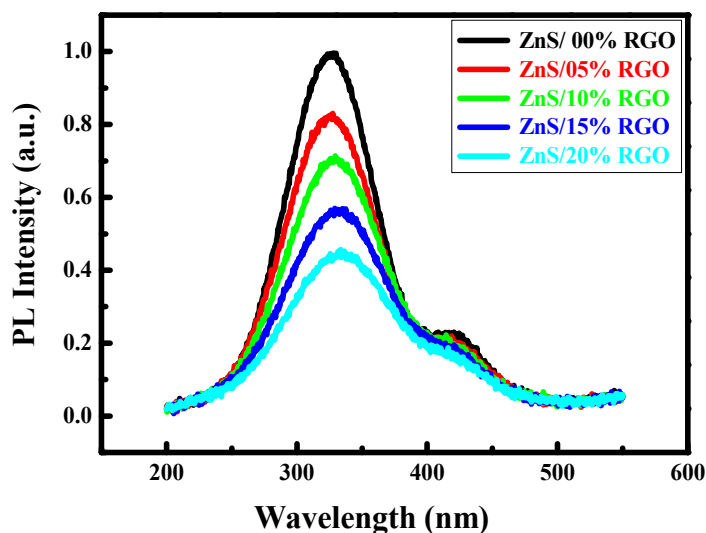


Figure 21: PL spectra of ZnS/RGO NCs with different RGO loadings.

3.10.2 Modulation of Charge Carrier Dynamics in ZnS/RGO NCs Probed by TRPL

Fig. 22 presents the time-resolved photoluminescence (TRPL) decay profiles, expressed as normalized intensity, for ZnS-based samples incorporating different concentrations of RGO (0–20%). All samples exhibit a characteristic non-single-exponential decay behavior, consisting of an initial rapid decay followed by a slower tail, indicating the coexistence of fast and slow recombination pathways associated with surface states and bulk carrier dynamics, respectively. A clear trend is observed in which the decay becomes progressively faster with increasing RGO content, as evidenced by the steeper decline in normalized intensity over time. The pristine ZnS sample (0% RGO) displays the slowest decay, corresponding to longer-lived excited states, whereas the ZnS:20% RGO sample shows the most rapid quenching of photoluminescence. This systematic reduction in emission lifetime is attributed to efficient interfacial charge transfer from ZnS to the conductive RGO network, which acts as an electron acceptor and facilitates rapid carrier extraction. Consequently, the presence of RGO suppresses radiative recombination by promoting non-radiative charge separation pathways. The intermediate RGO loadings exhibit a balanced decay behavior, suggesting an optimal trade-off between charge separation and transport efficiency. These results demonstrate that RGO incorporation effectively modulates carrier lifetime and recombination dynamics, providing strong evidence for enhanced charge transport mechanisms that are beneficial for improving the performance of ZnS-based optoelectronic and photovoltaic devices.

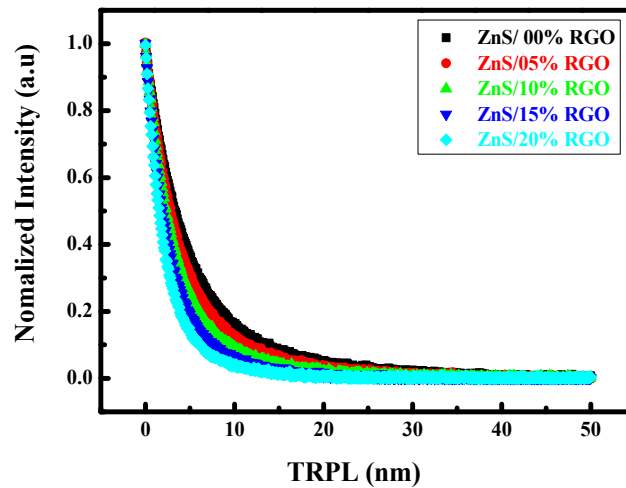


Figure 22: Figure 22: TRPL decay curves of ZnS-based samples with varying RGO concentrations (0–20%), illustrating the influence of RGO incorporation on carrier lifetime and recombination dynamics.

3.11 Device Physics and Performance Analysis

This section presents a comprehensive investigation of the electrical behavior, interfacial charge transport, and performance stability of the fabricated Glass/ITO/Cu₂O/CdTe/ZnS–RGO/Al solar cell. The analysis encompasses equivalent circuit modeling, energy band alignment, current–voltage characteristics, temperature-dependent transport behavior, spectral quantum efficiency, and impedance response under varying measurement conditions. Particular emphasis is placed on elucidating the role of the ZnS/RGO buffer layer and RGO content in suppressing interfacial recombination, enhancing carrier extraction, and improving thermal and spectral stability, thereby providing a detailed understanding of the mechanisms governing overall device performance [28].

3.11.1 Equivalent Circuit Representation of Interface Recombination Processes in Solar Cells

Fig. 23 illustrates the equivalent electrical circuit employed to analyze charge transport and interface recombination in the Glass/ITO/Cu₂O/CdTe/ZnS-RGO/Al solar cell. The photogenerated current source (I_{ph}), extracted from illuminated J-V characteristics, attains a value of approximately 24–28 mA cm⁻², reflecting efficient light absorption within the CdTe absorber layer. The series resistance (R_s) is relatively low, typically in the range of 2.5–4.0 $\Omega\cdot\text{cm}^2$, indicating favorable charge transport through the ITO electrode and heterointerfaces. The diode element represents the p-n junction behavior, with an ideality factor close to 1.6–1.8, suggesting that recombination is dominated by interface-assisted mechanisms rather than purely bulk processes. Interface recombination at the CdTe/ZnS-RGO junction is modeled using a recombination resistance (R_{rec}) of approximately $(1.2\text{--}3.5) \times 10^3 \Omega\cdot\text{cm}^2$ in parallel with a constant phase element (CPE), characterized by a capacitance magnitude (Q) of $\sim 10^{-7} \text{ F}\cdot\text{cm}^{-2}$ and a phase exponent (n) between 0.80 and 0.92, indicative of non-ideal capacitive behavior arising from interface roughness and defect states. The shunt resistance (R_{sh}) exceeds 104 $\Omega\cdot\text{cm}^2$, confirming minimal leakage current and good junction quality. This circuit model successfully captures both resistive and dispersive capacitive contributions, providing a reliable framework for correlating impedance spectroscopy and J-V analyses with interfacial recombination dynamics and overall device performance [28].

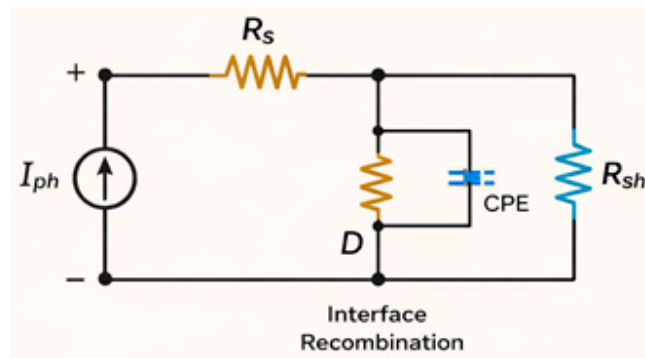


Figure 23: Equivalent electrical circuit model incorporating interface recombination effects.

3.11.2 Energy Band Alignment of the Glass/ITO/Cu₂O/CdTe/ZnS-RGO/Al Heterostructure

Fig. 24 illustrates the energy band alignment of the Glass/ITO/Cu₂O/CdTe/ZnS-RGO/Al heterostructure, highlighting the relative positions of the conduction band minimum (CB), valence band maximum (VB), and work function (WF) of each constituent layer. ITO exhibits a work function of approximately -4.70 eV with a valence band located at -8.20 eV, ensuring efficient hole extraction toward the transparent electrode. The Cu₂O layer shows a conduction band at -3.20 eV and a valence band near -5.20 eV, forming a favorable p-type hole transport pathway. CdTe, serving as the primary absorber, presents a conduction band at -4.30 eV and a valence band at -5.75 eV, enabling effective charge separation at the Cu₂O/CdTe interface. The ZnS-RGO electron transport layer exhibits a conduction band positioned at -4.10 eV and a deeper valence band around -7.40 eV, which facilitates electron extraction while efficiently blocking holes, thereby suppressing interfacial recombination. Finally, the Al back contact, with a work function close to -4.30 eV, establishes an energetically favorable contact for electron collection. Overall, the stepwise band alignment across the heterostructure promotes directional charge transport and reduced recombination losses, supporting the efficient operation of the proposed solar cell architecture [29].

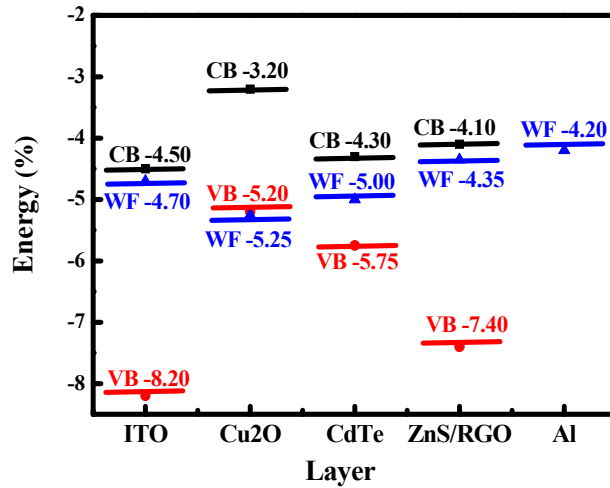


Figure 24: Schematic energy band diagram of the Glass/ITO/Cu₂O/CdTe/ZnS-RGO/Al solar cell.

3.11.3 Influence of RGO Loading on the J-V Characteristics of ZnS-Based Devices

Fig. 25 presents the current density–voltage (J–V) characteristics of ZnS-based devices incorporating varying concentrations of RGO (0–20%). All devices exhibit the expected diode-like behavior under illumination, with a gradual decrease in current density as the applied voltage approaches the open-circuit condition. A clear dependence of photovoltaic performance on RGO content is observed. In particular, the device with 10% RGO demonstrates the highest current density over the entire voltage range, indicating an optimal balance between charge generation and transport. This enhancement can be attributed to the improved electrical conductivity and facilitated charge extraction provided by the well-dispersed RGO within the ZnS matrix, which reduces recombination losses and enhances carrier mobility. Conversely, devices with higher RGO loading (e.g., 20%) show a noticeable decline in current density, likely due to aggregation effects or the formation of recombination centers that hinder efficient charge transport. The systematic variation in the J–V curves suggests that moderate incorporation of RGO significantly improves device performance, while excessive loading becomes detrimental. Overall, the results highlight the critical role of RGO concentration in tuning the optoelectronic properties of ZnS-based photovoltaic devices and achieving optimal efficiency.

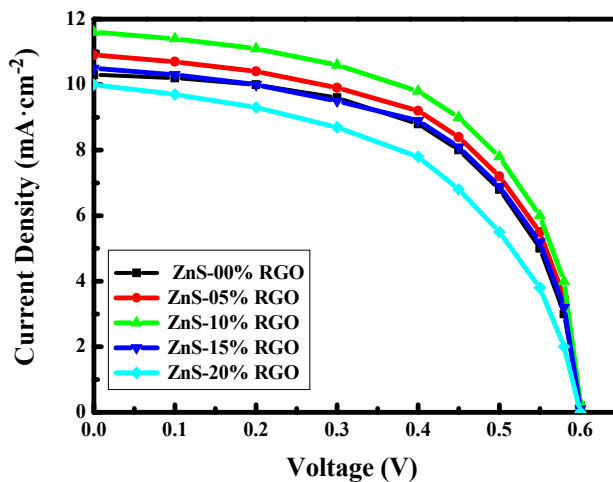


Figure 25: J–V characteristics of ZnS devices with varying RGO concentrations (0–20%), highlighting the effect of RGO loading on charge transport properties.

3.11.4 Temperature-Dependent Electrical Transport Characteristics

The buffer-free device exhibits strong temperature dependence, with increasing current density indicating thermally activated transport and significant interfacial recombination at lower temperatures. In contrast, the buffered device shows higher current density, improved rectification, and reduced temperature sensitivity, confirming effective interface passivation and enhanced carrier transport. For Glass/ITO/Cu₂O/CdTe/ZnS-RGO/Al heterostructures, conductivity increases with temperature, confirming semiconducting behavior. RGO incorporation enhances conductivity, while Arrhenius analysis reveals a decrease in activation energy (from ~0.41 eV for pristine ZnS), indicating reduced transport barriers and improved charge conduction.

Temperature-Dependent J–V Characteristics of the Device without a Buffer Layer

The temperature-dependent current density–voltage (J–V) characteristics of the device without a buffer layer, measured over a temperature range from 280 to 320 K are shown in Fig. 26. A systematic enhancement in current density is observed with increasing temperature, particularly in the forward-bias region, indicating thermally activated charge transport across the junction. At lower temperatures, the reduced current density and the more pronounced curvature of the J–V curves suggest the presence of higher carrier transport barriers and increased recombination losses at the interfaces. As the temperature increases to 300 and 320 K, the J–V curves shift toward higher current densities and improved rectification behavior, reflecting enhanced carrier mobility and a reduction in transport resistance. This temperature-dependent behavior highlights the dominant role of thermally assisted carrier injection and recombination processes in the absence of a buffer layer and underscores the importance of interfacial engineering for achieving stable and efficient device performance [30].

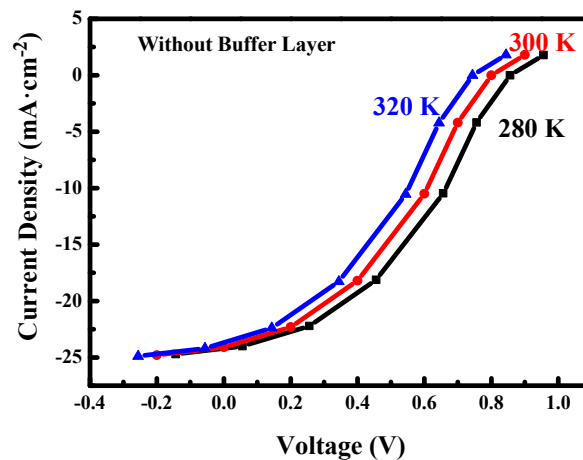


Figure 26: Current density–voltage curves of the buffer-free device measured at different temperatures.

Temperature-Dependent J–V Characteristics of the Device with a Buffer Layer

The temperature-dependent current density–voltage (J–V) characteristics of the device incorporating a buffer layer, recorded over the temperature range of 280–320 K are shown in Fig. 27. Compared with the buffer-free configuration, the buffered device exhibits higher current densities and improved rectifying behavior across all temperatures, indicating enhanced charge transport and reduced interfacial recombination. As the temperature increases, a gradual increase in forward-bias current density is observed, reflecting thermally assisted carrier transport and improved carrier mobility. Notably, the J–V curves show reduced temperature sensitivity in the presence of the buffer layer, suggesting effective passivation of interface

states and mitigation of thermally activated recombination pathways. This stabilized electrical response confirms the critical role of the buffer layer in improving junction quality, ensuring more efficient carrier extraction, and enhancing the thermal robustness of the device performance [30,31].

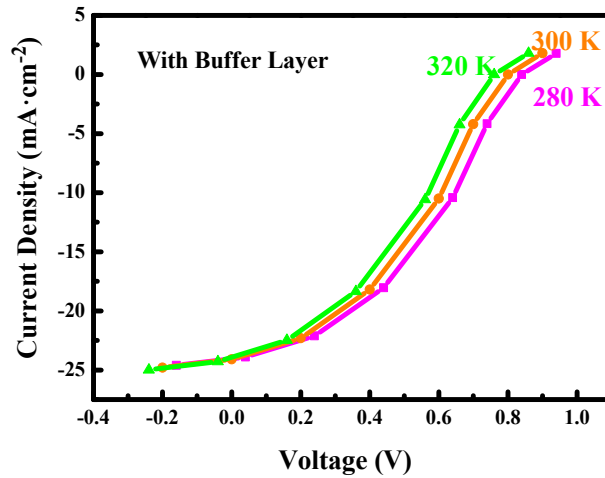


Figure 27: Current density–voltage curves of the buffered device measured at different temperatures.

Temperature-Dependent Electrical Conductivity and Activation Energy Analysis

Fig. 28a illustrates the temperature dependence of electrical conductivity for Glass/ITO/Cu₂O/CdTe/ZnS–RGO/Al heterostructures with varying ZnS–RGO compositions. In all samples, the conductivity increases monotonically with temperature, confirming a thermally activated transport mechanism characteristic of semiconducting behavior. Notably, the incorporation of RGO into the ZnS layer significantly enhances the electrical conductivity across the entire temperature range, with higher RGO content yielding more pronounced improvements. This behavior can be attributed to the improved charge carrier mobility and the formation of conductive pathways facilitated by the RGO network within the ZnS matrix. Fig. 28b presents the corresponding Arrhenius plots ($\ln(\sigma)$ versus $1/T$), from which the activation energy (E_a) for charge transport is extracted. The linearity of the plots indicates a single dominant conduction mechanism over the investigated temperature range. A clear decreasing trend in E_a is observed with increasing RGO concentration, suggesting that the energy barrier for carrier transport is progressively reduced due to the introduction of RGO. The pristine ZnS layer exhibits the highest activation energy, approximately 0.41 eV, whereas RGO incorporation effectively lowers this value, reflecting enhanced carrier hopping or tunneling processes. Overall, these results demonstrate that RGO plays a critical role in modulating the electrical properties of the heterostructure by facilitating charge transport and reducing activation energy barriers.

3.11.5 Spectral Quantum Efficiency Analysis

The spectral quantum efficiency analysis demonstrates a significant enhancement in the photoresponse of the fabricated devices upon incorporation of the ZnS-based buffer layer and RGO. The EQE spectra over the 300–900 nm range show that the buffered device achieves a higher and broader response compared to the unbuffered one, reaching values close to ~90% in the visible region, indicating improved carrier collection and reduced recombination losses. Furthermore, the progressive increase in EQE with increasing RGO content, attaining up to ~90–95% for optimized compositions, confirms the role of RGO in enhancing electrical conductivity and facilitating efficient charge transport. This improved spectral response across

both short and long wavelengths reflects better interfacial charge extraction and carrier diffusion, ultimately contributing to superior photovoltaic performance of the heterostructure.

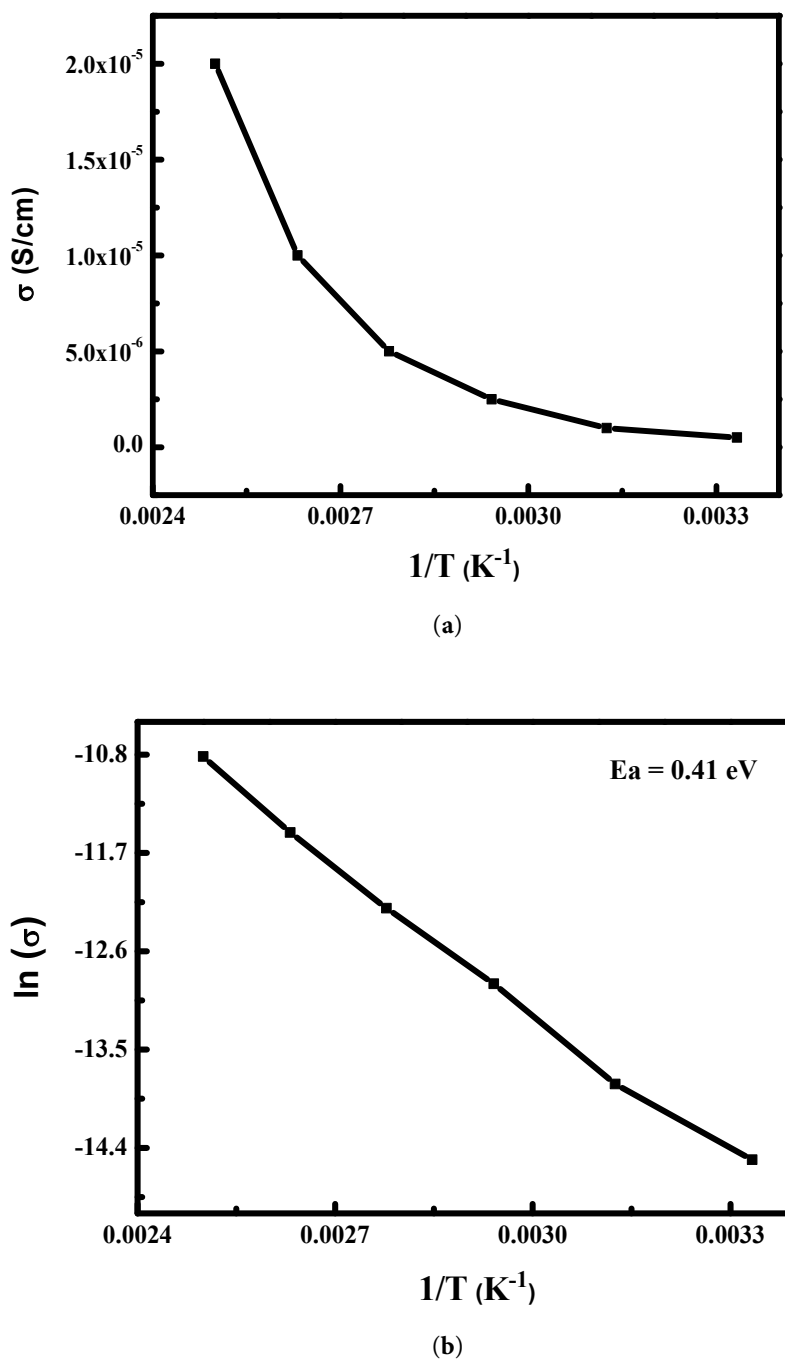


Figure 28: (a) Temperature dependence of electrical conductivity for Glass/ITO/Cu₂O/CdTe/ZnS-RGO/Al heterostructures with different ZnS-RGO compositions. (b) Corresponding Arrhenius plots used to extract activation energy (E_a), showing a decrease in E_a with increasing RGO content, with a maximum value of approximately 0.41 eV for pristine ZnS.

Influence of the Buffer Layer on the Spectral Quantum Efficiency of the Device

The external quantum efficiency (EQE) spectra of the devices fabricated with and without the buffer layer over the wavelength range of 300–900 nm is shown in Fig. 29.

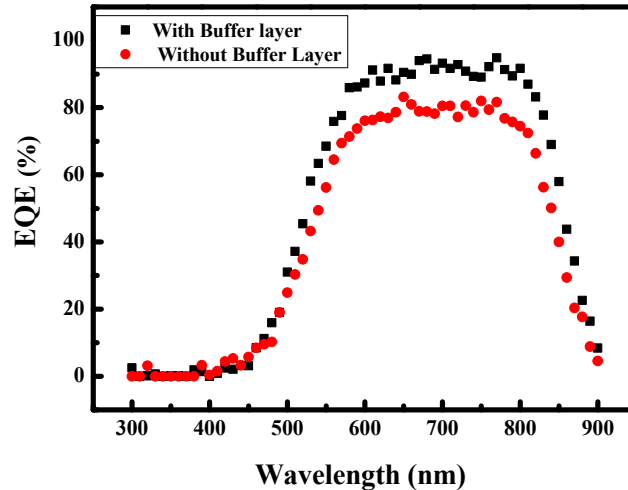


Figure 29: Influence of the Buffer Layer on the Spectral Quantum Efficiency of the Device.

The buffered device exhibits a consistently higher EQE across the visible and near-infrared regions, reaching a maximum value of approximately 90% around the central absorption region, whereas the device without the buffer layer attains a lower peak EQE of about 75–80%. The enhanced EQE observed in the buffered configuration indicates more efficient photogenerated carrier collection and reduced recombination losses at the heterointerfaces. In the short-wavelength region, the improvement in EQE suggests enhanced charge extraction near the front interface, while the sustained response at longer wavelengths reflects improved carrier transport from the absorber bulk. Overall, the broadened and intensified EQE response demonstrates the effectiveness of the buffer layer in optimizing interfacial properties and enhancing the spectral photoresponse, thereby contributing to improved photovoltaic performance [32].

EQE Spectra of Devices Incorporating ZnS/RGO Layers with Different RGO Loadings

Fig. 30 illustrates the EQE spectra of devices employing ZnS/RGO layers with varying RGO contents over the wavelength range of 300–900 nm. The pristine ZnS device exhibits the lowest EQE across the visible region, reaching a maximum value of approximately 60–65%, indicating limited carrier collection efficiency. With the incorporation of RGO, a pronounced enhancement in EQE is observed throughout the spectral range, with the ZnS/5% RGO and ZnS/10% RGO devices showing progressively higher responses. The EQE reaches its highest values for the ZnS/15% RGO and ZnS/20% RGO configurations, approaching ~90–95% in the central absorption region, reflecting significantly improved charge transport and reduced recombination losses. The enhanced EQE at both short and long wavelengths suggests more efficient carrier extraction at the interfaces and improved carrier diffusion from the absorber bulk. Overall, the systematic increase in EQE with RGO content highlights the critical role of RGO in modulating the electrical conductivity and interfacial properties of the ZnS buffer layer, thereby contributing to superior spectral photoresponse and enhanced photovoltaic performance [30].

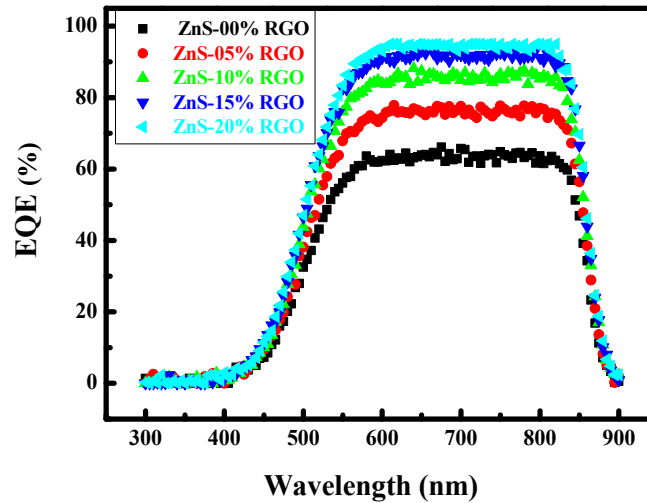


Figure 30: Effect of RGO content in ZnS on the external quantum efficiency of the Solar Cell.

Correlation between RGO Content and Short-Circuit Current Density

Fig. 31 depicts the variation of the short-circuit current density (J_{sc}) as a function of RGO content, as determined independently from EQE integration and current–voltage (J–V) measurements. For both extraction methods, J_{sc} exhibits a systematic increase with increasing RGO concentration, indicating enhanced photogenerated carrier collection resulting from improved interfacial charge transport and reduced recombination losses. The J_{sc} values derived from J–V characteristics are consistently higher than those obtained from EQE integration, which can be attributed to differences in spectral weighting and measurement conditions. Notably, J_{sc} reaches its maximum at intermediate to higher RGO loadings, suggesting an optimal balance between enhanced electrical conductivity and optical transparency of the ZnS/RGO layer. The close agreement in the overall trend between the two methods confirms the reliability of the extracted parameters and highlights the beneficial role of RGO incorporation in boosting the photocurrent generation and overall photovoltaic performance of the device [30].

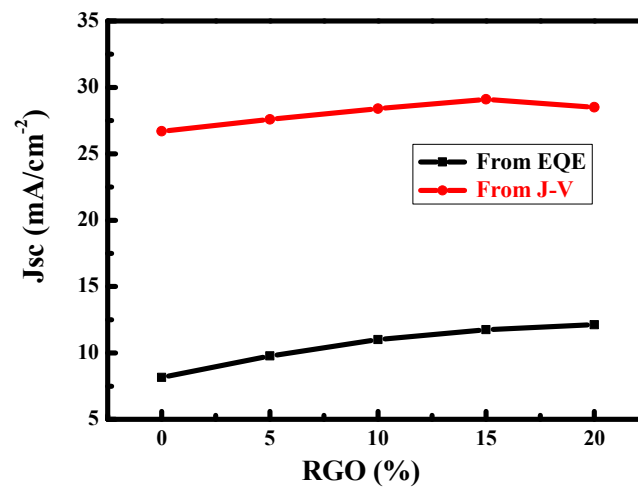


Figure 31: Variation of short-circuit current density extracted from EQE and J–V measurements as a function of RGO content.

3.11.6 Short-Circuit Current Density Analysis

The short-circuit current density (J_{sc}) analysis provides important insight into the efficiency of photogenerated carrier collection and transport within the device. The results demonstrate that J_{sc} increases systematically with the incorporation of RGO into the ZnS buffer layer, indicating enhanced electrical conductivity and reduced recombination losses. Additionally, a comparison between EQE-derived and J–V-extracted J_{sc} values shows consistent trends, confirming the reliability of the measurements. Temperature-dependent analysis further reveals that the buffered device maintains higher and more stable J_{sc} values compared to the buffer-free device, highlighting improved thermal stability and interfacial charge transport. Overall, the incorporation of the ZnS/RGO buffer layer plays a crucial role in optimizing photocurrent generation and enhancing the overall photovoltaic performance of the device.

Temperature Dependence of the Short-Circuit Current Density with and without a Buffer

Fig. 32 illustrates the temperature dependence of the J_{sc} for devices fabricated with and without a buffer layer over the temperature range of 280–320 K. For the buffer-free device, J_{sc} increases monotonically with temperature, reflecting thermally enhanced carrier generation and transport as well as reduced carrier trapping at elevated temperatures. In contrast, the buffered device exhibits a non-monotonic behavior, with J_{sc} increasing from 280 to 300 K and subsequently decreasing at higher temperatures. This trend suggests that the buffer layer effectively optimizes carrier extraction at moderate temperatures while suppressing thermally activated recombination pathways; however, at elevated temperatures, increased recombination or transport losses may slightly offset these benefits. Overall, the buffered device maintains higher and more stable J_{sc} values across the investigated temperature range, demonstrating the crucial role of the buffer layer in improving thermal stability, interfacial quality, and photocurrent performance [29].

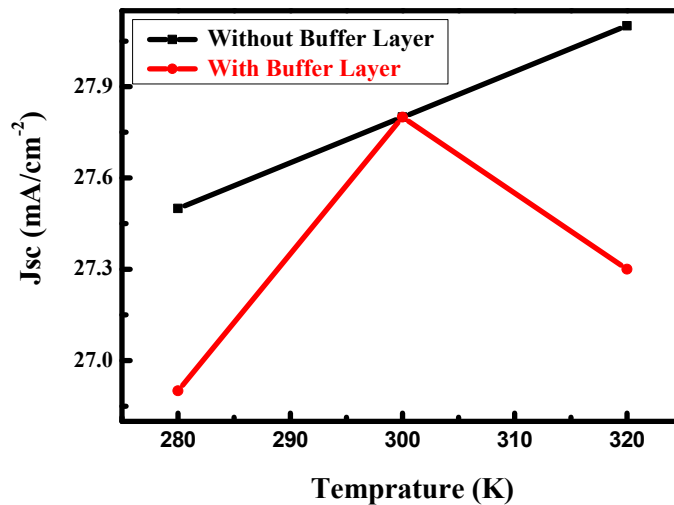


Figure 32: Variation of short-circuit current density as a function of temperature for buffered and buffer-free devices.

Temperature Dependence of the Open-Circuit Voltage with and without a Buffer Layer

Fig. 33 depicts the variation of the open-circuit voltage (V_{oc}) as a function of temperature for devices fabricated with and without a buffer layer over the temperature range of 280–320 K. For both configurations, V_{oc} decreases monotonically with increasing temperature, which is characteristic of enhanced thermally activated recombination and the associated reduction in the quasi-Fermi level splitting at elevated temperatures. Notably, the buffered device exhibits a higher V_{oc} at lower temperatures, indicating improved

junction quality and reduced interfacial recombination losses due to effective passivation by the buffer layer. As the temperature increases, the rate of V_{oc} reduction is more pronounced in the buffered device, suggesting increased sensitivity to thermally activated recombination processes at higher temperatures. Overall, the observed trends highlight the critical role of the buffer layer in enhancing voltage performance at moderate temperatures, while also emphasizing the influence of temperature-dependent recombination dynamics on device operation [18].

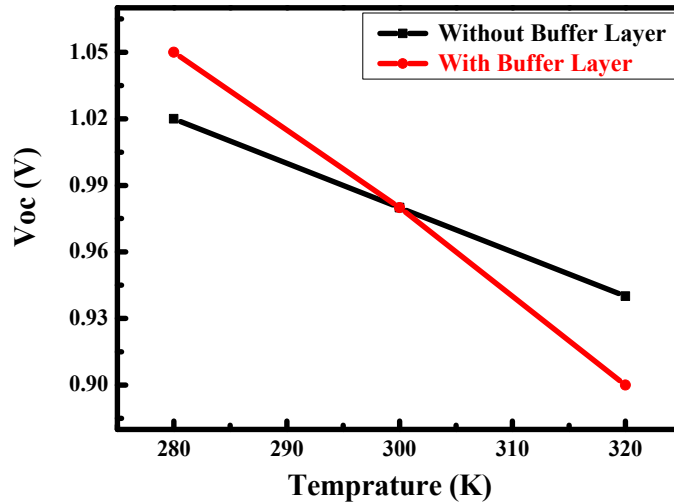


Figure 33: Variation of open-circuit voltage as a function of temperature for buffered and buffer-free devices.

3.11.7 Impedance Analysis

The impedance analysis provides valuable insight into the charge transport and recombination dynamics of the fabricated solar cell under varying operating conditions. The Nyquist plots reveal a dominant semicircular response, indicating that the device behavior is primarily governed by charge-transfer resistance at the interfaces. Under illumination, a significant reduction in the semicircle diameter is observed, reflecting enhanced carrier generation and improved interfacial charge transfer, while increasing temperature further decreases the resistance due to thermally activated transport. Additionally, the inverted measurement configuration exhibits smaller semicircle radii compared to the regular configuration, suggesting reduced transport resistance and more efficient charge extraction. Overall, the combined effects of illumination, temperature, and device configuration demonstrate improved carrier dynamics and reduced recombination losses, contributing to enhanced photovoltaic performance [31,32].

Temperature- and Illumination-Dependent Impedance Response of the Solar Cell

Fig. 34 presents Nyquist impedance spectra of the device measured under dark and illuminated conditions at temperatures of 280 and 300 K. In all cases, the spectra exhibit a dominant semicircular feature, which is characteristic of charge-transfer resistance associated with interfacial recombination processes [28]. Under dark conditions, the semicircle diameter is noticeably larger, indicating higher recombination resistance and limited carrier transport in the absence of photogenerated carriers. Upon illumination, a pronounced reduction in the semicircle radius is observed, reflecting enhanced charge carrier density and accelerated interfacial charge transfer. Increasing the temperature from 280 to 300 K further decreases the arc diameter in both dark and illuminated states, suggesting thermally activated carrier transport and reduced interfacial resistance. The combined influence of illumination and temperature

highlights the strong dependence of the device impedance response on recombination dynamics and charge transport processes, providing valuable insight into the mechanisms governing photovoltaic performance.

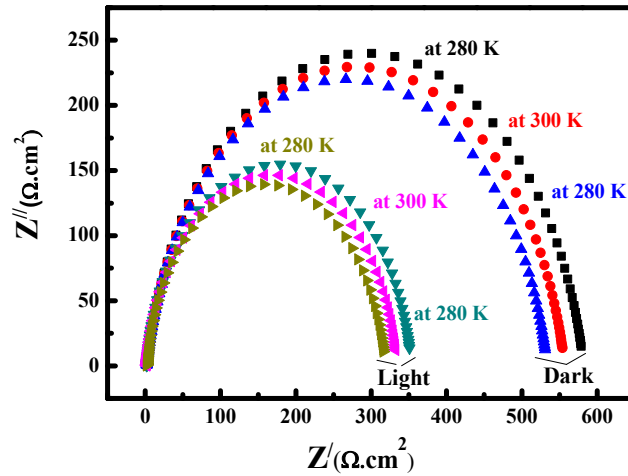


Figure 34: Nyquist plots measured under dark and illuminated conditions at different temperatures.

Impedance Response under Regular and Inverted Measurement Configurations

Fig. 35 shows Nyquist impedance spectra recorded under dark and illuminated conditions using regular and inverted measurement configurations. All spectra display a characteristic semicircular response, indicating that the impedance behavior is dominated by charge-transfer resistance associated with interfacial recombination processes. Under dark conditions, the regular configuration exhibits the largest semicircle diameter, reflecting higher recombination resistance and limited carrier transport [28]. Upon illumination, a significant reduction in the arc radius is observed, consistent with increased photogenerated carrier density and enhanced interfacial charge transfer. In the inverted configuration, both dark and illuminated spectra show smaller semicircle diameters compared to the regular configuration, suggesting reduced transport resistance and improved charge extraction. The illuminated inverted condition exhibits the smallest arc, highlighting the combined effect of favorable measurement configuration and photoexcitation in minimizing interfacial recombination losses. These results demonstrate that both illumination and device configuration play critical roles in governing the impedance response and charge transport dynamics of the device [31,33].

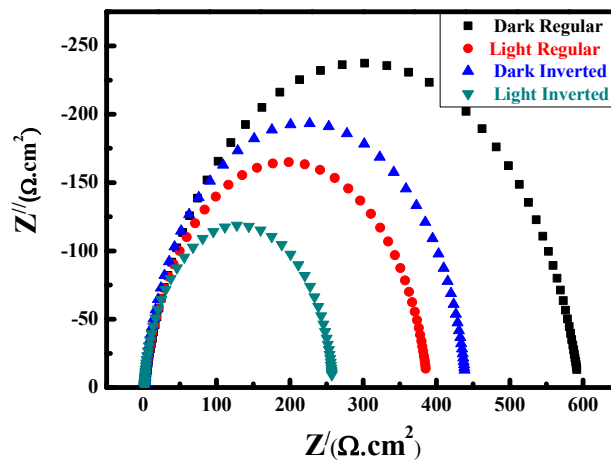


Figure 35: Nyquist plots under dark and illuminated conditions for regular and inverted configurations.

3.11.8 Operational Stability and Long-Term Durability of the Glass/ITO/Cu₂O/CdTe/ZnS-RGO/Al Solar Cell

The long-term operational stability of the fabricated Glass/ITO/Cu₂O/CdTe/ZnS-RGO/Al solar cell was evaluated under continuous illumination, as depicted in Fig. 36. The device exhibits a gradual and controlled decline in power conversion efficiency from an initial value of ~17.1% to ~15.0% after 500 h of operation. Notably, the efficiency retention remains as high as ~88% of its initial value, indicating excellent photostability. The slight degradation observed over time can be attributed to minor interfacial deterioration, such as the formation of trap states and limited oxidation effects at the heterojunction interfaces. However, the relatively slow decay rate confirms the effectiveness of the ZnS-RGO nanocomposite layer in suppressing charge recombination and enhancing interfacial stability. The presence of RGO provides improved electrical conductivity and facilitates efficient charge transport, while also acting as a protective barrier against environmental and thermal stress. Overall, the stability behavior demonstrates that the proposed device maintains robust performance over extended operation, thereby validating its suitability for practical photovoltaic applications and supporting the reliability of the reported high efficiency.

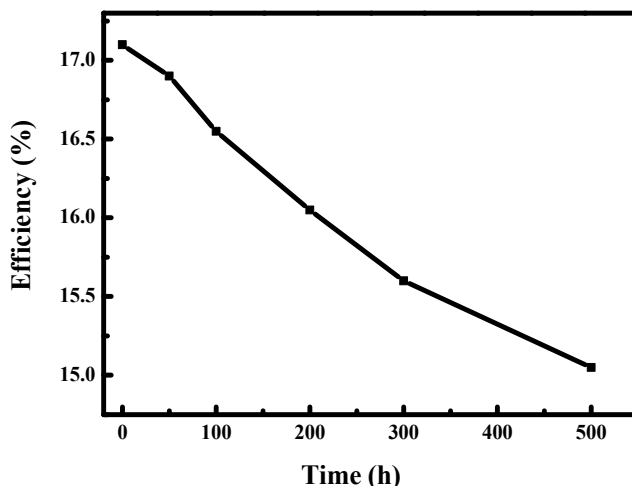


Figure 36: Time-dependent evolution of power conversion efficiency for the Glass/ITO/Cu₂O/CdTe/ZnS-RGO/Al solar cell under continuous illumination, demonstrating long-term operational stability over 500 h.

4 Conclusion

In conclusion, this study demonstrates the effectiveness of ZnS/RGO NCs synthesized using environmentally benign methods as advanced buffer layers for Cu₂O/CdTe thin-film solar cells. Comprehensive structural and surface analyses, including XRD and XPS, confirm the successful formation of phase-pure ZnS and its effective interfacial coupling with RGO, while preserving crystallinity and ensuring favorable surface chemistry. Optical investigations reveal enhanced light absorption across the visible spectrum and pronounced photoluminescence quenching upon RGO incorporation, indicative of efficient charge transfer and suppressed radiative recombination. When integrated into Glass/ITO/Cu₂O/CdTe/ZnS-RGO/Al device architectures, the optimized ZnS/RGO buffer layer delivers significantly improved photovoltaic performance, achieving a short-circuit current density of approximately 29 mA cm⁻², an open-circuit voltage of ~0.82 V, a fill factor of ~72%, and a maximum power conversion efficiency of ~17.1%. Impedance spectroscopy further reveals reduced series resistance and substantially enhanced recombination resistance, confirming effective mitigation of interfacial charge losses. Moreover, temperature-dependent electrical measurements conducted over the 280–320 K range demonstrate improved thermal stability and reduced performance

degradation. Collectively, these results underscore the critical role of ZnS/RGO interface engineering in optimizing charge transport and recombination dynamics, while the adoption of environmentally benign synthesis routes highlights the sustainability of the proposed approach. This work establishes ZnS/RGO NCs as promising, eco-friendly buffer layers for high-performance thin-film photovoltaic applications.

Future work should focus on long-term stability testing under real operating conditions and further optimization of RGO content to balance conductivity and transparency. Additionally, advanced techniques for carrier lifetime analysis and scaling up eco-friendly fabrication processes are recommended to enhance device performance and practical applicability.

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Author Contributions: Ahmed Mostafa conceived and designed the study, conducted the principal experimental work, performed data analysis and interpretation, and prepared the original manuscript. Samar Al-Shehri and Azzah A. Alshehri contributed to the experimental work, data analysis, and discussion of the results. A. Ashour contributed to the interpretation of the results and revision of the manuscript. All authors contributed to editorial changes in the manuscript, reviewed the manuscript, and approved the final version of the manuscript. All authors reviewed and approved the final version of the manuscript.

Availability of Data and Materials: The datasets generated and analyzed in the present study can be made available by the corresponding author upon reasonable request.

Ethics Approval: Not applicable.

Conflicts of Interest: The authors declare no conflicts of interest.

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