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By Universitas Muhammadiyah Sidoarjo

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Academia Open

Vol. 11 No. 1 (2026): June
DOI: 10.21070/acopen.11.2026.13176

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Structural, Optical, and Morphological Evaluation of Ag-Doped ZnO/CuO/Porous Silicon Heterojunctions for Optoelectronic Applications: Evaluasi Struktur, Optik, dan Morfologi Heterojunctions ZnO/CuO/Silikon Porous yang Doping Ag untuk Aplikasi Optoelektronik

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Abstract

General Background Renewable energy technologies increasingly rely on advanced heterojunction architectures to improve light harvesting and charge transport in optoelectronic devices. **Specific Background** Multilayer systems combining ZnO, CuO, and porous silicon offer complementary electronic and optical properties, while plasmonic silver incorporation provides additional light-matter interaction pathways. **Knowledge Gap** However, a systematic experimental correlation between Ag doping concentration, nanostructural evolution, optical behavior, and photovoltaic response in ZnO/CuO/PSi heterojunctions remains insufficiently explored. **Aims** This study aims to fabricate and evaluate Ag-doped ZnO/CuO/porous silicon heterojunction devices and to examine the role of silver concentration on structural, optical, and electrical characteristics. **Results** Structural and morphological analyses reveal refined crystallite sizes and pronounced morphology evolution with increasing Ag content, accompanied by enhanced visible-light absorption and bandgap reduction to 2.78 eV at 7% Ag doping. Photovoltaic measurements demonstrate a maximum open-circuit voltage of 3300 mV, a fill factor of 45.09%, and a power conversion efficiency of 0.3125% for the optimized device. **Novelty** The work provides an integrated experimental assessment linking plasmonic Ag doping to interface engineering and device-level performance within a single heterojunction framework. **Implications** These findings offer practical guidance for designing cost-effective optoelectronic devices and advancing plasmonic-assisted multilayer solar cell architectures.

Keywords: Ag-doped ZnO, CuO Heterojunction, Porous Silicon, Plasmonic Nanoparticles, Photovoltaic Devices

Key Findings Highlights:

Progressive silver incorporation induces significant morphological and crystallographic modification in multilayer structures.

Visible-light absorption increases markedly due to combined plasmonic and bandgap engineering mechanisms.

Optimized heterojunction configuration exhibits superior voltage output and charge collection behavior.

Introduction

Solar energy has become one of the most prospective sources of renewable and sustainable energy sources, which attract more and more attention to the world over the last decades as it has the potential to offer clean and sustainable alternatives to traditional fossil energy sources. As the environmental issues of global warming and climate change continue to rise, there has been strategic need to develop solar cells with high efficiency and low costs as a means of global energy security and resource sustainability. Here, solar cell based on thin film has become an option because of the various attributes associated with it such as light weight construction, mechanical flexibility, and ability of large area construction through affordable manufacturing methods as opposed to the conventional crystalline silicon solar cells.

The innovations in the field of thin-film technology have not just been in enhancing the solar cell efficiency but has also been applied in a variety of high-technology use in optical and electronic devices such as sensors, light-emitting diodes, and photodetectors. This is because of the fact that these films can manipulate electronic and optical properties by manipulating their thickness, chemical composition or interface engineering. Therefore, the development of hybrid multilayer structures creates possibilities to achieve a high improvement of optoelectronic performance and allows using a wide range of solar radiation. Multi-junction solar cells are one of the latest designs in the area, which is based on combination of several layers of semiconductors with various bandgaps, and therefore, can absorb a broad spectrum of the solar spectrum with minimum loss of energy.

The transition metal oxides (TMOs) have become prominent in photovoltaic and optoelectronic utilization following their appropriate bandgaps, various oxidation states, stable crystalline structures and unique optical and electrical features like high absorption coefficients and chemical stability. An example of such materials is copper oxide (CuO) and zinc oxide (ZnO). ZnO is an n-type semiconductor, which has a wide bandgap (3.2-3.4 eV), a high transparency, and a high mobility of the electron, and this property is excellent in transparent conducting electrodes and electron transport layers. On the other hand, CuO is a p-type semiconductor that has a smaller bandgap (1.2-1.7 eV) and has large visible light absorbing capacity making it an effective hole transport layer and light absorber [6, 7].

Porous silicon (PSi) is one technology that has transformed the silicon-based photovoltaic technology because Canham discovered its powerful photoluminescence at room temperature in 1990. The porous silicon, in contrast to bulk crystalline silicon, an indirect bandgap material with a low ability to emit light, show quantum confinement effects at the nanoscale (usually 1-100 nm) of size. This quantum confinement alters the electronic structure of its material to form an indirect bandgap into a quasi-direct bandgap and has great benefits in optical absorption and emission characteristics. The nanoporous structure has a number of benefits: it offers more surface to trap light by multiple internal reflections, minimized surface reflection losses, heightened separation of charge carriers at heterojunction interfaces, and is compatible with different deposition modes of oxide semiconductors.

Noble metal nanoparticles, and especially silver (Ag), have been proposed as a potent method of enhancing the performance of photovoltaics by a process called plasmonic enhancement. Silver nanoparticles are known to possess localized surface plasmon resonance (LSPR) in the visible spectrum, which results in strong local electromagnetic fields which enhance the absorption of light in the surrounding semiconductor materials significantly. Ag nanoparticles can offer several advantages when used in heterojunction solar cells: optical absorption via plasmonic near-field coupling, charge carrier generation and collection, electron trapping centers to reduce electron-hole recombination, and bandgap tuning of host semiconductors. These plasmonic effects are very dependent on the size, shape, concentration, and distribution of Ag nanoparticles and must be optimized, with care, to trade off enhancement mechanisms and possible recombination losses.

The study is concerned with the formulation and production of a multilayer heterojunction architecture that is founded on Ag-doped ZnO/CuO/PSi/Si system with the view of producing high efficiency and cost effective solar cells and optoelectronic devices. These materials are chosen on the basis of their complementary characteristics: n-type ZnO is useful as an excellent electron conductor which has high transparency and p-type CuO designs are characterized by strong capability of absorbing visible light and hole conduction, porous silicon designs form nanostructures with natural improvement of light capture and reflections, and latent silicon designs have crystalline structure and high efficiency in the absorption of light and conduct of holes. The addition of silver nanoparticles at optimal levels will seek to utilize the plasmonic enhancement effects so as to enhance optical absorption and charge carrier dynamics.

Multiple aims are fulfilled by the combination of such materials in a multilayer heterojunction system: better absorption of the solar spectrum in the UV- visible-NIR range, lower optical losses due to anti-reflection and light trapping, better electrical conduction between layers due to optimal band alignment, and lower processing costs of the material by a solution method. Moreover, the offered structure is not confined to the application of solar cells only but can be utilised in other new optical and electronic devices like photodetectors, light-emitting diodes, and optical communication devices.

This study is important in that it fills the gap between the potential of theories and their practical applications by introducing a comprehensive research based on both theoretical study and experimental observation. The systematic investigation of the synthesis of materials, structural characterization, optical properties, and device performance furnishes some of the basic understanding of the physics of heterojunctions as well as nanoscale interfaces engineering. Knowledge of the correlation between silver doping level, morphology development, optical properties, and photovoltaic properties can be used to rational design the next generation optoelectronics device. The results will lead to the creation of more effective and sustainable solar energy technology in addition to the new role of thin-film semiconductors in the current electronic and photonic systems capable of converting power to high conversion efficiency at low manufacturing cost, creating stable

heterojunction interfaces with low defect-recombination rates, optimizing light management using plasmonic and nanostructural design and building scalable fabrication methods that can be used in manufacturing. This study offers a holistic way to design advanced optoelectronic devices by offering systematic research on the impact of silver nanoparticle doping on structural, optical and electrical properties of ZnO/CuO heterojunctions on porous silicon substrate, which enables the design of such devices with specific performance characteristics to be used in various applications in renewable energy devices and photonic systems[13-15].

Experimental Methods

2.1 Materials and Chemicals

N-type silicon wafers, hydrofluoric acid (HF, 40%), ethanol (99.9% purity), copper chloride (CuCl_2), sodium hydroxide (NaOH), zinc acetate dihydrate ($\text{Zn}(\text{CH}_3\text{COO})_2 \cdot 2\text{H}_2\text{O}$), silver target (99.99% purity), and deionized water were taken as received with no additional purification.

2.2 Synthesis of Porous Silicon

Photo-electrochemical etching was done on n-type silicon wafers to form porous silicon layers in an electrolyte solution of HF and ethanol. The silicon wafers were drained and placed in Teflon electrochemical cell with platinum as the counter electrode. Photo-assisted anodization was done with a steady current density of 20 mA/cm^2 and under light to produce uniform nanopores structure of 20 minutes. The resultant PSi samples were washed with ethanol and dried in the nitrogen gas.

2.3 Preparation of Silver Nanoparticles

Silver nanoparticles were prepared through the pulsed laser ablation in liquid technique (PLAL). A silver target was moistened in the deionized water and ablated with Nd:YAG laser of 1064 nm wavelength, 8 Hz repetition rate and 100 pulses. The colloidal suspension that was formed contained the spherical Ag nanoparticles with average diameter of 15-45nm.

2.4 Synthesis of Ag-Doped ZnO Nanoparticles

Silver doped zinc oxide nanoparticles were produced through hydrothermal decomposition process. Zinc acetate precursor solution was added to silver nanoparticle suspension in three different concentrations (3, 5 and 7 percent by weight). The mixtures were put through hydrothermal treatment to produce Ag-doped ZnO nanocrystals at varying levels of doping.

2.5 Deposition of CuO Thin Films

The films of copper oxide were made through the chemical precipitation process. The aqueous solution of 0.2 M copper chloride was reacted with sodium hydroxide to form CuO precipitate which was deposited on porous silicon substrates to form the p-type hole transport layer.

2.6 Device Fabrication

The entire heterojunction devices were prepared by sequential deposition of porous silicon as n-Si substrate, deposition of copper oxide as p-type material as CuO layer and deposition of n-type electron transport material as Ag-doped zinc oxide layer. Electrical characterization of the device was done by applying metal contacts to complete the structure.

2.7 Characterization Techniques

The analysis was carried out on X-ray diffraction (XRD) to identify crystal structure, phase composition, and size of the crystallite of the sample using Scherrer equation. The chemical bonding and functional groups were determined by the use of Fourier-transform infrared spectroscopy (FTIR). The morphology of the surface and the particle size distribution were observed with the help of field emission scanning electron microscopy (FE-SEM). To measure bandgap energies and optical absorption properties, measurement of optical absorption spectra was performed with the help of UV-Vis spectrophotometry. Dark and light measurements Current-voltage (I-V) measurements were performed to measure photovoltaic parameters:

open-circuit voltage (V_{oc}), short-circuit current (I_{sc}), fill factor (FF) and power conversion efficiency. (η).

Results and Discussion

3.1 Structural Characterization by X-Ray Diffraction (XRD)

3.1.1 Silver Nanoparticles

An X-ray diffraction of the silver nanoparticles prepared through pulsed laser ablation in liquid (PLAL) provided diffraction peaks with 2 theta values of 38.10, 44.30, 64.40 and 77.40, which belong to (111), (200), (220) and (311) crystallographic

planes of face-centered cubic (FCC) silver structure (JCPDS card No. 04-0783). Majority peak of 38.10 indexed to plane (111) shows that it is preferentially oriented in this direction which is characteristic of metallic silver nanoparticles. The mean crystallite size determined by Debye-Scherrer equation was 25.20 nm and individual crystallite size of 24.01 to 26.82 nm at various diffraction peaks respectively. The size distribution is relatively small implying homogenous nucleation and growth of the material during the laser ablation.

The quality of silver nanoparticles was also measured by calculating the dislocation density (δ) and microstrain (η). The density of dislocation was 13.90×10^{14} to 17.34×10^{14} lines/m², and the microstrain was 9.41×10^{-4} and 15.07×10^{-4} . These comparative low values are a good indicator of the low lattice defects and strain that ensures high crystalline quality of the synthesized nanoparticles. The PLAL method does form nanoparticles with fewer defects than any of the chemical synthesis methods because the speed of quenching prevents oxidation and contamination of the crystalline structure in the swift liquid medium.

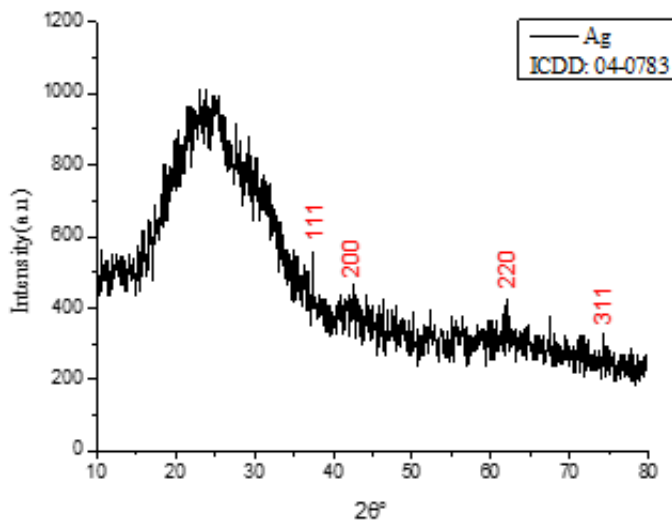


Figure 1. Figure (1): X-ray diffraction (XRD) pattern of the prepared silver material.

2θ (Deg.)	FWHM (Deg.)	Crystallite size (nm)	Average size (nm)	Crystallite $\delta \times 10^{14}$ lines/m ²	$\eta \times 10^{-4}$
38.10	0.35	24.01	25.20	17.34	14.96
44.30	0.35	24.51			