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Influence of Silar Cycle on The Energy Bandgap of Iron Copper Sulphide (FeCuS) Thin Films Deposited on SLG Substrate

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Abstract

This study aims to investigate the influence of SILAR cycle on the energy bandgap of Iron Copper Sulphide (FeCuS) thin films deposited on soda-lime glass substrate (SLG). A Successive Ionic Layer Adsorption and Reaction (SILAR) method is one of the chemical methods for making uniform and large area thin films, which is based on immersion of substrates into separately placed cationic and anionic precursors. The technological importance of chemically deposited iron copper sulphide (FeCuS) using SILAR technique vis-à-vis the effect of SILAR cycle on the energy band gap of the deposited thin films has not been unraveled. Thin films of Iron Copper Sulphide were grown on soda-lime glass substrate (SLG) by a simple solution based Successive Ionic Layer Adsorption and Reaction (SILAR) technique at room temperature (300 K) with EDTA, TEA and NH_4OH as complexing agents at different SILAR cycles (20, 30 and 40 cycles) of deposition. The thin films grown were characterized using Avantes UV-VIS spectrophotometer (Avalight-DH-S-BAL) in the wavelength range 200-1000nm and Four Point Probe machine (Keithley 4ZA4 2400 Sourcemeter, manufactured by Tektronix Company). The optical properties considered revealed high absorbance and reflectance but low transmittance in the UV region; low values of absorbance and reflectance accompanied with high transmittance in the VIS region. Moreover, the resistivity of the grown thin film varied from 9.480 x 10^6 Ωm to 4.366 x 10^7 Ωm in order of increasing SILAR cycle, direct band gap of 3.76e V, 3.51e V and 3.42e V were obtained. These properties suggest that the films are suitable for solar cell and optoelectronic applications.

Key Words

SILAR cycle, Iron copper sulphide (FeCuS), Energy band gap, SLG substrate, Thin films

Introduction

Photovoltaic came into existence as a rapidly growing field because of the global search for an alternative source of power generation. It is a direct means for detection and conversion of solar radiations into electrical energy. The solar energy is a free gift of nature to humankind, which is pollution-free, can be sensed, controlled and used to produce power using solar cell modules. Woodford [1] predicted that the sun has enough fuel onboard to drive our Solar System for another five billion years and solar panels can turn this energy into an endless and convenient supply of electricity.

Many researchers have succeeded in depositing iron copper sulphide (FeCuS) thin films using chemical bath deposition (CBD) [2, 3] and solution growth technique (SGT) [4]. A successive ionic layer adsorption and reaction

(SILAR) method is one of the chemical methods for making uniform and large area thin films, which is based on immersion of substrates into separately placed cations and anions sources. The scope of SILAR technique in the field of solar cell fabrications is enormous as this method has excellent material utilization efficiency, good control over the deposition process along with the film thickness, low cost and large-scale deposition capability on virtually any type of substrate which makes it very attractive, relatively simple, easy, economical and reproducible. There is considerable interest in the deposition of ternary derivative material, due to the potential of tailoring both the lattice parameters and the band gap by controlling depositions parameters [5] like the SILAR cycles.

SILAR technique has numerous advantages over other techniques such as uniform film deposition, control of thickness, precise maintenance of deposition temperature, low cost and wastage of material is avoided. The technique could also be used to deposit compound materials on a variety of substrates such as insulators, semiconductors and metals. To the best of our knowledge, SILAR technique has not been employed to synthesize the technological importance of iron copper sulphide (FeCuS) and there are fewer reports on the preparation of solar cells fabrication with FeCuS materials deposited by SILAR technique.

Experimental Details

Stoichiometric quantities of analytical grade reagents of ferrous nitrate, cuprous chloride and thiourea served as precursors, ethylene diamine tetraacetate (EDTA) and triethanolamine (TEA) were used as complexing agents with deionized water and ammonia solution.

The deposition method adopted in this study is Successive Ionic Layer Adsorption and Reaction (SILAR). FeCuS thin films were deposited on glass (SLG) substrates (76.2 mm x 2.5 mm x 1.2 mm) when immersed into the precursors and is carried out using four beakers system (Beakers I, II, III and IV) at room temperature. In order to remove the organic and inorganic impurities, the glass substrates were first degreased in concentrated trioxonitrate (V) acid for 48 hours, washed with detergent, rinsed in distilled water and dried in air. The degreased cleaned surface provide nucleation center for the growth of the films, hence yielding highly adhesive and uniformly deposited films on the surface of the substrates.

A four-beaker system solution was obtained by first preparing 5 ml of 0.4 M ferrous nitrate, 5 ml of 1 M cuprous chloride, 3 ml of 14 M ammonia solution, 3 ml of 7.4 M TEA, 3 ml of 0.1 M EDTA was placed into 50 ml beaker (Beaker I) and followed by magnetic stirring at room temperature to obtain a homogenous solution. Then, 20ml of distilled water was put into two different 50 ml beakers (Beakers II and IV). Finally, 20 ml of 1 M thiourea was placed into another 50 ml beaker (Beaker III). The solutions in the beakers I and III served as cationic and anionic precursors. The ferrous nitrate, cuprous chloride and thiourea served as sources of Fe^{2+}, Cu^{2+} and S^{2-} respectively while the TEA and EDTA as complexing agents, slowed down the reactions for the formation of solid thin films on the substrates and the ammonia solution to stabilize the pH of the mixtures. The substrate was kept vertically in each beaker at every immersion to prevent it from slanting or falling in the beaker. The deposition was done at room temperature for 80 seconds dip time per cycle.

One SILAR cycle consisted of four steps: adsorption of both iron and copper species for 20 s, rinsing with distilled water for 20 s to remove excess adsorbed or loosely bounded iron and copper species, reaction with thiourea precursor solution for 20 s to form stable FeCuS and rinsing with purified water for 20 s to remove excess or unreacted species or powdery FeCuS. By repeating such deposition cycle 20, 30 and 40 times, FeCuS thin films were obtained respectively. After the deposition, the substrates were removed and allowed to dry in Laboratory open air. The samples deposited at 20, 30 and 40 cycles were labeled FeCuS-20(0.4 M), FeCuS-30(0.4 M) and FeCuS-40(0.4 M) respectively.

The optical analyses data of the thin films were obtained from Avantes UV-VIS spectrophotometer (Avalight-DH-S-BAL) in the wavelength range 200-1000 nm and the electrical characterization was examined with the aid of Four Point Probe machine (Keithley 4ZA4 2400 Source meter, manufactured by Tektronix Company) for measurement of current (I) and voltage (V).

Results and Discussion

Figure 1 shows that the samples have reflectance values which are less than 15% and 33% (with the sample deposited at 40 cycles having the least and highest values of 3.7% and 32.5%) for both at the UV and NIR regions respectively. Results from reflectance spectra showed that these properties mostly decrease and at another time increase with SILAR cycle. This provides wide latitude for the applications of the thin film. Average reflectance was found to be below 30% for all films.

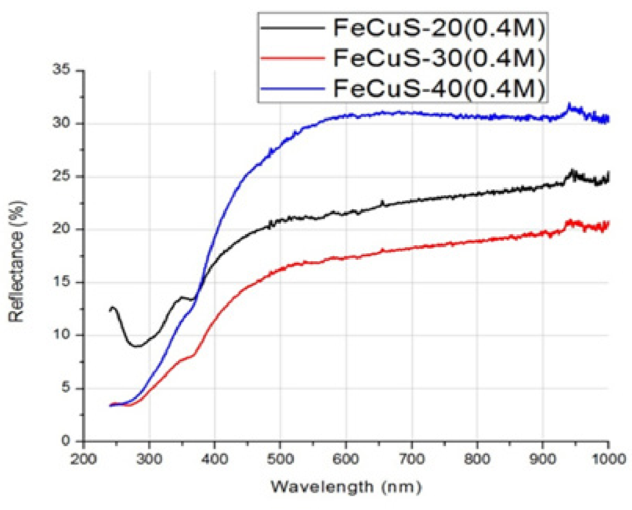

Figure 1: Graph of reflectance against wavelength of the grown thin films.

For wavelength above 500 nm, it is observed from the transmittance graph (Figure 2) that the deposited films at 20

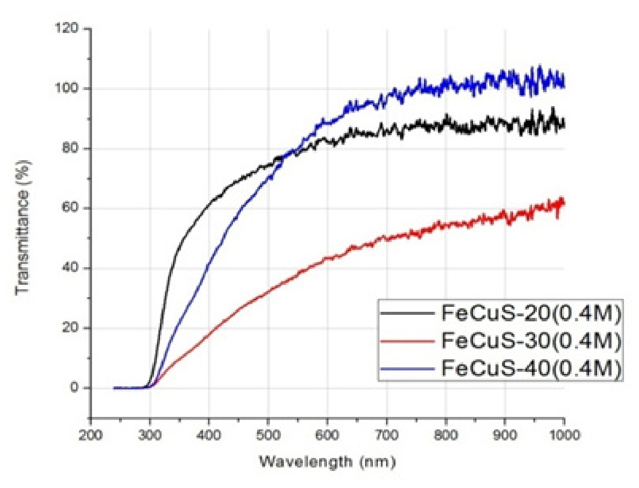

Figure 2: Graph of transmittance against wavelength of the grown thin films.

and 40 cycles both have high transmittance above 80% while the sample deposited at 30 cycles has the least transmittance of 60%. The transmittance varies most times as SILAR cycle and at another time inversely proportional. In all the films below 450 nm there was a sharp fall in the percentage transmittance of the films which indicates a strong increase in photon absorption [6]. It denotes that some states have been created in the Fermi-level between the conduction and valence band. This can also be attributed to the increase in fundamental absorption as photon striking increases with increase in carrier concentration [7]. As shown on the graph, average transmittance at wavelength (λ) = 800 nm was found to be between 80% and 90%. These values of fairly low and high transmittances in UV and VIS-NIR regions respectively are in agreement with previous researchers [2, 4, 8, 9]. The results further revealed the utilization of the materials in solar cell. The optical absorption spectra of the thin films shown in figure 3 it was observed that all the deposited samples exhibit high absorbance in the UV region of the electromagnetic spectrum between wavelengths of 200 nm and 300 nm which decreased with increasing wavelength of solar radiation. However, the absorbance of the thin film is between 2.5 and 4.7. The increase in absorption occurs when the photon energy reaches the value of the energy gap where electronic transfers occur between the valence band and conduction band. The film shows relatively low absorbance in the NIR regions of the spectrum (less than 1.0). This is in consonance with the reports of [2, 4, 8] as they also observed high absorbance in the UV region and low absorbance in the VIS-NIR region.

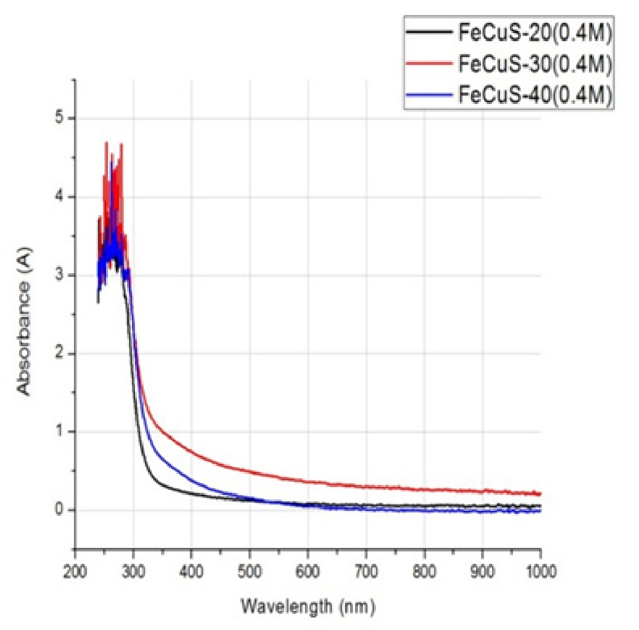

Figure 3: Graph of absorbance (A) against wavelength of the grown thin films.

In order of increasing SILAR cycle, direct band gap of 3.76e V, 3.51e V and 3.42e V were obtained from the graph of (αhv)2 as a function of the photon energy (hv) plotted for direct allowed transition (Figure 4), through the intercept on energy axis after extrapolation of the straight-line section in the high-energy region of the plot. It is observed that the band gap values decreased as the SILAR cycle increased. There is an obvious contrast between the range of these direct band gap values and the results of previous researchers [2-4, 8]. The values obtained were found to be higher than previous researchers which could be attributed to the variation in the molar concentrations of the precursors used, the deposition time, and impurities from the environment where the research was conducted. The band-gap of the window layer should be high and the layer be thin as possible to maintain low series resistance [10, 12]. This is to ensure that the window layer does not absorb any of the incident light and to allow maximum photon energy to reach the absorber layer where it is needed for generation of electrons.

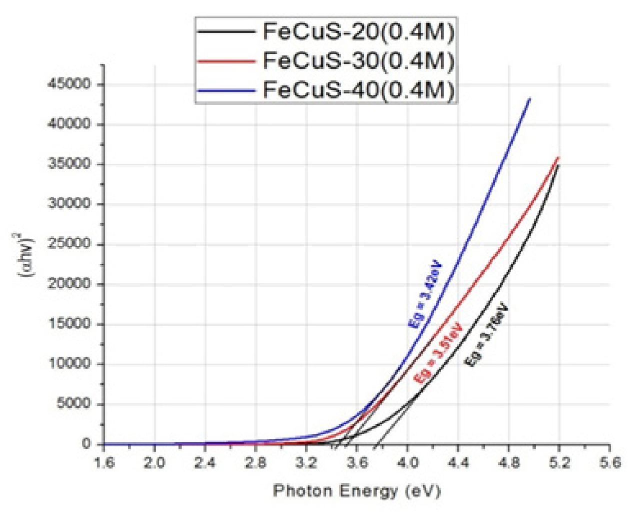

Figure 4: Graph of (αhv)2 against Photon energy in eV of the grown thin films.

The resistivity of the grown thin film varied from 9.480 x 10^6 Ωm to 4.366 x 10^7 Ωm as shown in table 1. This enhances the electrical property of the film and makes it a good material for solar applications. Resistivity should not be too high or low due to the inevitable defects in solar cells during the actual production process [11]. The results of the films deposited showed that the resistivity decreases as SILAR cycle increases.

Table 1: Electrical results of the thin films.

Thin film Samples	Voltage (V)	Current (A)	Sheet resistance, R_s (Ωm^{-2})	Resistivity (Ωm)
FeCuS-20 (0.4M)	4.370×10^{-1}	9.075×10^{-8}	2.183×10^{7}	4.366×10^{7}
FeCuS-30 (0.4M)	4.685×10^{-1}	3.223×10^{-8}	6.590×10^{7}	1.318×10^{7}
FeCuS-40 (0.4M)	3.107×10^{-1}	2.972×10^{-8}	4.740×10^{7}	9.480×10^{6}

Conclusion

The deposition and characterization of FeCuS thin films have been successfully achieved. The results showed that the energy band gap values of the thin films decreased as the SILAR cycle increased. Other solid-state properties of the materials were influenced as the SILAR cycles increased. This provides wide latitude for applications of the thin films in the following: solar cell fabrication, screening off UV radiation, low transmittance and low reflectance in UV region, coating of poultry buildings, eye glasses coating, solar thermal conversion, solar control, anti-reflection coating and window layers in solar cells. Anticipation is that the growing of the thin film with required specific cycle will lead to more applications of the ternary compound semiconductor.

Recommendations

It is recommended that other types of substrates such as Si wafer, and titanium foil should be used in the deposition of FeCuS thin films using SILAR technique. Other methods of thin film deposition such as electrodeposition, chemical vapour deposition etc. be used and results compared with that of this work. All other characterization such as Energy Dispersive X-ray analysis (EDX), X-Ray Diffraction (XRD), etc. should also be considered. Scaling up of the applications of the result in this work is also recommended.

Conflict of Interest

No conflict of interest among authors.

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