

Influence of Solvents on Properties of Aluminium Sulphide Thin Films Synthesized by Chemical Spray Pyrolysis Technique

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ABSTRACT

In this paper, the influence of solvents on the structural properties of spray-deposited AIS thin films has been studied. Different predetermined mixtures were prepared from ethylene glycol, deionized water and alcohol solvents and sprayed on heated glasses *via* a simple and reliable technique known as spray pyrolysis. XRD patterns confirmed cubic and tetragonal phases of synthesized AIS material.

Keywords: AIS, Thin film, Spray pyrolysis

INTRODUCTION

Aluminium sulfide is a group III-VI chalcogenide compound with semiconducting properties [1]. It finds applications in tanning chemicals, papermaking, preparation of hydrogen sulphide, and production ethanethiol [2] and manufacturing cathodes used in lithium-sulfur solid-state batteries [3].

The listed applications are because of AIS wide bandgap of about 4.1 eV which is one of the highest among chalcogenide semiconductors [4]. The constituents of AIS are available in abundant and can be a possible replacement for cadmium sulfide which is an n-type partner in commercialized thin-film solar cells, such as Copper Indium Gallium Selenide (CIGS) and Cadmium Telluride (CdTe) solar cells [5]. It can also be used as a buffer layer in a carbon nano-tube growth [6]. The techniques used in the synthesis of AIS thin films in most reported works are wet chemical technique which includes spray pyrolysis [7], chemical bath deposition [8], successive ionic layer and reaction (SILAR) [9], chemical vapor deposition (CVD) [10] and others. Among the listed chemical methods, spray pyrolysis stands out as a simple technique for low-cost deposition [11]. It is applicable over a wide area, industrially scalable and stoichiometry can be easily controlled. It is common that deposition parameters in spray pyrolysis play a significant role in enhancing structural and opto-electronic properties of the final product [15]. One of the deposition parameters is the type of solvent used in preparation. Starting from predetermined solution that can be linked to behavior of the evaporating solution [12]. The use of deionized, distilled or double distilled water in preparation of pre-determined mixture containing aluminium salt and thiourea had been reported in most works on AIS thin films,

though complexing agents are sometimes added to enhance the stability of the solution [13-15]. The effect of solvent on properties of AIS has not been adequately investigated. The aim of this study is to investigate the effect of solvent on the structural properties of AIS thin films deposited by spray pyrolysis technique with an objective to correlate solid state properties of AIS thin film to solvent properties.

EXPERIMENTAL DETAILS

Pre-determined solutions containing 0.020 M of aluminium acetate dihydrate ($\text{AlC}_4\text{H}_6\text{O}_4 \cdot 2\text{H}_2\text{O}$) and 0.25 M of water-soluble organic compound thiocarbamide ($\text{CS}(\text{NH}_2)_2$) were prepared with solvents; ethylene glycol, deionized water and alcohol. All reagents were ordered from British Drug House England (BDH). The high concentration of thiocarbamide will act as a boost for Sulphur reduction during pyrolysis. Each solution was stirred continuously for an hour until a clear solution was obtained without the inclusion of any advanced agent except for the solution prepared with deionized water in which a drop of H_2SO_4 was added to prevent precipitation in the mixture. The solution was sprayed on soda lime glasses heated to 350°C at the rate of

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2mL/min. The air was used as the carrier gas at a pressure of 30 psi. Spray to target distance was maintained at 30 cm. The resulting AIS thin films were classified as specimens AIG, AIA and AID, where AIG represents sprayed AIS thin film sample obtained from ethylene glycol, AIA represents sprayed thin film specimen obtained from alcohol and AID represents sprayed thin film specimen obtained from deionized water. AIG, AID and AIA were annealed in a muffled furnace at 350°C for one hour. Weight of the films and substrates was measured using Adventurer balance (model: Adventurer Pro AV313 d = 0.0001g).

Characterization of AIS Thin Films

The structural characterization of the samples was done using an X-ray diffractometer with a lynx eye detector using a copper target ($\text{Cu}\alpha$, 1.5418 Å). All X-ray diffraction (XRD) data for the specimens was recorded at current and acceleration voltage of 25mA and 40 kV, respectively. The grain growth analysis of samples /crystallite size was carried out by using Hitachi scanning electron microscope (SEM).

RESULTS AND DISCUSSION

Structural Properties

The XRD patterns of AIG, AID and AIA are shown in **Figures 1-3**, respectively. The pattern of AIG shows diffraction peaks corresponding to (111), (220) and (311) reflection planes of cubic AIS which are observed at 2θ equaling 38.6°, 48.1° and 57.2°. The broadening observed at the peaks is an evidence of the nanocrystalline nature of the specimen. There was no evidence of secondary phase in the specimen. Crystallite size was calculated using Eq. 1 and 3.65 nm was obtained as the crystallite size of AID. The pattern of AID shows diffraction peaks at 2θ equaling 30.9°, 38.5°, 48.1°, 56.5° and 69.5° which can be indexed to (101),

(111), (112), (220) and (400) respective reflection planes which are the planes of cubic and hexagonal structure of AIS. However, some peaks corresponding to aluminium oxide (AlO) are observed at 2θ equaling 38.6° and 38.4°. The presence of oxides of Aluminium can be attributed to decomposition of microsized droplets of predetermined mixture during open air spraying. The calculated crystallite size for AID is 8.160 nm. The pattern of AIA shows peaks at 2θ equaling 30.4°, 33.6° and 38.10 which can be indexed to (111), (101) and (200) reflection planes of cubic and hexagonal phases of AIS respectively. Other peaks that cannot be indexed to planes of AIS were observed in the diffractogram of AIA. A crystallite size of 264.8 nm was obtained for AIA. The variation in crystallite size from crystalline in AIA and AID to nanocrystalline in AIG due to solvent influence can be explained by taking the vapor pressure and polarity of the solvents into account. Slower evaporation rate induced by lower vapor pressure may have been responsible for small grain size of AIG (**Figure 3**). The increase in crystallites size with corresponding decrease in polarity can be observed in **Figure 3**. It can be a suggestion that a reduction in solvent polarity causes loose coiling of bonds, which results in higher aggregate formation. There is an improvement in the structural properties of AIS thin film synthesized using ethylene glycol by post-deposition annealing at 350°C and without the use of hydrazine which is toxic. **Table 1** shows the polarities and vapor pressures of the solvents used in preparing the AIS predetermined solution.

$$G = \frac{0.9\lambda}{\beta \cos\theta} \quad (1)$$

Where G is the crystallite size, λ is the $\text{CuK}\alpha$, β is the full width at half maximum and θ is the angle of diffraction.

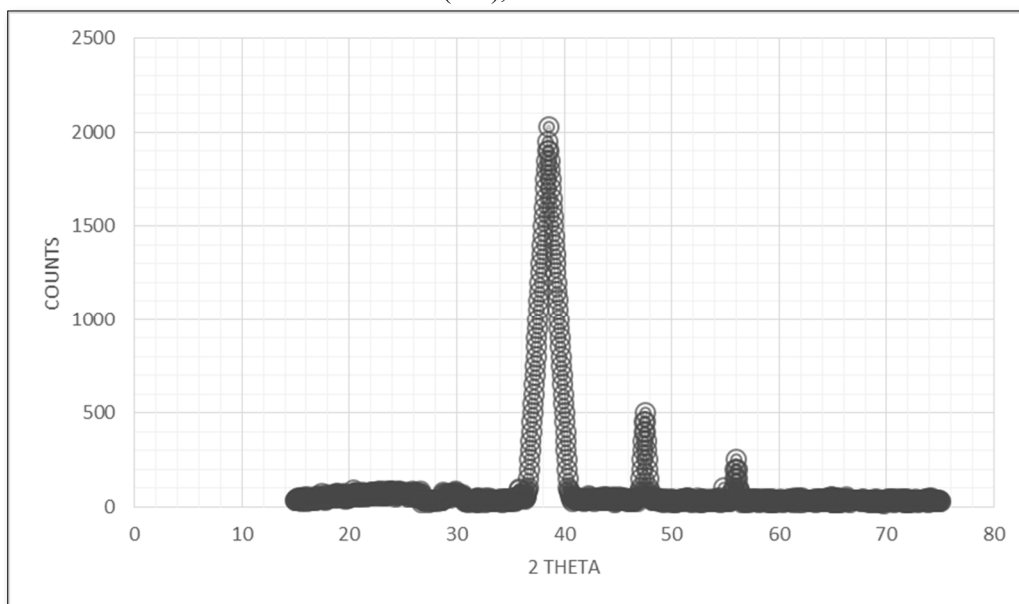


Figure 1. XRD Patterns for AIS thin films prepared using ethylene glycol solvent.

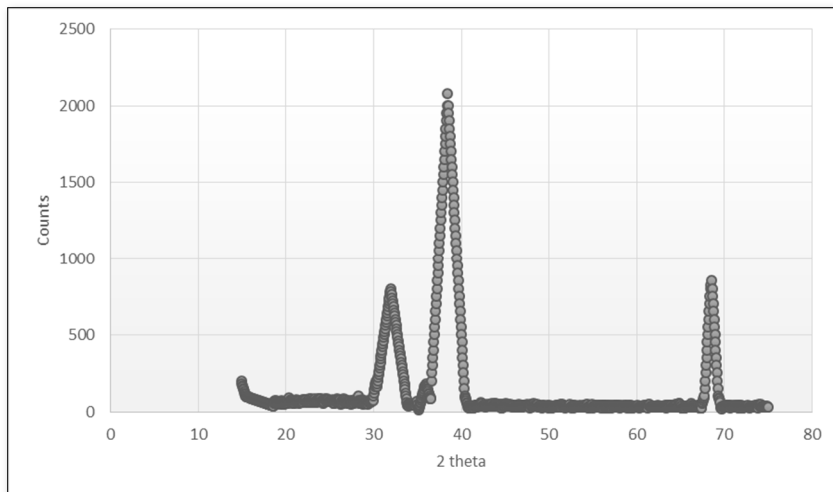


Figure 2. XRD Patterns for AIS thin films prepared using deionized water solvent.

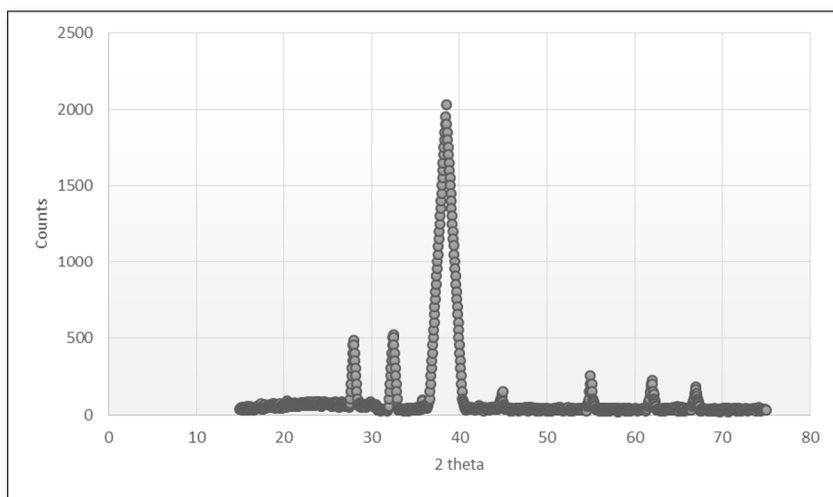


Figure 3. XRD Patterns for AIS thin films prepared using alcohol solvent.

Table 1. Dipole moments and vapor pressures of solvents.

S/No	Solvent	Dipole Moment (Debye)	Vapor Pressure (kPa)
1	Alcohol	1.650	5.950
2	Water	1.850	2.450
3	Ethylene Glycol	2.745	0.008

The composition of the solvent with varied AIS thin films was analyzed by energy dispersive X-ray spectroscopy (EDX). The composition of AIG, AID and AIA are shown in Table 2. The AIS compound is confirmed by the peaks of aluminium (Al) and sulphur (S) in the XRD. It can also be observed that all synthesized films are highly Sulphur-deficient, which may be attributed to affinity of sulphur

towards oxygen leading to formation of sulphur (IV) oxide which is lost during pyrolysis [15]. However, the highest atomic percentage of sulphur can be found in specimen AIG, as shown in Table 2, which may be due to drying retardation during the spraying process. Oxygen peaks observed may be due to air used as carrier gas and the carbon peak may be attributed to C=O functional group in AIG.

Table 2. Elemental composition of solvent with varied AIS thin films.

S/No	Specimen	Al Atomic %	S Atomic %	O Atomic %	C Atomic %
1	AlG	52.46	14.75	19.50	7.21
2	AID	59.29	10.25	30.46	Not available
3	AIA	67.00	8.60	20.10	4.30

CONCLUSION

In this study, AIS thin films have been synthesized via a chemical technique: spray pyrolysis. The influence of solvents on the structural properties of spray-deposited AIS thin films has been studied. Ethylene glycol, deionized water and alcohol are solvents used in preparation of the predetermined mixture containing aluminium salt and thiocarbamide. The films developed from predetermined mixture with ethylene glycol used as solvent exhibited cubic phase of AIS only, while those developed from deionized water and alcohol showed both cubic and tetragonal phases. It can be suggested from the small crystallite size (2.67 nm) of AlG that the film is nanocrystalline, while AIS thin film synthesized from deionized water has the largest crystallite size of 250.60 nm. The dents are absent in the surface of ethylene glycol-synthesized films. In the case of AIS films prepared from deionized water and alcohol, some voids can be noticed though the particle sizes are more distinguishable compared to those of ethylene glycol.

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