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Structural and Optical properties of CIAGS thin films

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ABSTRACT

CIGS thin films are proved to be the better candidate to be used as absorber layer in thin film solar cells. Addition of small amount of aluminum not only gives the provision in band gap tailoring but also has reduced the compound formation temperature. CIAGS thin films are prepared on glass substrates by thermal evaporation. Copper, indium, aluminum, gallium and selenium are used as source materials. Samples are annealed at different temperatures in the range 150^{o}C - 500^{o}C to study the compound formation. Bruker D8 Advance is used to study the structural properties of as-deposited and annealed samples. As-deposited results show the appearance of Indium peaks as is obvious from the layout of the deposited structure. Compound formation started at 150^{o}C and above. Variation in crystallite size and micro strain is also observed as a function of temperature. JA Wollam M2000 variable angle spectroscopic ellipsometer is used to study the optical properties of thin films. Variation in the band gap, refractive index and extinction coefficient is studied as a function of temperature and varying aluminum content.

1. INTRODUCTION

For increasing energy demands to search renewable energy sources such as solar energy is the most promising sustainable energy resource (Reddy 2013). Silicon is the most important technology in the production of solar due its high efficiency, but due to its high cost researcher trying to find another technology to decrease the cost of the materials to fabricate the solar cell. Thin film can see as proper replacement with silicon technology due to less expensive, fewer materials used, less manufacturing process as they are made very thin film around 35-360 nm. The reason after this replacement is low cost manufacturing solar cell because its uses smaller amount of material and the deposited layer are much thinner than the other solar cells. But this technology based solar cells have lower efficiency. In thin film technology, CdS/CdTe and CIS have been

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much attention due to high efficiency but these materials have some adverse effects on environment so the researchers are trying to improve the efficiency of the solar cell by using different materials. Copper Indium Gallium Selenide is found better with other silicon technology (Tyagi 2013).

These days one of the main priorities in the photovoltaic solar energy conversion is to lower the production costs (Baek 2011, Kaigawa 2011) good shape ability, light weight (Chen 2012) while maintaining high efficiencies. In the field of the copper indium gallium diselenide (CIGS) based solar cells, CIGS is one of the best leading positions with reported highest efficiencies values of up to 20.3% (Rousset 2011, Seyrling 2011, Wang 2012).

Copper indium gallium selenide is a member of the I–III–VI_2 group of chalcopyrite semiconductors with large band gap (Garcia 2013). For recently developed, quaternary Cu (In, Ga) Se_2 is used as an absorber layer to increase both the conversion efficiency and voltage. CIGS has large absorption coefficient (10^5 cm^{-1}) and maximum solar absorption region (Liu 2011). For developing of high efficiency and low cost thin film solar cells, Cu (In, Ga) Se_2 is a promising material (Li 2012, Blosch 2012, Baji 2013). It has a variable band gap, which can be varied from 1.05eV ($CuInSe_2$) to1.68eV ($CuGaSe_2$) (Faraj 2012, Liu 2013).

Chalcopyrite thin films are typically prepared by different techniques such as vacuum evaporation, sputtering deposition, electrodeposition, electron beam deposition, molecular beam deposition, and selenization (Faraj 2012, Men 2012).But the method mostly used for deposition of chalcopyrite CIGS thin films is vacuum based evaporation to produced high efficiencies (Kaigawa 2011, Wu 2013).

In our research work, we prepared Copper Indium Gallium Aluminum Selenium Cu (In, Ga_{1-x}, Al_x) Se_2 by doping Aluminum in CIGS thin films to vary the band gap of material, which improves the voltage and efficiency of solar cells. After preparing Cu (In, Ga_{1-x}, Al_x) Se_2 thin films (x=0,0.2,0.4,0.6,0.8,1.0), the deposited thin films subsequently annealed for 15 minutes at 250˚C to improve the crystallinity of thin films.

2. Experimental Details

The thin films of Cu (In, Ga_{1-x}, Al_x) Se_2 were prepared by Sequential Elemental Layer (SEL) technique using resistive heating in the sequence of Cu-In-Ga/Al-Se. the films were deposited on glass substrate at room temperature. Before the deposition was carried out, the glass substrates were dipped in acetone and place in the HQ Kerry ultrasonic for 15 minutes, and then placed in isopropyl alcohol and put again in ultrasonic bath for 15 minutes to improve the adhesive properties and to remove any organic contamination.

In our experimental work, we used Edwards 306 coating unit to deposit the materials. To create vacuum in the work chamber for the deposition of thin film, a pumping system is used. High vacuum was achieved using rotary pump (10^{-3} Torr) and diffusion pump (10^{-6} Torr). Then the source was heated near the melting point of evaporant and the films were deposited after the source was thermally stabilized.

The films were also deposited onto single crystal Si with Aluminum back contact and glass substrate at room temperature, and this process is repeated for different concentration of x(x=0,0.2,0.4,0.6,0.8,1.0). After films deposited, annealing was performed at 250˚C temperature for 15 minutes in controlled atmosphere.

Structural properties of the films were carried out using Bruker D8 Advance X-ray Diffrcatometer operated using Copper Kα radiations (λ=1.5405Å). Optical properties of the films were studied using Variable Angle Spectroscopic Ellipsometer (VASE) at an angle of incidence of 45˚. Four probe measurements were carried out for studying the electrical properties at room temperature.

3. Results and Discussions

In experimental work, Cu (In, Al_x, Ga_{1-x}) Se_2 thin films with different composition(x=0, 0.2, 0.4, 0.6, 0.8, 1.0) were deposited by using evaporation techniques. The two important compounds used in solar cells are Cu(In,Ga)Se_2 and Cu(In,Al)Se_2. In this work, we introduced Aluminum in CIGS thin films first time with the sequence Copper, Indium, Gallium, Aluminum, Selenium. Addition of the Aluminum causes the variation in the band gap. For the conformation the films were characterized structurally and optically.

Crystal phase of all the samples were acquired by XRD using CuKα (λ=1.54060 A˚). XRD pattern correspond to CIGAS thin films shown in Fig. 1. CIGAS thin films exhibit strong chalcopyrite diffraction peaks of (213) for CIGS and (200) for CIAS. The identification of the various peaks present in different composition(x=0, 0.2, 0. 4, 0.6, 0.8, 1.0) was shown in Fig. 1 but the highest peaks of all compositions was investigated. .However, some minor peaks of all composition were also present in XRD pattern which belonged to CIGS, CIAS. It was observed that all the highest diffraction peaks of all composition were matched with JCPD card no.24-344, belonging to the CIAS and JCPD card no.40-1488 used to determent for CIGS peaks. For x=0.0 the XRD pattern show amorphous behavior whereas when aluminum was added to the compound the films show polycrystalline behavior. With increase in aluminum concentration the diffraction peaks shifts to slightly higher angle. No peaks corresponding to the oxides and aluminum can be seen for the entire concentration range studied indicating that aluminum is successfully incorporated into the host lattice.

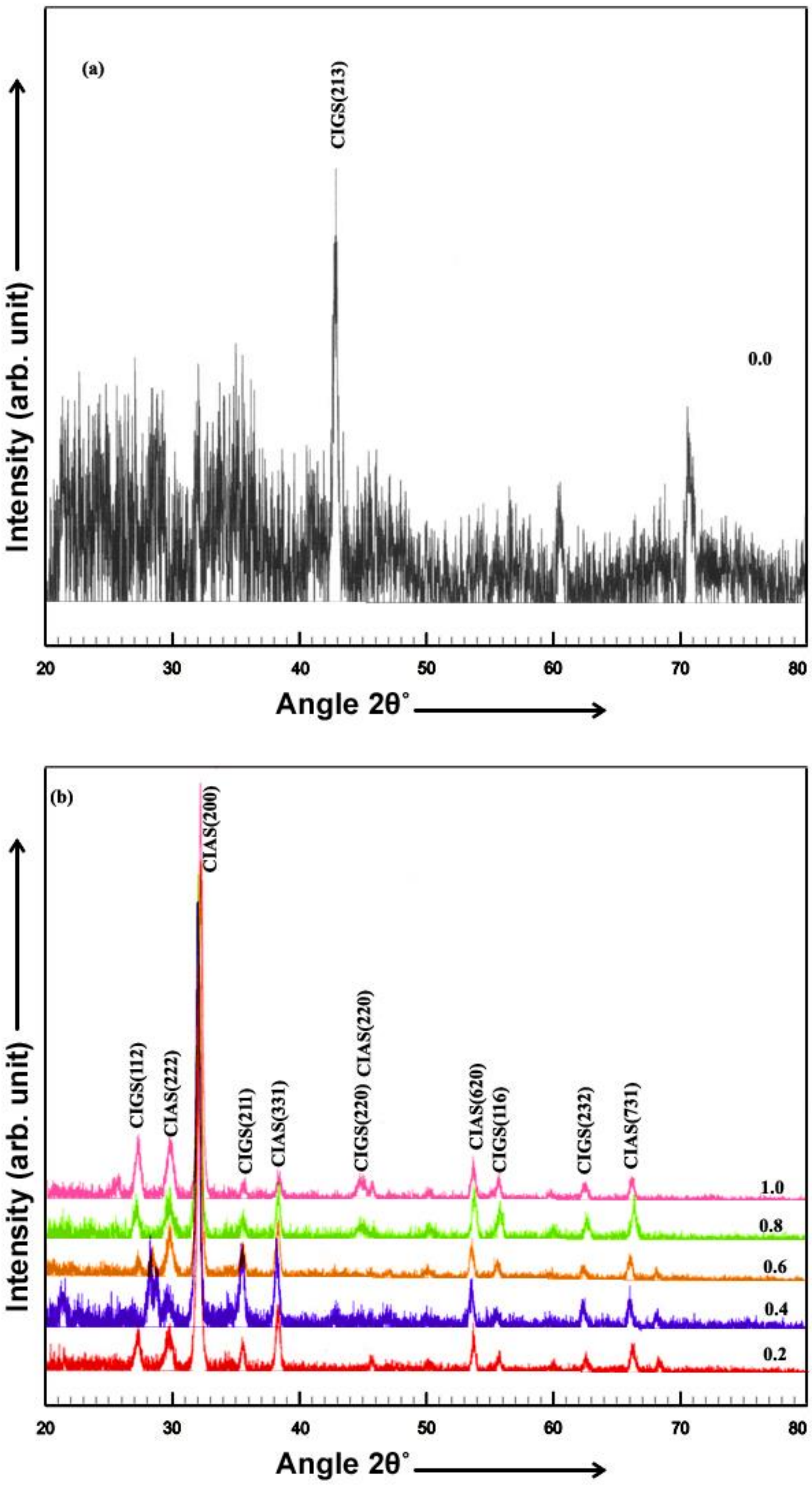

Fig. 1 XRD patterns of Cu (In, Al_x, Ga_{1-x}) Se_2 thin films

The crystallite size of all peaks of CIAGS films was determined using Scherer equation given in Eq. (1)

$$t = \frac{0.9\lambda}{B\cos\theta} \quad (1)$$

Where, t is represented crystallite size of material and B show the Full Width Half Maximum (FWHM).

For CIGAS at x=0, the highest diffraction peak of CIGS is observed at 42.68˚ 2θ value. According to JCPDS card, the highest peak appeared at the plane (213).The calculated value of grain size is 42.66nm. At x=0.2, highest peak of CIAS is observed at 32.04˚ (2θ) and the calculated grain size is 41.35nm. At x=0.4, the highest diffraction peak of CIAS observed at angle 2θ is 32.04˚ and grain size is 41.35nm. At x=0.6, the highest peak of CIAS observed at 32.04˚ 2θ value and the grain size is 41.35nm. At x=0.8, the highest peak of CIAS observed at 31.76˚ and the grain size is 41.31nm.At x=1.0, the highest diffraction peak of CIAS is observed at 32˚ 2θ value. The calculated value of grain size is 41.34nm. The main peak appears of all the composition between 32˚-42.68˚with slow changes of 2θ. The grain sizes are plotted as function of aluminum concentration in Fig. 2.

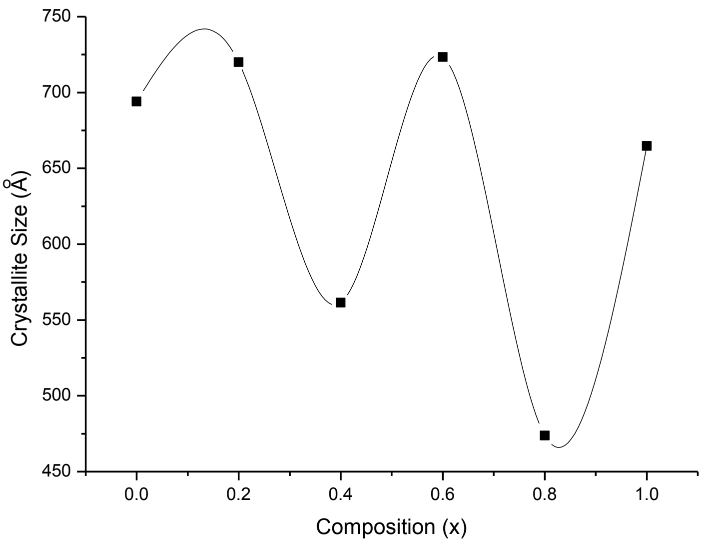

Fig. 2 Crystallite size as a function of aluminum concentration

In order to obtain polycrystalline CIAGS thin films, layers of different composition x=0, 0.2, 0.4, 0.6, 0.8, 1.0 were sequentially evaporated after addition of Aluminum in amorphous CIGS thin film. Polycrystalline CIAS thin films with chalcopyrite structure and orientation along the (200) plane have been obtained as was shown in XRD pattern All CIAGS thin films were characterized by optically obtaining their Transmission, Reflection and absorption.

Fig. 3 show absorption spectra of CIAGS films. The films are highly absorbing in the visible region and infrared region. As these films are further intneded to be utlized for solar cell applications for which high absorption fo the films is an importat requirement.

The high absorption of the films in our case indicate their potential use as an absorber layer in solar cells.

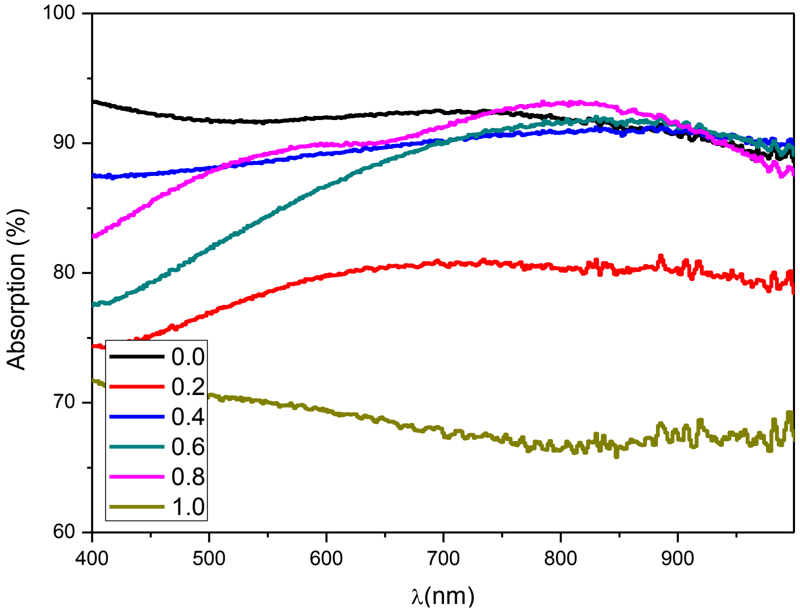

Fig. 3 Effect of aluminum incorporation on absorption of CIAGS films

The band gaps of the films are calculated using transmission curves and is plotted as a function of aluminum concentration in Fig. 4. It can be seen that with increaseing aluminum content the band gap decreases till x=0.6%. With further increas in aluminum content the band gap starts to increas again.

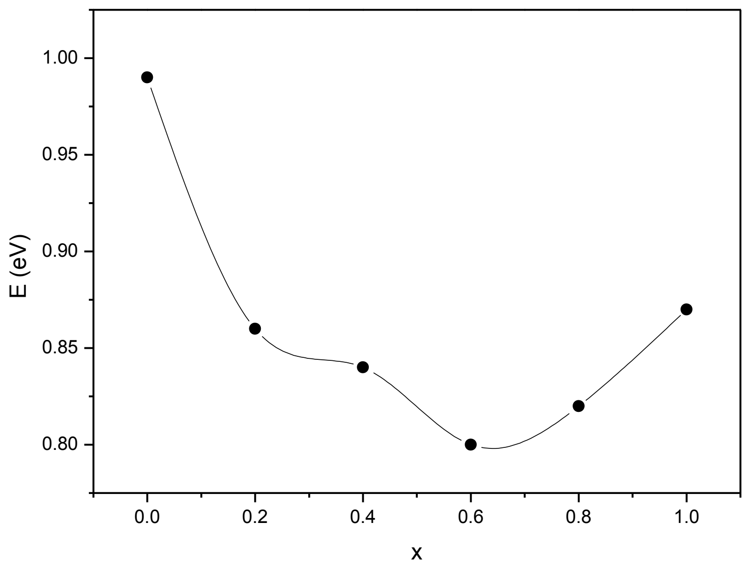

Fig. 4 Band gaps of Cu (In, Al_x, Ga_{1-x}) Se_2

For studying the refractive index and extinction coefficient Variable Angle Spectroscopic Ellipsometer was used operated at an angle of incidence of 65˚. Fig. 5 show refractive index and extinction coefficient for Cu(In,Ga_{1-x},Al_x)Se_2 films. High values of refractive index and extinction coefficient indicate that films are highly dense as well as highly absorbing in the wavelength range studied. Refractive index and extinction coefficients are also plotted as a function of aluminum content in Fig. 6 and it can be seen that as the aluminum concentration increases the refractive index as well as extinction coefficient increases.

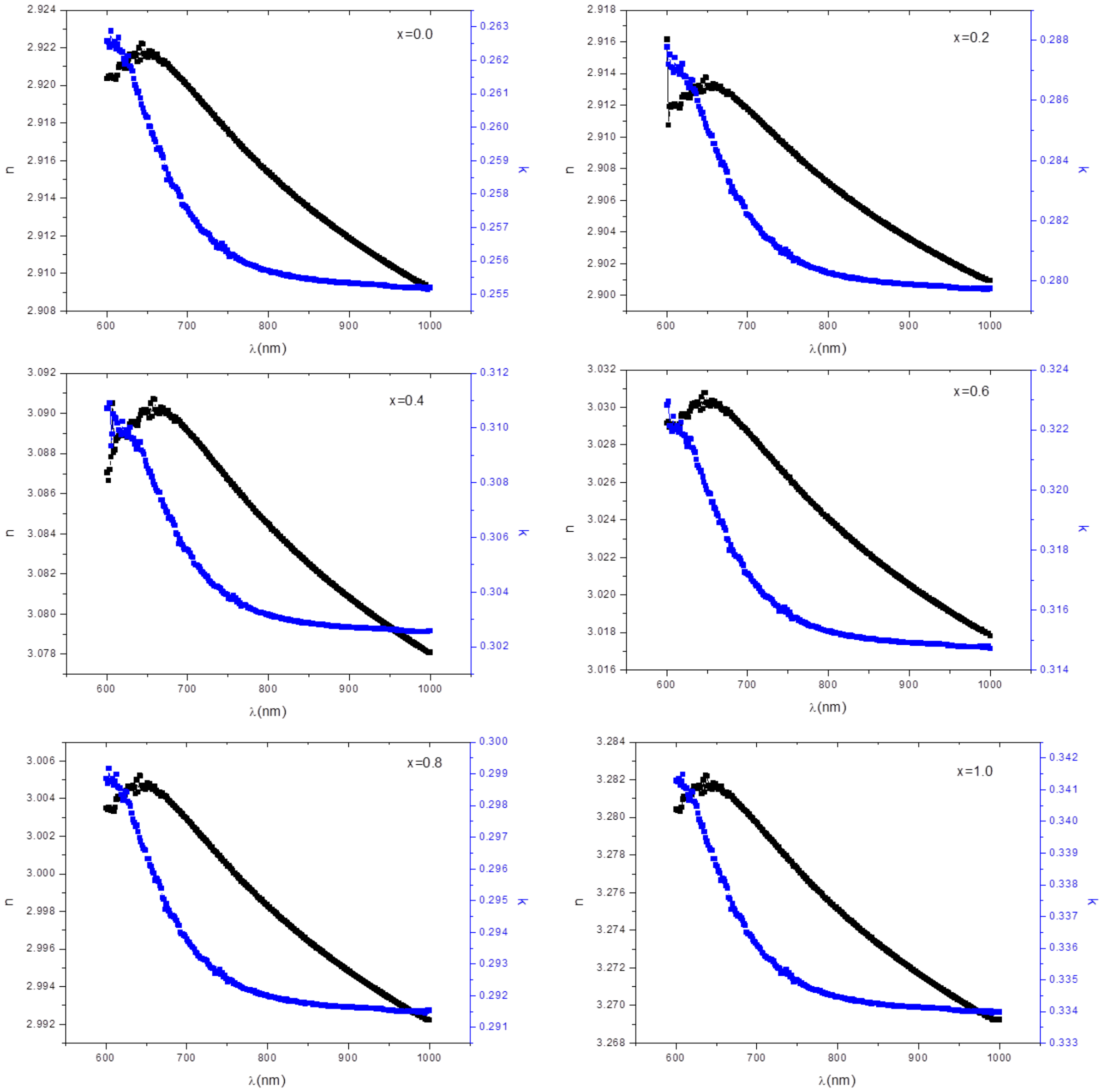

Fig. 5 Refractive index and extinction coefficients for Cu (In, Ga_{1-x}, Al_x) Se_2

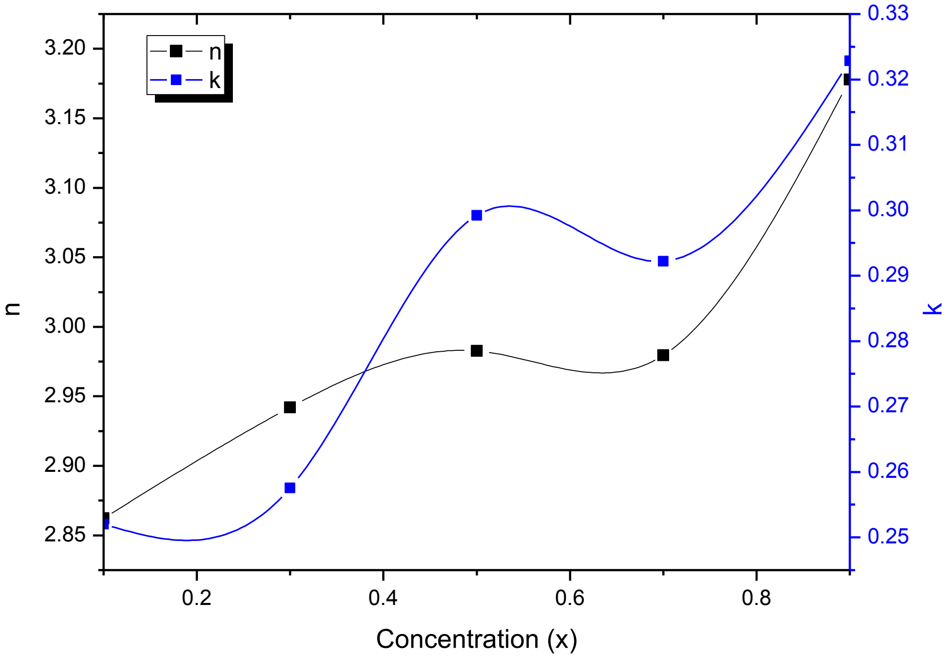

Fig. 6 Refractive index n and extinction coefficient k as a function of aluminum concentration

Conclusion

Structural and optical properties of a new compound material Cu (In, Al_x, Ga_{1-x}) Se_2 (CIAGS) have been investigated. CIAGS has been prepared by sequential elemental layer technique. XRD patterns show that addition of aluminum helps in achieving polycrystalline nature of the films. The films are highly absorbing with high values of refractive index and extinction coefficient. The band gap of the films decreases as the aluminum content in the films increases.

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