Thermoelectric properties of InSe and AlSe bilayer thin films

Sarita Boolchandani[a]*, Subodh Srivastava[b] & Y K Vijay[b]

[a]Department of Electronics & Communication, Vivekananda Global University, Jaipur 303 012, India
[b]Department of Physics, Vivekananda Global University, Jaipur 303 012, India

The indium selenide (InSe) and aluminum selenide (AlSe) bilayer thin films of different thickness ratio have been prepared using thermal evaporation at vacuum. Thermoelectric behaviour of each sample has been determined temperature regions of 300-310 K. The thermoelectric behaviour of prepared thin films has been found to improve when samples have been annealed in vacuum at 70 ºC and 100 ºC. Electrical studies have been carried out of each film before and after annealing.

Keywords: Indium selenide, Aluminum selenide, Bilayer thin films, Thermoelectric power

1 Introduction

III-VI group compound element semiconductors are of great importance because of their intrinsic vacancy structure and physical properties which make it attractive for application of electronic device, solar energy and thermal energy conversion to electrical energy[1-11].

Thermoelectric materials are the most attractive in waste heat conversion application as they can directly convert thermal energy to electricity. The direct conversion between electrical and thermal energy and also can be used for heating and cooling application which is based on Peltier-Seebeck effect is known as thermoelectricity[12,13]. A low thermal conductivity, large electrical conductivity and high Seebeck coefficient (S) are the requirement of high performance thermoelectric materials. The S as a function of temperature is one of the significant methods to analyze electronic properties of solids. It can be used to determine the concentration of majority carrier, type of semiconductor, fermi level position, etc.[14-17].

In the present work, a set-up is designed to measure Seebeck coefficient in the vacuum with variation in temperature of 300-310 K. The setup is very compact, low power and easy to operate. The S is measured by applying a temperature gradient in longitudinal direction. The distance between the thermocouples is 7 mm.

The molecular beam epitaxy, evaporation technique, chemical vapor deposition, electrochemical atomic layer epitaxy, thermal evaporation method, chemical bath deposition are some of the methods used for the growth of III–VI materials like aluminum selenide (AlSe) and indium selenide (InSe)[18-28].

In this paper, InSe and AlSe bilayer thin films having different thickness ratio, prepared using thermal vacuum evaporation method have been studied. In order to enhance the properties of thin films, annealing at 70 ºC and 100 ºC is done in vacuum. The structural properties are analyzed through scanning electron microscopy (SEM), the electrical properties are measured in terms of electrical conductivity and thermos electric power at room temperature.

2 Experimental

The samples of InSe and AlSe bilayer thin films of the same overall thickness (2500 Å) and different thickness ratio (In:Se/Al:Se=25:75, 50:50, 75:25) were deposited by the thermal evaporation method on a clean glass substrate under a vacuum of 10^{-6} mbar. The electrical and thermo-electrical power parameter measurements of the films were carried on to investigate the effect of annealing temperature and thickness ratio.

Pure aluminum tablets (99.99%) and selenium powder (99.99%) procured from Sigma-Aldrich and indium ingots (99.99%) from Alpha Aesear were used as source material for the evaporation. The materials were placed in molybdenum boats and tungsten

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*Corresponding author (E-mail: saritaboolchandani@gmail.com)

filament which were heated indirectly by passing high current through the electrodes. First, the selenium was deposited on aluminum contacts then aluminum/indium thin film was deposited to make a bi-layer structure through proper masking. The simple glass slides were used and were cleaned with soap water, distilled water and then with acetone. When high vacuum (10^{-6} mbar) is achieved in the vacuum chamber, slow current is applied to heat the material. The film deposition on the substrate was controlled through the source shutter. On optimization of all parameters (vacuum, evaporation rate, etc.), the source shutter was removed to start film deposition on the substrate. The evaporation rate was maintained at ~6 Å/s. The rate thickness and evaporation rate of the as-deposited films was monitored using quartz crystal monitor attached with the unit. To achieve the metastable equilibrium the deposited films were kept inside the vacuum deposition chamber for 24 h. The prepared bilayer thin film and its schematic diagram is shown in Fig. 1 (a,b).

Thermoelectric power measurement of prepared samples have been carried out in the temperature range 300-310 K by taking pure metallic copper (Cu) as reference metal. The thermoelectric module TEC1-12715 was used for temperature difference on the two ends sample. The module was sandwiched between two copper plates. Experiment set up used for sample testing is shown in Fig. 2. The chamber was pumped first by using a rotary pump to a rough vacuum of 10^{-3} Torr. Only one end of the sample was heated leading to the creation of a hot junction and a cold junction to create a temperature gradient on the sample.

The thermocouple used for measurement of the temperature gradient (dT) is of K type. The sample under investigation was kept on copper plates and the wires of the module were connected to a constant power supply to provide the voltage to the thermoelectric module in order to maintain the temperature difference. A multimeter was used to measure the voltage difference across the film. The Seebeck coefficient (S) was measured using the relation given below:

$$S = \frac{dV/dx}{dT/dx} \qquad \qquad \text{… (1)}$$

or, $S = \dfrac{dV}{dT}$ $\qquad \qquad$ … (2)

here dV is the voltage difference across the film, dT is the temperature gradient and dx is the distance between thermocouples.

3 Results and Discussion

3.1 Scanning electron microscope (SEM) characterization

The SEM images of InSe and AlSe bi-layer is shown in Fig. 3. Figure 3 shows that big cluster have appeared in spherical shape and are distributed on the entire surface which shows that thin films are granular in nature.

3.2 Electrical characteristics

Electrical conductivity was measured using a two-terminal configuration by applying a voltage (≈ 10 V) to the sample and measuring the current through it using a Keithley 2450 source meter. The measurements were carried out at room temperature and the variation of conductivity with different thickness ratio of InSe and AlSe thin films is shown in Fig. 4 (a,b).
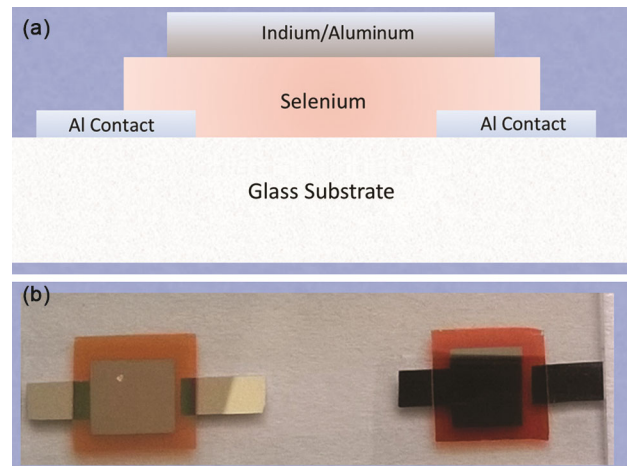

Fig. 1 — (a) Schematic representation of thin film structure and (b) prepared thin films.

Fig. 2 — Experimental setup for calculation of thermoelectric power.
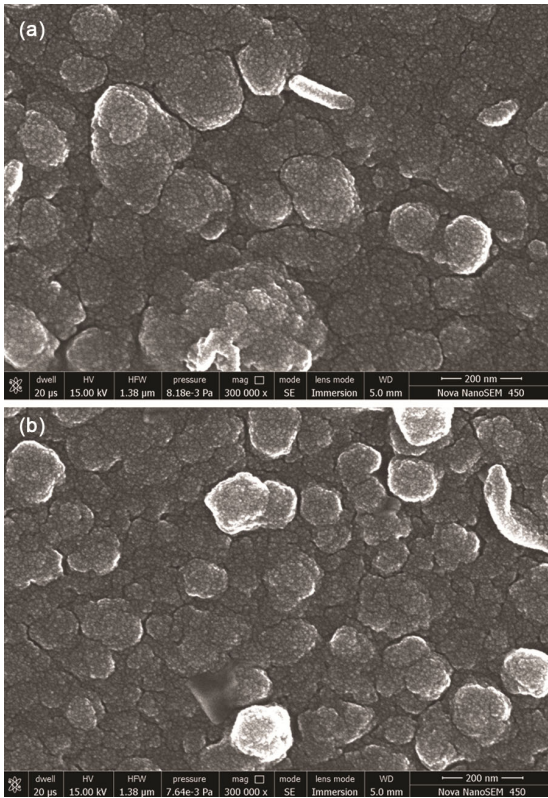
Fig. 3 — SEM images for 50:50 of (a) InSe and (b) AlSe.

Figure 4 (a,b) revealed that on increasing the In or Al concentration in InSe or AlSe bi-layer thin films, the room temperature film conductivity is increased in comparison to that of lower concentration. It may be due to the interaction of metal with the Se thin film layer, which leads to decrease in potential difference at the interface of bi-layer thin film which in turn, is expected due to crystalline structure of InSe and AlSe bi-layer thin films, so the electrical resistance of composite films decreases or conductivity increased.

3.3 Thermoelectric power

The thermoelectric power, *i.e.*, Seebeck coefficient (*S*) of each sample is calculated by keeping one end of the sample at hot junction while the other end to the cold junction. The temperature on the hot side is increased linearly and the thermo *emf* is noted. From the slope of thermo *emf* v/s ΔT graph *S* is measured. The thermoelctromotive force, *i.e.*, Seebeck coefficient of the prepared InSe and AlSe bilayer thin films with different thickness ratio and annealing temperature is shown in Fig. 5 to Fig. 8.

As shown in Figs 5 and 6 the increase of conductivity with decreasing Se ratio in composition indicates the semi-metallic behaviour. Also, from
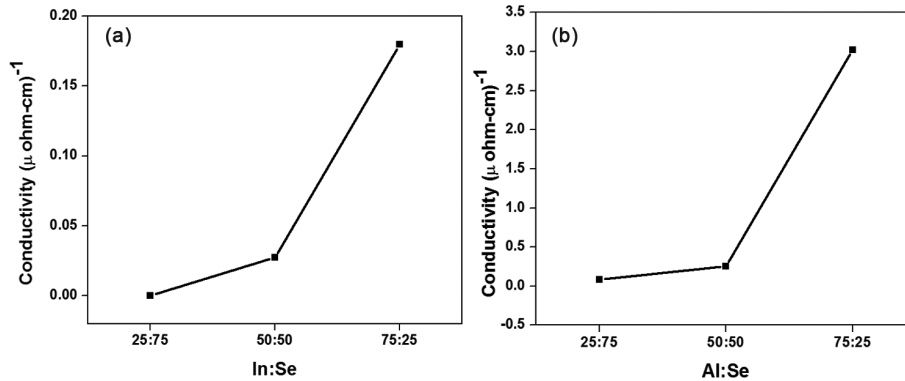

Fig. 4 — (a) Graph between conductivity and InSe ratio and (b) graph between conductivity and AlSe ratio.
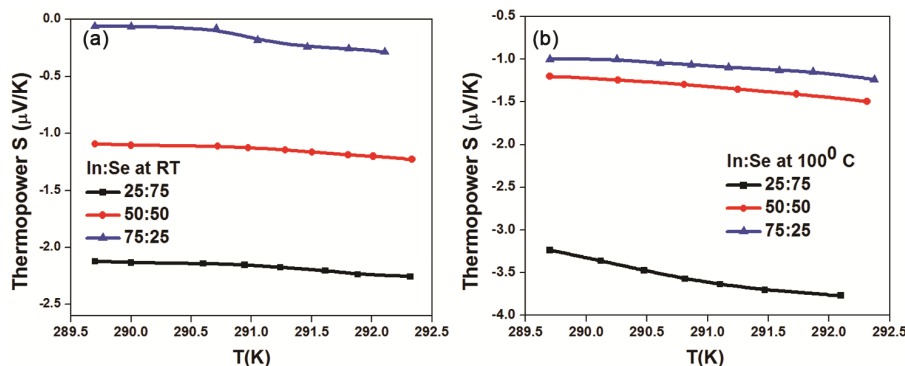

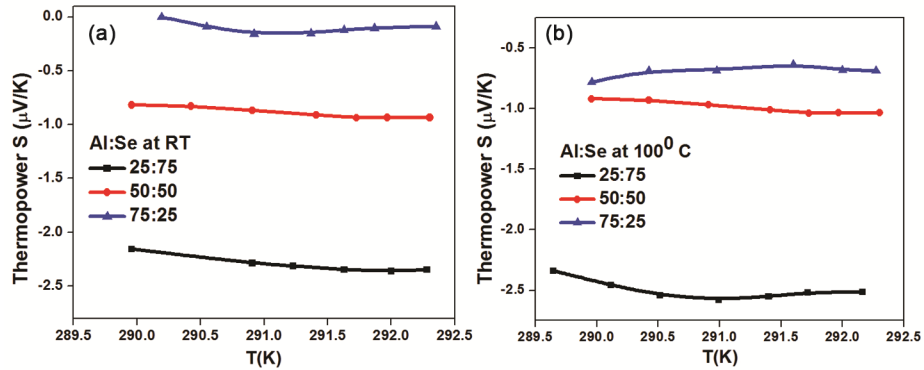Fig. 5 — Graph between thermoelectric power and temperature for InSe (a) RT and (b) 100 ºC.

Fig. 6 — Graph between thermoelectric power and temperature for AlSe (a) RT and (b) 100 ºC.
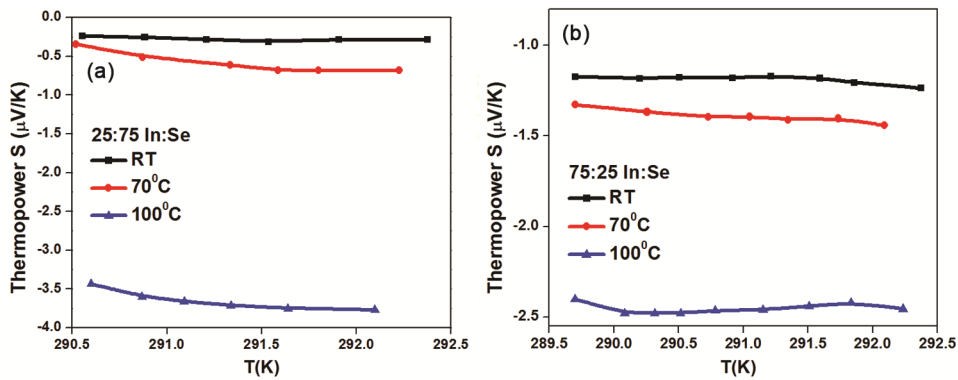

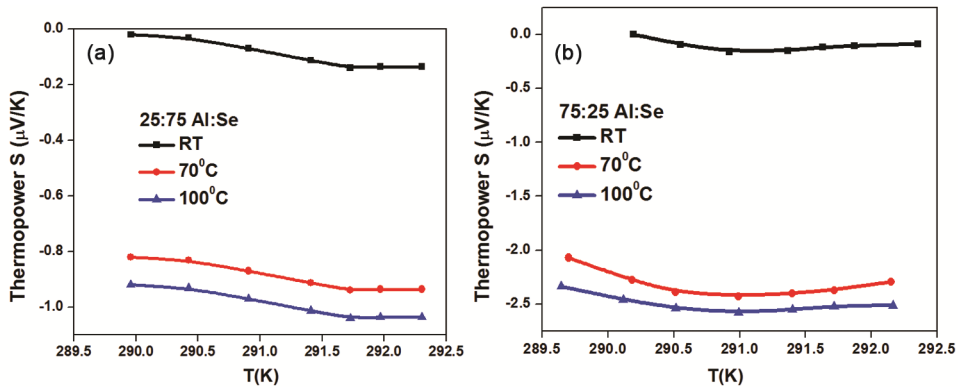Fig. 7 — Graph between thermoelectric power and temperature for InSe (a) 25:75 and (b) 75:25.

Fig. 8 — Graph between thermoelectric power and temperature for AlSe (a) 25:75 and (b) 75:25.

these figures it is cleared that as Se composition is increasing or metal (In/Al) composition is decreasing the S increases.

Seebeck coefficient increases with the increase in annealing temperature, *i.e.*, annealing improves the thermoelectric behaviour of the thin films is shown in Figs 7 and 8. This may be due to the better crystallinity and elimination of grain boundaries of the thin films at higher annealing temperatures.

The S of the deposited films has found to be negative, *i.e.*, electrons are predominant charge carriers which confirm the n-type behavior of Indium Selenide and Aluminum Selenide thin films[29].

4 Conclusions

Indium selenide and aluminum selenide bilayer thin films with different thickness ratio have been prepared by vacuum evaporation technique. The films were uniform and had good adherence to the substrate. SEM images show that the thin films are granular in nature. The increase in conductivity with increase in In: Se or Al:Se ratio in the films confirms the semiconducting

behavior of the InSe and AlSe films. The negative value of Seebeck coefficient confirms the *n*-type behaviour of deposited InSe and AlSe thin films.

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