

Optimization of composition of quaternary alloy-based film and effect of Annealing for phase change optical recording Material

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Abstract

There is ever-increasing demand to store and retrieve a large amount of information at a very fast rate. For such high-speed data transfer, optical recording media using a direct overwrite scheme is very popular. Various optical recording materials have been used for this technique but quaternary alloy-based thin films are the most promising material. AgInSbTe alloy-based films are the most popular material for optical data storage. In this paper, the results of Diffraction (XRD) & Scanning Electron Microscopy (SEM) analysis of as-deposited and annealed film of optical storage material $(\text{AgSbTe})_x(\text{In}_{1-y}\text{Sb}_y)_{1-x}$ has been presented. These films have higher linear densities and crystallization is dominated by growth which results in lower jitter. In this paper detailed analysis of the morphology of film surface with change in composition has been done. Also, the effect of annealing on surface morphology has been discussed. The results have been presented that show the crystallization of the film increases with an increase in annealing temperature. Furthermore, the selection of the most appropriate conditions for alloy-based thin film with optimized composition and annealing temperature to obtain good optical contrast that is more suitable for optical storage has been presented here.

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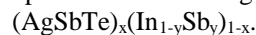
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I. Introduction

With the audio and video information available on the internet, the need for storing large amount of information has arisen. The area of optical recording has progressed rapidly. Read-only disc standards (such as CD-Audio and CD-ROM) have been supplemented by recordable (CD-R) and rewritable (CD-RW) technologies, allowing users to create customized CDs[1]. Storage of encyclopaedia, text message, pictures etc. requires fast and efficient archival storage. For large amount of digital storage and to increase the data transfer rate, high speed recording in phase change optical discs is becoming popular. Direct over write methods are more prevalent for high speed and high-density optical recording medium. For high speed CD-RW phase change chalcogenide materials are preferred, such as GeSbTe or AgInSbTe alloys. A compact disc is a optical storage medium with digital data recorded on it. Compact discs have three layers (dielectric layer, recording layer and heat sink layer)[2]. The crystallization process of the phase change layer or recording layer utilized in phase change optical discs is critical [3]. The mechanism used for phase change recording is based on transition of phase between amorphous and crystalline state of the recording material using a highly focused Laser beam. Once the Laser is switched off, the molten state cools down and get amorphous. Further for erasing the recorded mark, recrystallization is done using different low power Laser for a longer period of time. Recording of digital information is in the amorphous marks in-between crystalline spaces. For phase change recording media is sputter deposited on pre-grooved polycarbonate substrate. The groove structure acts as a gradient for the reflected beam and is used for radial tracking [2]. For groove only phase change recording formats, AgInSbTe is a preferred material. Such material has many advantages: high linear densities, lower jitter, and good optical contrast[4]. Lower jitter is due to the growth of crystalline edge towards the mark centre. This results in well-defined edges and good optical contrast [5].

Experimental Details

Quaternary alloy based thin films were developed for the following composition



Alloy Preparation

Bulk alloy of different composition was prepared by melt quenching technique[6]. For the typical composition as mentioned above the value of x chosen to be less than 0.5 and y =0.7. The constituent Elements were of 5N purity melted together in evacuated sealed quartz ampoules at a pressure of 10^{-6} torr. The ampoules

were kept in rocking furnace at about 1150⁰C for about 30 hours to ensure homogenization of the melt. The samples were then quenched in liquid nitrogen to ensure the amorphization of the alloy. Further, the quenched samples were removed from ampoules by keeping the ampoules in a mixer of HF for about 24 hours. Quartz gets dissolved in HF and grey colour alloy samples were obtained as reported earlier[7].

Film Preparation

Thermal evaporation technique was used to deposit films of prepared alloy with a thickness of approximately 100 nm on the glass substrate at a high vacuum of 10⁻⁶ torr. Glass and quartz slides were thoroughly cleaned before the films were deposited. The substrate is washed in a weak detergent solution to remove the detergent solution from the slides, followed by a hot water spray rinse and an overflow rinse. The organic material is then converted to water soluble compounds using a boiling solution hydrogen peroxide (Teepol). Finally, the substrate is immersed in hot distilled water and then soaked in De-ionised water for about 15 minutes to remove the peroxide layer. The substrate is then dried before being put into a vacuum system for film deposition. Various films were prepared with different compositions of 'x' (x = 0.2, 0.3, and 0.4) and y = 0.7. ESCA750 (Shimadzu corporation, Japan) was used to verify the composition of as-deposited films. The compositional variation was on the order of 2% when compared to the original values of component elements.

II. Result and Discussion

XRD Analysis

Due to its four element alloy characteristics and lack of stoichiometric composition different crystalline phases such as Sb, AgInTe₂, AgSbTe₂, found to co-exist after annealing [8]. It has been found that crystalline phases formed after thermal annealing depends on the alloy composition and annealing temperature. The film so prepared were analysed through XRD and it was found to amorphous in nature. Thermal annealing of as-deposited film was done at different temperature range from 200⁰C to 400⁰C (200, 300 & 400⁰C) through radiant heating under the vacuum of 10⁻⁵ torr for one hour. The analysis of the annealed films was again carried out and XRD trace the evolution of crystallization after heat treatment of films [8]. It has been observed that the crystalline phases formed after thermal annealing depends on the alloy composition as well as annealing temperature. As reported earlier [8], for each composition of x, it is observed that AgInTe₂ peak intensity continues to increase with increasing annealing temperature up to 400⁰C. However peak intensity of Sb decreases with decreasing composition of x and increasing annealing temperature. It has been observed that Sb phase almost disappears for x=0.2, annealed at 400⁰C, it shows that Sb phase may have decomposed at higher temperature. Based on the result obtained earlier[9] we have analysed only x=0.2 and y=0.7 composition. In the fig.1 we have shown the XRD graphs for x=0.2, y=0.7 composition for different annealing temperatures and as-deposited films. We have further calculated different parameters like interlayer spacing, crystallite size, strain and dislocation density with the help of published work [10][11][12], as listed in table 1.

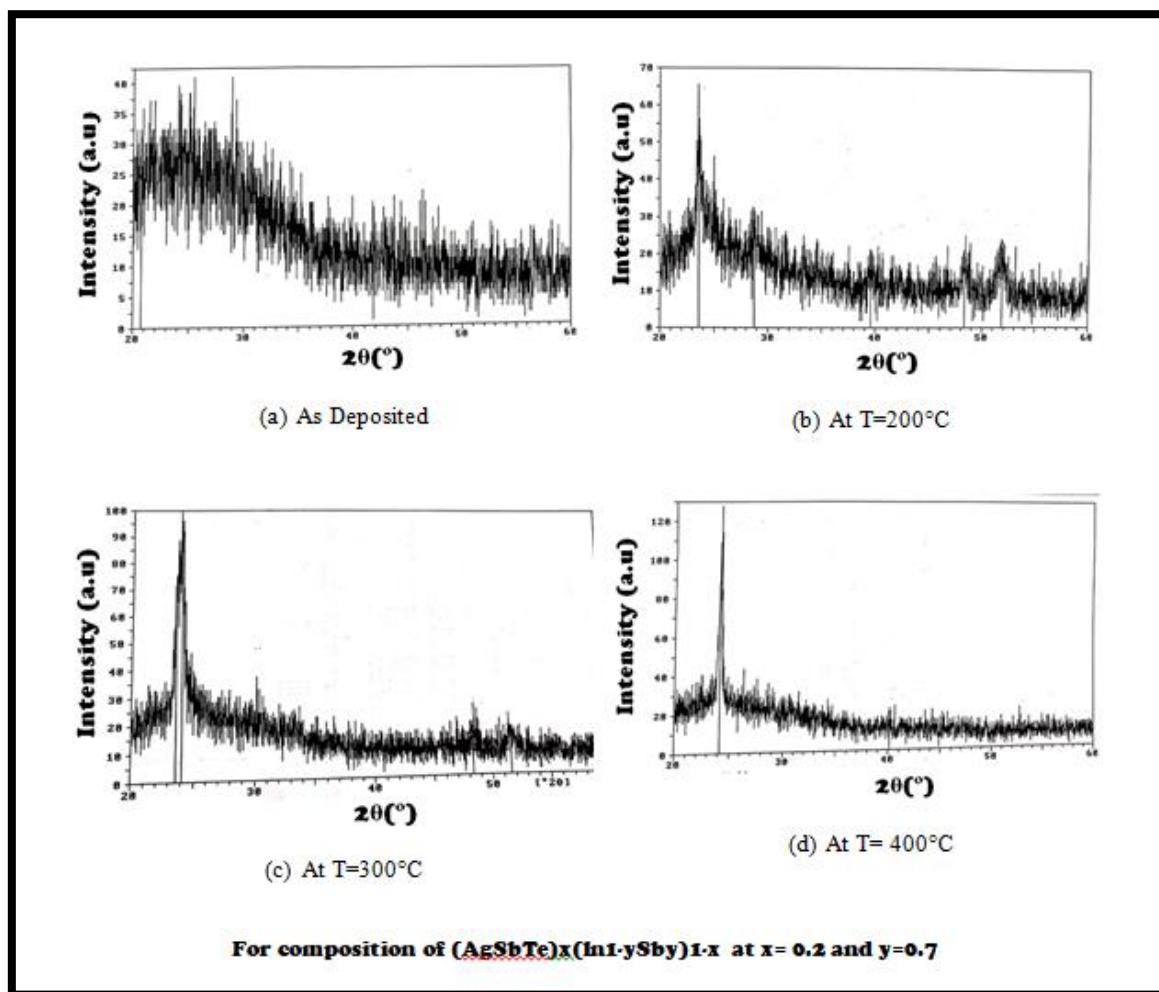


Fig1. Showing the XRD graphs of composition at different annealing temperature

Annealing temperature	200 °C	300 °C	400 °C
Interlayer spacing (Å)	3.767	3.693	3.691
FWHM (radians)	0.035	0.014	0.010
Crystallite size (Å)	40.58	101.58	135.39
Strain	-0.018	-0.007	-0.005
Dislocation density ($\times 10^{11} \text{ cm}^{-2}$)	60.72	9.69	5.45

Table1: Showing various XRD parameters of composition at different annealing temperature

The crystallite size as well as crystallinity increasing with increase in annealing temperature. All films are compressively strained if has a negative value. The average strain and dislocation density values in films decrease as the annealing temperature rises. This type of microstrain change is associated to the crystallization process in polycrystalline thin films[13].

SEM Analysis

The change in the morphology of the as-deposited and annealed films for various temperature of $(\text{AgSbTe})_x(\text{In}_{1-y}\text{Sb}_y)_{1-x}$ film, with $x=0.2$ and $y=0.7$, was studied using Scanning Electron Microscopy (SEM). SEM analysis clearly shows the change in the grain size with change in annealing temperature. The detailed SEM analysis of various films has been presented here as shown in fig.2. For a good phase change recording material, the amorphous and crystalline state should have good optical contrast at recording wavelength. This was observed in SEM images where it clearly shows the grains size changes as the annealing is done and clearly shows the crystalline spots with good optical contrast. This shows the change of phase from amorphous to crystalline with increase in the annealing temperature.

It can be seen from the morphology of the film that as-deposited film has irregular shaped structures which are not explicit. However, as samples are annealed the crystalline grains are more explicit and rounder in

shape and large crystal structures with good optical contrast. In table2, we have calculated the grain size with rise in annealing temperature from the SEM images, it is rising from 0.1905 to 0.583 μm with increase in the annealing temperature from room temperature to 400 $^{\circ}\text{C}$

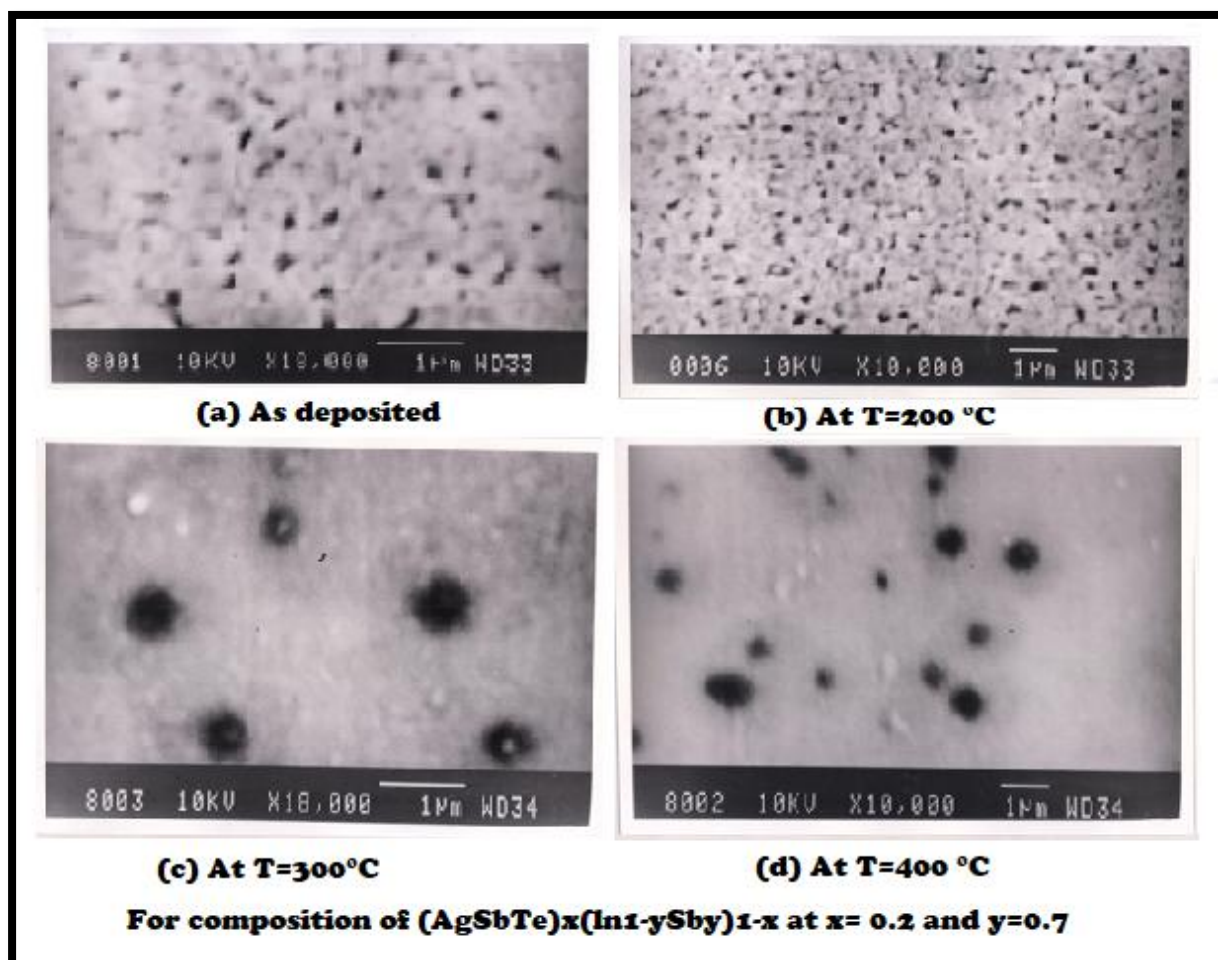


Fig.2: Showing the surface morphology of films at different annealing temperature

Annealing Temperature($^{\circ}\text{C}$)	Grain Size(μm) $x = 0.2, y = 0.7$
As-Deposited	0.1905
200	0.25
300	0.50
400	0.583

Table2: showing the grain size of films at different annealing temperature

III. Conclusion

The quaternary chalcogenide alloy-based films were prepared with $(\text{AgSbTe})_x(\text{In}_{1-y}\text{Sb}_y)_{1-x}$ typical composition, where $x=0.2$ and $y=0.7$ using melt quenching technique. It can be seen from XRD analysis, Table1, that the interlayer spacing is not affected much, however, FWHM and dislocation density are decreasing with rise in annealing temperature. Decreased in FWHM implies that as the annealing temperature rises, peak is getting sharper which means change of phase from amorphous to crystalline. This is also supported by crystallite size and strain values. Increase in crystalline size implies more crystallization with increase in annealing temperature. The results were further verified using SEM. SEM analysis shows the increase in the grain size with increasing temperature. Above analysis clearly shows that as the annealing temperature is increased the material gets crystalline and optical contrast is clearly visible in Fig2. Since the crystallites are clearer and sharper at higher temperatures. It is attributed to the growth dominated process in chalcogenide-based films. Due to this the phase change is faster also the growth starts from the edges to the mark hence good optical contrast is achieved.

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