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Optical properties of Se or S-doped hydrogenated amorphous silicon thin films with annealing temperature and dopant concentration

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ABSTRACT

We report the effects of the thermal annealing and dopant concentration on the optical properties of Se or S-doped hydrogenated amorphous silicon thin films. The Se and S-doped a-Si:H (a-Si,Se:H and a-Si,S:H) thin films were prepared by glow discharge plasma enhanced chemical vapor deposition (GD-PECVD) on 7059 corning glass. The films were subsequently annealed in vacuum in the temperature range from 100 to 500 °C. Influence of doping and annealing was examined by means of optical transmission spectroscopy of the films in the wavelength range of 300–1100 nm taken at room temperature. The absorption coefficients and refractive indices decreased as the annealing temperature increased from 100 to 300 °C and then increased again as the annealing temperature further increased to 500 °C, while the highest bandgap was observed at 300 °C for all of the samples. For a given dopant concentration bandgap was observed to be higher in a-Si,S:H than a-Si,Se:H thin films.

1. Introduction

For the past few years hydrogenated amorphous silicon (a-Si:H) thin films have been the subject of extensive studies due to their efficient optical and electrical properties, which allow them to be used in a variety of fields, such as photovoltaic solar cells [1–4], large area arrays of electronic devices [5], photosensors for the detection of bio-molecules [6,7], micro-electro-mechanical systems (MEMSs) [8,9], gas sensors [10,11], pixel detectors for high energy particles [12,13], optical imaging [14], photodetectors [15] and so on. Hydrogenated amorphous silicon thin films and its alloys are promising candidates for the realization of advanced opto-electronic devices. However, the light-induced degradation of a-Si:H remains one of the major challenges for the application in modern technological devices, especially in the field of photovoltaics [16,17]. The photodegradation behavior was found to be suppressed by the inclusion of hydrogen during the growth of the thin films by heat treatment in vacuum and other environments [18–21]. Mandal et al. also investigated the reduction in the photodegradation of doped and undoped a-Si:H after coating it with a polymer (polystyrene) [22].

The advantage of using a-Si:H is that it allows materials to be deposited inexpensively over large areas. The performance and light-soaking behavior of hydrogenated amorphous silicon (a-Si:H) solar cells with absorber layers deposited under non-constant silane concentration, increased open-circuit voltage that is up to 40 mV higher than that of a conventional amorphous silicon solar cell at initial efficiencies above 9% [23,24]. Apart from solar cells, most studies on devices have focused on thin film transistors in which the gate metal is coated with an insulator, usually hydrogenated amorphous silicon nitride or silicon dioxide [25]. The doping of hydrogenated amorphous silicon with tetrahedrally coordinated elements such as C or Ge and halogens such as F or

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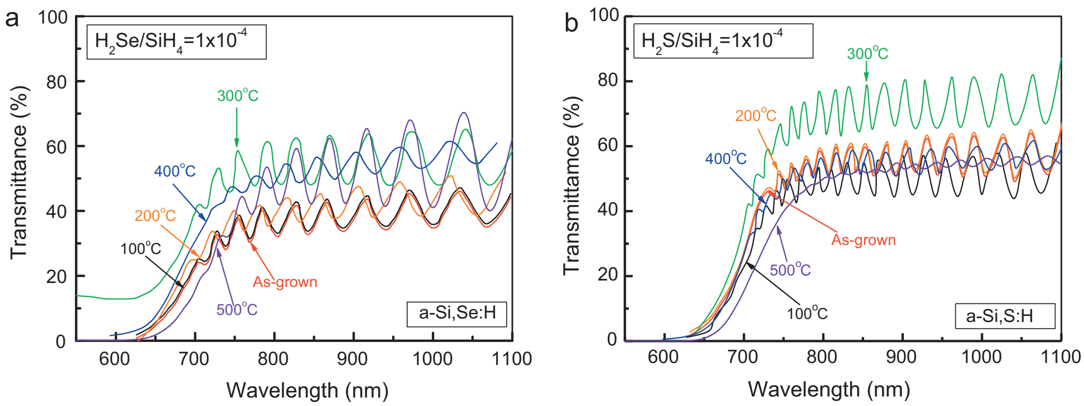

Fig. 1. Typical transmission spectra of Se and S-doped hydrogenated amorphous silicon (a-Si,Se:H and a-Si,S:H) thin films in the wavelength range from 600 to 1100 nm for annealing temperatures in the range from 100 to 500 °C (a) $H_2Se/SiH_4 = 1 \times 10^{-4}$ and (b) $H_2S/SiH_4 = 1 \times 10^{-4}$.

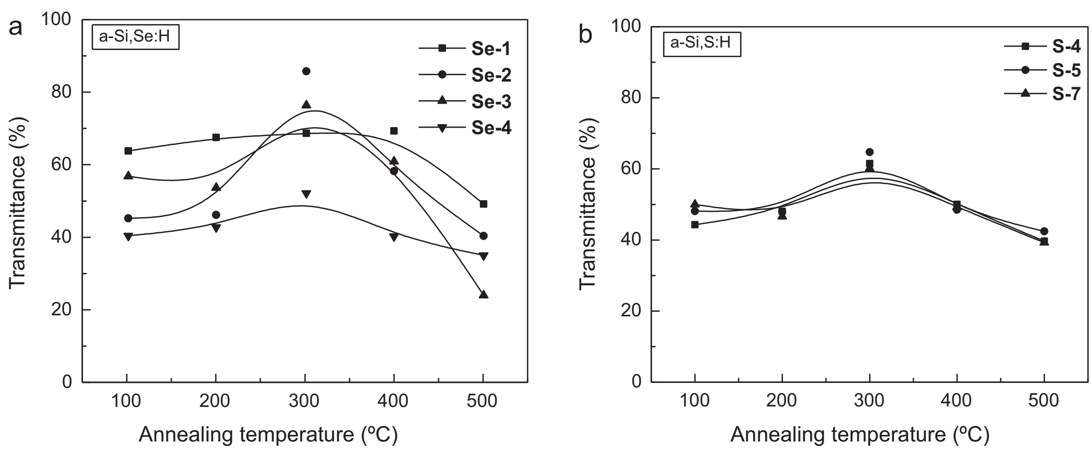

Fig. 2. Transmittance (%) versus annealing temperature (°C) for (a) a-Si,Se:H thin films prepared using different doping gas ratios, Se-1($H_2Se/SiH_4 = 1 \times 10^{-1}$), Se-2 ($H_2Se/SiH_4 = 1 \times 10^{-2}$); Se-3 ($H_2Se/SiH_4 = 1 \times 10^{-3}$); Se-4 ($H_2Se/SiH_4 = 1 \times 10^{-4}$); and (b) a-Si,S:H thin films prepared using different doping ratios S-4 ($H_2S/SiH_4 = 1 \times 10^{-4}$); S-5 ($H_2S/SiH_4 = 1.1 \times 10^{-5}$); S-7 ($H_2S/SiH_4 = 6.8 \times 10^{-7}$).

Cl has been studied by several groups [26,27]. The a-$Si_{1-x}C_x$:H films are generally obtained by PECVD, through the decomposition of silane (SiH_4) or disilane (Si_2H_6) and a hydrocarbon gas (CH_4, C_2H_4, C_2H_6, etc.). Films can also be formed using gases that have both Si and C atoms, i.e., di-, tri- and tetra-silylmethane, $C(SiH_3)_4$, tetra-methylsilane, $Si(CH_3)_4$, and others. So far, carbon doped hydrogenated amorphous silicon (a-Si,C:H) has emerged as a leading contender among the large bandgap alloys used in amorphous silicon based photovoltaic devices by serving as a light absorbing window layer in multi-junction cells [28–30]. However, the problems of long-term stability and efficiency of this material have not been solved. The major cause of this instability is photoinduced degradation of a-Si:H thin films due to light-induced metastable Si dangling-bond defects [31–36].

It is documented that prolonged illumination of a-Si:H with bandgap light reduces photoconductivity [16]. It has been suggested that light-induced reduction in photoconductivity may be caused by the creation of metastable recombination centers in the

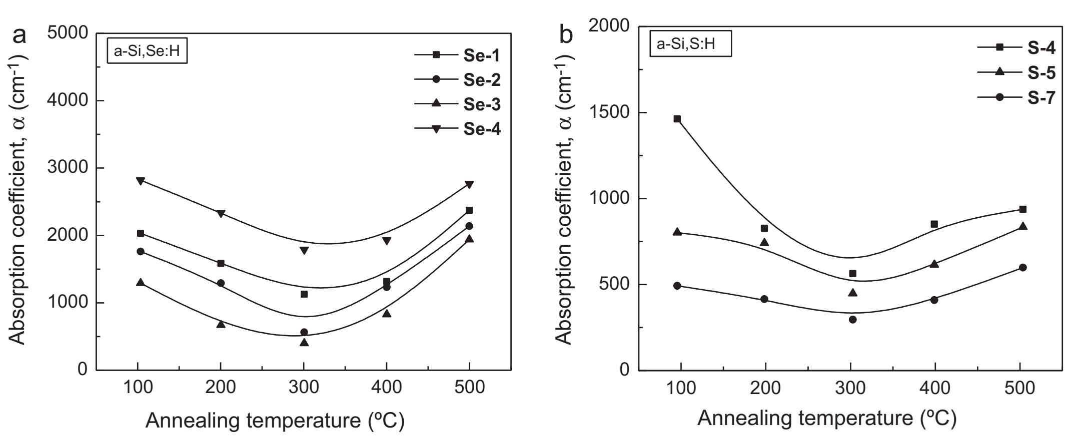

Fig. 3. Variation of absorption coefficient with annealing temperature at a wavelength of 750 nm for (a) a-Si,Se:H thin films; (b) a-Si,S:H thin films.

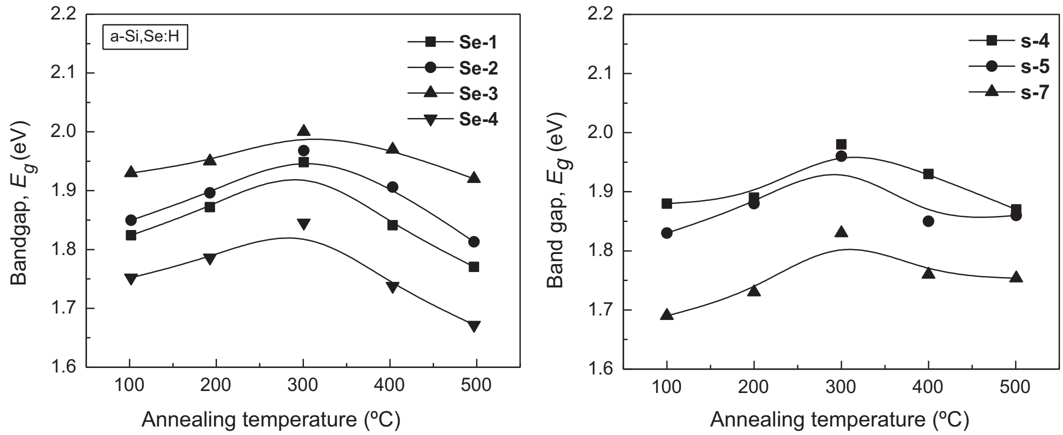

Fig. 4. Variation of bandgap with annealing temperature at a wavelength of 750 nm for (a) a-Si,Se:H thin films and (b) a-Si,S:H thin films.

gap. This behavior can be a limiting factor in optoelectronic applications of a-Si:H. The doping of double donors of group VI elements such as Se or S in a-Si:H showed significant reduction in the degradation of photoconductivity [37–39]. Lately, Sharma et al. have investigated the effect of Se or S doping on a-Si:H and studied the change in conduction mechanism with temperature [40,41]. Use of Se and S as dopants in films grown by capacitively coupled RF glow discharge decomposition of silane (SiH_4), hydrogen sulfide (H_2S) and hydrogen selenide (H_2Se) diluted in helium (He) results in silicon dangling bonds being terminated in the same way as hydrogen [42]. Hydrogenated amorphous silicon is the basis of an expanding large-area-electronics industry, which started with solar cells for consumer electronic devices and today covers an expanding number of applications, as mentioned above. The effect of the composition and annealing on the optical properties of Se or S-doped a-Si:H should be considered when evaluating their operational characteristics.

Thermal annealing has been found to reverse many of the light-induced features in the infrared absorption spectrum of a-Si:H. Annealing studies have emerged as an important tool that provide insight into the various material properties, especially in a-Si:H [43], a-Si,C:H [44], amorphous silicon-rich nitride films [45], etc. Annealing conditions and dopant concentrations influence optical constants of semiconductor thin films. This is necessary to precisely model spectral response for electro-optical devices. The electrical transport in Se or S-doped a-Si:H is sensitive to doping concentration. Doping can also have an effect on the temperature dependence of dc dark and photoconductivity [39,40]. To the best of our knowledge, there is no paper in the literature reporting the comparison of optical properties of Se or S-doped a-Si:H thin films under annealing conditions as well as dopant concentrations.

The main objective of this study is to estimate the effect of dopant concentrations and annealing temperature on the optical properties of Se and S-doped hydrogenated amorphous silicon thin films deposited on corning 7059 glass at 230 °C. In particular, we attempted to find the optimum conditions under which high efficiency recovery of the degraded amorphous silicon could be achieved. The films with different gas ratios were subsequently annealed in vacuum at temperature ranging from 100 °C to 500 °C. The optical transmission spectra were taken immediately after heat treatment and the optical constants, such as bandgap, refractive indices, absorption coefficient and dielectric constant, were calculated and compared with those of un-annealed films taken at room temperature.

2. Experimental

The a-Si,Se:H and a-Si,S:H thin films were prepared in an RF plasma glow discharge (13.56 MHz) using plasma enhanced chemical vapor deposition (PECVD) by the decomposition of H_2Se and H_2S vapors mixed with silane gas (SiH_4) on 7059 corning glass at a substrate deposition temperature of 230 °C, respectively [41,42]. The thicknesses of the films measured by a surface profiler (Dektak stylus–Veeco Instruments Inc.) ranged from 0.24 to 5.44 μm. The absence of sharp peaks in the X-ray diffraction (XRD Phillips-Holland Diffractometer, model X pert PRO) patterns confirmed the amorphous nature of the films. The details of the samples used for the study of the optical properties are given in Table 1. The selenium and sulfur concentrations (Se/Si and S/Si) increased with increasing doping gas ratios (H_2Se/SiH_4 and H_2S/SiH_4). After depositing a-SI:H thin films, all the samples were taken out of deposition chamber and annealed in a microprocessor controlled vacuum fur-

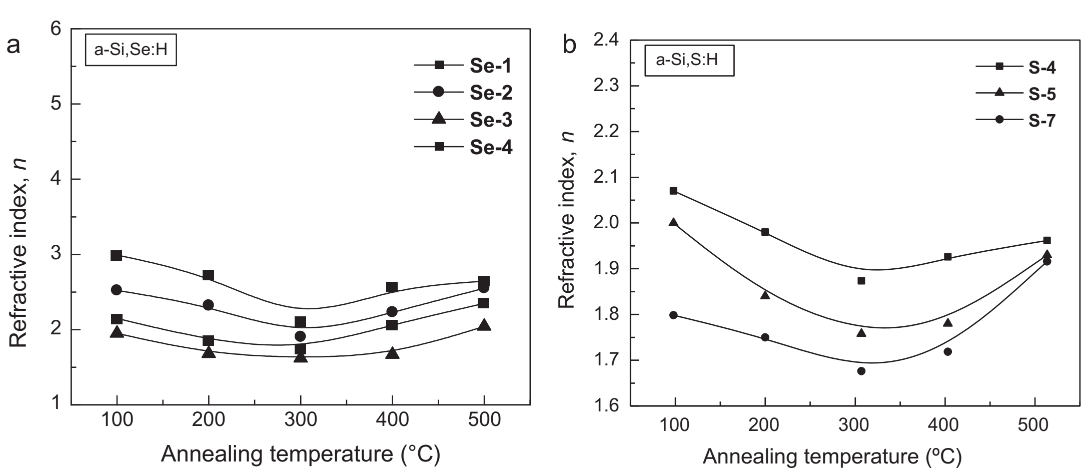

Fig. 5. Variation of refractive index with annealing temperature at a wavelength of 750 nm for (a) a-Si,Se:H thin films and (b) a-Si,S:H thin films.

Table 1
The abbreviation of the samples of Se and S-doped a-Si:H thin films used for the study of the optical properties as a function of the doping gas ratios (H_2Se/SiH_4 and H_2S/SiH_4) with respect to the Se/Si and S/Si concentrations of the films.

	Sample	H_2Se/SiH_4 (doping gas ratio)	Se/Si (doping concentrations)
a-Si,Se:H	Se-1	1.0×10^{-1}	2.50×10^{-1}
	Se-2	1.0×10^{-2}	2.01×10^{-2}
	Se-3	1.0×10^{-3}	2.18×10^{-3}
	Se-4	1.0×10^{-4}	1.60×10^{-4}
	Sample	(H_2S/SiH_4)	S/Si
a-Se,S:H	S-4	1.0×10^{-4}	1.20×10^{-3}
	S-5	1.1×10^{-5}	1.80×10^{-4}
	S-6	6.8×10^{-7}	1.70×10^{-6}

nace at temperatures ranging from 100 °C to 500 °C in a vacuum (~2×10^{-6} torr) for an hour. The transmittance responses of these samples were measured after each annealing. The optical transmission measurements of the Se and S-doped a-Si:H thin films were obtained using a Shimadzu spectrophotometer (3700 UV–VIS–NIR) with a spectral bandwidth of less than 1.8 nm (resolution of ±0.5 nm, photometric accuracy of ±0.0005 absorbance units and repeatability of 0.2 nm).

3. Results and discussion

Fig. 1(a) and (b) shows the typical transmission spectra for a-Si,Se:H (doping gas ratio of $H_2Se/SiH_4 = 1 \times 10^{-4}$) and a-Si,S:H (doping gas ratio of $H_2S/SiH_4 = 1 \times 10^{-4}$) films, respectively. The variations of the transmittance with the annealing temperature, as well as the doping concentration, at a typical wavelength of ~750 nm for the Se and S-doped a-Si:H are shown in Fig. 2(a) and (b), respectively. The maximum transmittance was observed at an annealing temperature of 300 °C for most of the samples. The optical transmittance increased as the annealing temperature increased from 100 to 300 °C. Thereafter, the transmittance decreased as the annealing temperature further increased to 500 °C.

The absorption coefficient, α, was calculated with an accuracy of ±2% in the weak and medium absorption regions using the relation [46–48]:

$$\alpha = \frac{1}{d}\left[\ln\frac{1}{T}\right]$$

where d is the thickness of the films and T is the transmittance. Fig. 3(a) and (b) shows the variations of the absorption coefficients versus annealing temperature of the Se and S-doped a-Si:H thin films, respectively. The absorption coefficients were also varied with doping concentrations. The value of α decreased with increasing annealing temperature from 100 to 300 °C and then decreased as the annealing temperature further increased up to 500 °C. In the case of Se-doped a-Si:H films, for a given annealing temperature, α decreased with increasing doping concentration from $H_2Se/SiH_4 = 10^{-4}$ to 10^{-3} and then increased as the doping concentration increased from $H_2Se/SiH_4 = 10^{-3}$ to 10^{-1}. While in the case of S-doped a-Si:H, α increased with increasing doping concentration from $H_2S/SiH_4 = 6.8 \times 10^{-7}$ to 10^{-4} at a given annealing temperature. However, the absorption coefficients of the Se-doped a-Si:H thin films were observed to be approximately 1 order of magnitude higher than those of the S-doped a-Si:H thin films.

The optical bandgap of the Se and S-doped a-Si:H films was calculated using Tauc's relation [42,48]:

$$\alpha = \left(\frac{B}{h\nu}\right)(h\nu - E_g)^{\gamma}$$

where B is the proportionality constant, $h\nu$ is the photon energy, and $\gamma = 2$ for an indirect bandgap which depends on the electronic transitions (energy-momentum). Fig. 4(a) and (b) shows the variations of the bandgap versus annealing temperature for the Se and S-doped a-Si:H thin films, respectively. The value of the bandgap was observed to be the highest at an annealing temperature of 300 °C for both types of film and for all doping concentrations. From these figures, it can be seen that the bandgap of the a-Si,S:H and a-Si,Se:H films varies with the doping gas ratios, as well as with the annealing temperature. For example, for a gas ratio of H_2Se/SiH_4 and $H_2S/SiH_4 = 1 \times 10^{-4}$ at an annealing temperature of 300 °C, the bandgap was observed to be 1.85 eV in a-Si,Se:H films and 1.97 eV in a-Si,S:H films, respectively. A smaller bandgap was observed in the case of the a-Si,Se:H films with a similar gas ratio, this may be because of larger binding energy of the Si–S bond than that of the Si–Se bond [49,50].

The optical absorption measurements show that the incorporation of Se and S into a-Si:H films increases the bandgap of the material and results in increased disorder and higher defect densities [42]. It might be expected that the annealing would remove many of the defects during sample preparation, which are responsible for the performances of the devices [51]. Decrease of the bandgap in films, when subjected to annealing above 300 °C, shows the optimum shift. This optimum shift can also be explained based on the formation of nano-clusters of Si–Se [43].

The refractive indices, n, in the weak and medium absorption regions for all of the a-Si,Se:H and a-Si,S:H films were calculated with an accuracy of 1–2% using the relation [46].

$n = [M + (M^2 - s^2)^{1/2}]^{1/2}$ where, $M = ((s^2 + 1/2)) + 2s((T_M - T_m)/(T_M T_m))$ and T_M and T_m are the values of the maximum and minimum transmission at a particular wavelength, respectively. Envelopes were drawn through the maxima and minima of the transmittance curve, and calculated based on the assumption that these envelopes correspond to the transmittance of integral half wave and quarter wave thickness. These fringes are used to calculate the refractive index. Refractive index of the substrate was determined using the transmission spectrum of the substrate alone and was observed to be 1.51. The variations of the refractive indices versus annealing temperature of the Se and S-doped a-Si:H thin films at a typical wavelength, $\lambda = 750$ nm are shown in Fig. 5(a) and (b) respectively. The values of n were observed to be the lowest at an annealing temperature of around 300 °C for all of the Se and S-doped samples. Thus, it appears that the quality of the a-Si,S:H and a-Si,Se:H films improves up to an annealing temperature of 300 °C and deteriorates beyond this temperature. Annealing at 300 °C in a vacuum for one hour could be a viable alternative to a-Si,C:H films as a wide bandgap semiconductor material.

4. Conclusions

The optical properties of Se and S-doped a-Si:H thin films were investigated with respect to thermal annealing conditions and dopant concentrations. The annealing temperature at which the best optical constants were obtained for the Se and S-doped a-Si:H thin films was experimentally found to be 300 °C. The deterioration of the optical properties was observed when heated above 300 °C. Above this temperature a clear, decrease in transmittance, increase in the absorption coefficients, decrease in the bandgap and increase in the refractive indices were observed. The values of the absorption coefficients observed were approximately 1 order of magnitude higher in the Se-doped a-Si:H thin films than in the S-doped a-Si:H thin films. The value of the bandgap was observed to be smaller in the a-Si,Se:H films than in the a-Si,S:H thin films.

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