Transport Properties of Conversion Materials for Digital Radiography

Jae Hyung Kim^a
School of Computer Aided Science, Inje University,
Eobang-dong, Gimhae-si, Gyeongnam 621-749, Korea

Chang Hee Park

Department of Radiologic Technology, Daegu health College, Taejeon-dong, Buk-gu, Daegu 702-722, Korea

Sang Hee Nam

Medical Image Research Lab., Inje University, Eobang-dong, Gimhae-si, Gyeongnam 621-749, Korea

^aE-mail: jhkim@physics.inje.ac.kr

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Applying the moving photo-carrier grating (MPG) technique and time-of-flight (TOF) measurements, we studied the transport properties of stabilized amorphous selenium typical of the material used in direct conversion X-ray imaging devices. For MPG measurement, we obtained electron and hole mobility and the recombination lifetime of a-Se films with arsenic (As) additions. We found an apparent increase in hole drift mobility and recombination lifetime, especially when 0.3 % As was added into a-Se film, whereas electron mobility decreased with the addition of As due to the defect density. For TOF measurement, a laser beam with pulse duration of 5 ns and wavelength of 350 nm was illuminated on the surface of a-Se with a thickness of 400 μ m. The measured hole and electron transit times were about 8.73 μ s and 229.17 μ s, respectively.

Keywords: Carrier mobility, Charge transport, Conversion material, Moving photo-carrier grating(MPG), Recombination, Time of flight (TOF), Transient time

1. INTRODUCTION

Amorphous selenium (a-Se) is of particular interest within a wide variety of electronic device applications, not only due to its commercial value as a xerographic photoreceptor material but because of its interesting physical properties[1,2]. While traditionally a-Se films were employed in xerography, more recently these materials have been used as the X-ray photoconductor in flat-panel X-ray image detector[3]. The amorphous selenium film currently being studied for use as an X-ray photoconductor is not pure a-Se but rather a-Se alloyed with 0.2-0.5 % As (most frequently 0.3 % As) and doped with chlorine (Cl) in the 10-30 ppm range; this is also known as stabilized a-Se[4,5]. A small amount of As added to a-Se film enhances the thermal stability of the amorphous state, but larger amounts induce undesirable hole traps in a-Se: As films. Doping a-Se film with small amounts of Cl is necessary to reduce the deep traps associated with arsenic atoms[6,7].

The mobility and recombination lifetime of electrons and holes in semiconductors are important parameters that determine the performance of many devices such as solar cells or thin film transistors[8,9]. The moving photo-carrier grating (MPG) technique can be used to determine the carrier mobility and recombination lifetime of these electrons and holes. While several previous studies have conducted MPG measurements on the transport properties of amorphous silicon (a-Si) samples[10-12], no studies to date have used this method to examine transport phenomena in a-Se films. Therefore, we investigated the transport properties of $a-Se_{1-x}$ As_x films, focusing on the underlying electron and hole drift mobility and recombination lifetime. We determined how adding As to a-Se film influenced recombination lifetime. We also investigated time of flight (TOF) regarding drifting electrons and holes in stabilized a-Se film to determine the transit time of holes and electrons.

2. EXPERIMENT

2.1 Sample fabrication and absorption spectra

The starting materials for samples were prepared by mixing 99.999 % a-Se and 99.999 % As (Nippon Rare Metal Co., Japan) in a weight ratio of 0.1, 0.3, 1, 5, and 10 %. As-doped a-Se films were deposited on a Corning glass using the thermal evaporation technique under 10⁻¹ Torr. Prior to film deposition, the Corning glass was washed using an ultrasonic cleaner, rinsed with deionized water, and blown dry using N_2 gas. We measured the transmissions for 30 µm a-Se_{1-x}As_x $(0 \le x \le 0.1)$ films using a visible spectrophotometer to determine the absorption of a-Se:As films, and applied these absorption spectra to estimate the bandgap energy (E_g) of a-Se_{1-x} As_x . A-Se:As films in MPG measurement had a thickness of about 30 µm. Parallel gold electrodes with a 1-mm spatial separation were coated using thermal evaporation to measure the weak current flowing in a-Se:As film.

The starting materials for TOF measurement were prepared by adding 0.3 wt% As and 30 ppm Cl to a selenium pallet with a purity of 99.999 % (Nippon Rare Metal Co.) to improve conduction and thermal properties [4]. The stabilized a-Se layer with a thickness of 400 μ m was placed on a heated indium tin oxide (ITO) substrate using the thermal evaporation method. A semitransparent gold layer was deposited onto the upper surface of the a-Se layer as an upper electrode.

2.2 MPG measurement

Figure 1 presents the experimental setup used for MPG measurement. Two coherent laser beams of wavelength λ formed an interference pattern on the sample surface. The beams impinged under an angle δ , resulting in an intensity grating with a spatial period $\Lambda=\lambda/[2\sin(\delta/2)]$. A well-defined frequency difference Δf was introduced between the two beams, so that the intensity grating moved with a velocity $v_{gr}=\Lambda\Delta f$. This movement was achieved through acousto-optic modulators in each laser beam. We used the method described by Haken et al. to calculate the current density induced by the moving intensity grating [10].

Light intensity at the surface of the sample had a spatial and temporal dependence[11-13]:

$$I(x,t) = I_1 + I_2 + 2\sqrt{I_1I_2}\cos(kx - \omega_{or}t),$$

where I_1 and I_2 are the intensity of the two beams, k is the spatial frequency $(k=2\pi/\Lambda)$, and ω_{gr} is the angular grating velocity $(\omega_{gr}=2\pi\upsilon_{gr}/\Lambda)$. MPG measurements were based on the line $\lambda=532$ nm.

This technique uses a spatially and temporally modulated light intensity to generate photo-carriers, causing a modulation of carrier density. The different mobilities of electrons and holes produce a phase shift between the charge distributions, and the resulting internal

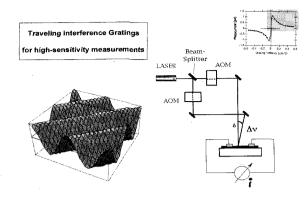


Fig. 1. Experimental setup for measuring the moving photo-carrier grating (MPG).

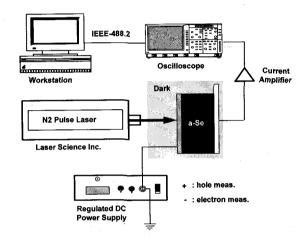


Fig. 2. Schematic diagram of the time-of-flight (TOF) measurement.

electric field creates a grating-velocity-dependent short circuit current j_{sc} . The very existence of j_{sc} indicates that electrons and holes have differing mobilities, and analysis of the sign and the shape of $j_{sc}(v_{gr})$ enables the values of carrier mobility and their recombination lifetime to be determined. In a-Se samples, the short circuit current typically ranges from 10^{-12} to 10^{-13} A, which can be measured using an electrometer. We applied the MPG technique to a-Se_{1-x} As_x (between x=0 and 0.1) films, and found that the laser angle δ for MPG measurement samples was 21.1°, which gives the grating period of Λ =1.45 μ m.

2.3 TOF measurement

Figure 2 is a schematic diagram illustrating the principle of TOF measurement. We applied a voltage across the a-Se layer sandwiched between the Au and ITO electrodes to collect charges. The applied bias (V) appeared across the thickness of the a-Se layer since the external resistance was much less than the a-Se resistance. We used a short light pulse of 5 nm from a laser light source (350 nm) to photo-generate the free

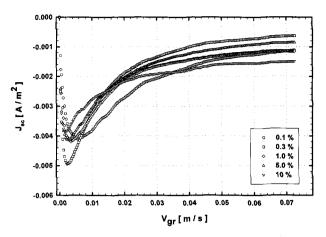


Fig. 3. Current density as a function of v_{gr} .

charges. The transit across the a-Se layer produced a measurable current in the external circuit. We monitored the transient voltage, R_L , using an oscilloscope (LeCroy LC 334AM, USA) as a photo response signal.

3. RESULTS AND DISCUSSION

3.1 Absorption spectra and energy bandgap

Arsenic (As) atoms have only five electrons in their outermost shells $(4s^24p^3)$, while selenium (Se) atom have six $(4s^24p^4)$. The presence of As in Se leaves vacancies called holes in the electron structure of Se atoms. Therefore, the addition of As to a-Se film increases hole mobility because it produces an acceptor energy level, just above the highest filled band. We estimated the bandgap energy E_g of a-Se_{1-x} As_x $(0.001 \le x \le 0.1)$ from absorption spectra obtained using the visible spectrophotometer. The energy bandgap of a-Se_{1-x} As_x films decreased from 2.22 to 1.83 eV when 0.001-0.1 % As was added to a-Se films.

3.2 MPG measurement

Figure 3 presents the short circuit currents measured for a-Se_{1-x} As_x (0.001 $\le x \le 0.1$) films as a function of v_{gr} . Short circuit currents were zero when $v_{gr} = 0$ and decreased linearly for small values of v_{gr} . After reaching a minimum point, j_{sc} increased steadily up to the highest grating velocity that we applied. MPG curves for a-Se:As film differed from those for a-Si:H[11,13]. The MPG curves of a-Se:As film were inverted compared to those of a-Si:H; this was caused by positive photo-carrier charges (holes). The dominant mobility carriers are holes for a-Se films, whereas they are electrons for a-Si:H films[13].

We obtained carrier mobilities μ_n and μ_p by fitting the measured short circuit current to the theoretical expression

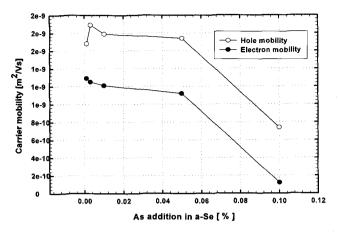


Fig. 4. Electron and hole drift mobility as a function of As addition.

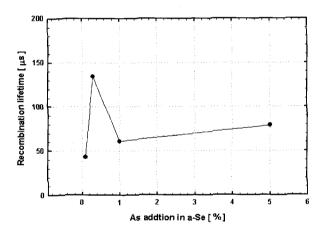


Fig. 5. The recombination lifetimes for a-Se:As films as a function of As addition.

derived by Haken et al.[10]. Electron and hole drift mobility for a-Se_{1-x} As_x films are plotted as a function of As addition in Fig. 4. Hole drift mobility appeared to increase with the addition of As at x=0.003 (0.001 $\le x \le 0.01$), whereas electron drift mobility decreased with the addition of As. Hole mobility decreased due to defect density of deep hole traps when x exceeded 0.003, after hole mobility increased with small additions of As (0.001 $\le x \le 0.003$)].

Figure 5 illustrates how recombination lifetime is dependent on the addition of As to a-Se films. This behavior makes sense considering that holes dominantly recombine after being excited from being localized into extended states. We found an apparent increase in τ_R when x=0.003 ($0.001 \le x \le 0.1$), and that τ_R increased with As addition when As exceeded x=0.01. This change is probably a result of two factors: small additions of As (up to x=0.003) to a-Se films enhance the electric conductivity of a-Se_{1-x} As_x films, and further addition of As induces undesirable hole traps in a-Se samples[14].

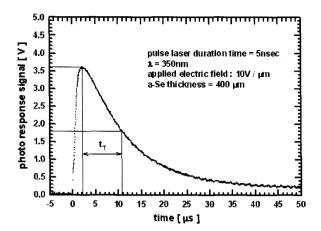


Fig. 6. Hole photo response signal.

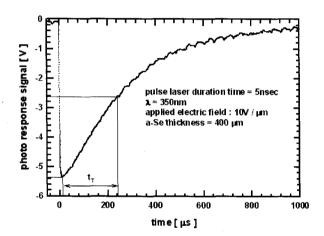


Fig. 7. Electron photo response signal.

3.3 TOF measurement

Figures 6 and 7 present transient current TOF signals for an a-Se layer (400 μ m). Transient TOF waveforms were taken after the application of 10 V/ μ m across the a-Se layer. Results indicate that the electric field remained uniform across the a-Se film layer, suggesting the presence of hole and electron trapping within the a-Se layer. The transit time became dependent on an applied electric field to raise charge collection as a theoretical anticipation value.

At a voltage bias of 10 V/ μ m, hole and electron transit times were 8.72 and 229.2 μ s, respectively. TOF transient photocurrents exhibited behavior similar to that in a-Se_{0.966}Te_{0.034} alloy photoreceptor film, as reported by Kasap[15].

Table 1 lists hole and electron transit times as a function of the applied electric field. The table shows that the transit times were inversely proportional to the applied electric field, up to 10 V/um.

Table 1. Hole and electron transit times as a function of the electric field.

Electric field	Transit time [µs]	
[V/µm]	Hole	Electron
4	24.9688	565.1245
5	19.0840	450.8741
6 .	16.2088	370.5562
7	13.8562	337.2540
8	10.9794	299.5000
9	9.9339	247.5556
10	8.7267	229.1730

4. CONCLUSION

Hole drift mobility appeared to increase with the addition of As at x=0.003 $(0.001 \le x \le 0.05)$. In contrast, electron drift mobility did not appear to change as a function of As addition. Hole mobility decreased due to defect density of deep hole traps when x exceeded 0.003, whereas hole mobility increased with the addition of small quantities of As ($x \le 0.03$). Among non-crystalline semiconductors such as a-Se:As film (the class of materials investigated in this study), any photo-generated carrier generally exhibits multiple trapping before recombination[10]. Thus, space- and time-dependent electron and hole densities appear to include free and trapped photo-carriers in a-Se:As films, which have a high density of localized states extending from band edges into the bandgap. Moreover, mobilities μ_n and

 μ_p are not free carrier mobilities, but rather multiple trapping processes that are strongly dependent on the addition of As to a-Se films.

 τ_R increased with the addition of As, up to 5 %. Recombination lifetime in a-Se:As (0.3 %) film also appeared to increase. This change was probably caused by two factors. In contrast, the addition of small quantities of As to a-Se:As films (up to x=0.003) enhanced the electric conductivity of a-Se_{1-x} As_x films, while adding more As induced undesirable hole traps in a-Se samples[15,16].

We also measured hole and electron transit times in stabilized a-Se using a time-of-flight technique. Hole and electron transit times were strongly dependent on the electric field applied to collect the carriers. The measured hole and electron transit times were about 229.17 and 8.73 μ s at 10 V/ μ m, respectively.

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