# Deposition of hydrogenated amorphous silicon (a-Si:H) films by hot-wire chemical vapor deposition (HW-CVD) method: Role of substrate temperature

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## Abstract

Hydrogenated amorphous silicon (a-Si:H) thin films were deposited from pure silane ($SiH_4$) using hot-wire chemical vapor deposition (HW-CVD) method. We have investigated the effect of substrate temperature on the structural, optical and electrical properties of these films. Deposition rates up to $15\,\text{Å}\,s^{-1}$ and photosensitivity $\sim 10^6$ were achieved for device quality material. Raman spectroscopic analysis showed the increase of Rayleigh scattering in the films with increase in substrate temperature. The full width at half maximum of TO peak ($\Gamma_{TO}$) and deviation in bond angle ($\Delta\theta$) are found smaller than those obtained for P-CVD deposited a-Si:H films. The hydrogen content in the films was found $<1\,at\%$ over the range of substrate temperature studied. However, the Tauc's optical band gap remains as high as 1.70 eV or much higher. The presence of microvoids in the films may be responsible for high value of band gap at low hydrogen content. A correlation between electrical and structural properties has been found. Finally, the photoconductivity degradation of optimized a-Si:H film under intense sunlight was also studied.
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*Keywords:* Amorphous silicon; Chemical vapor deposition; Raman spectra; Thin films

## 1. Introduction

During the last two decades and more, hydrogenated amorphous silicon (a-Si:H) has been studied extensively as a basic material for thin film solar cell applications. Variety of deposition methods have been used to yield material with good opto-electronic properties and higher deposition rates [1–3]. Out of these, only plasma chemical vapor deposition (P-CVD) has been established for industrial application. However, the device quality a-Si:H films prepared by P-CVD method at optimized deposition parameters show lower growth rates [4] and constrains the film deposition to a narrow substrate temperature range [5]. The lower growth rates increase the process

operation time and hence the production cost. The narrow substrate temperature range involves the complexity of control on hydrogen, which is responsible for the light-induced degradation of electronic properties [6]. Furthermore, the possibility of various gas phase reactions leading to the formation of higher order silanes in plasma [7] and the bombardment of growing film with high energy ions add to creation of defects in a-Si:H films which hampers the electrical properties [8]. Therefore, investigations of alternate deposition methods, which allow high deposition rates and device quality, are desirable.

Hot-wire chemical vapor deposition (HW-CVD) or simply "hot-wire method" has received considerable attention in recent years as an alternative deposition method for a-Si:H [9–12] because it is capable of improving film stability [12] and of achieving higher deposition rates [10]. In hot-wire method, no discharge has to be sustained

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and only particles with thermal energies are involved. This method appears to be capable of providing easy control on the production of unknown higher order silanes whose gas phase reactions generally lead to the incorporation of undesirable excess hydrogen in the film. Also, this method involves few parameters which can be easily optimized and the film growth process involves primary general radicals (atomic Si and atomic H) released from the hot filament [11,13]. However, some reports have indicated that the films deposited by hot-wire method do not show any systematic correlation between the deposition parameters and the resulting film properties [11]. On the other hand, some reports [12,14] have shown that films with very low hydrogen content are electronically as good as device quality films produced by P-CVD which contain large quantity of hydrogen ($>15$ at%). Moreover, hot-wire method grown films showed less light induced degradation [15]. Therefore, detailed careful investigations of the synthesis and properties of hot-wire method grown a-Si:H films are required.

In this paper, we present the electrical, optical and structural properties of a-Si:H films deposited by hot-wire method as a function of the substrate temperature. An attempt has been made to deposit highly photosensitive low hydrogen content a-Si:H films by HW-CVD. In the present study, we have obtained device quality a-Si:H films with higher photosensitivity, low hydrogen content at high deposition rate.

## 2. Experimental procedures

The films were deposited simultaneously on corning #7059 glass and c-Si wafers in a HW-CVD system, details of which have been described elsewhere [16]. Pure silane ($SiH_4$), Matheson Semiconductor Grade was used as source gas. The pressure during deposition was kept constant by using manual throttle valve. The temperature of the filament was maintained at $1900 \pm 25\,°C$ due to reason mentioned elsewhere [17]. The substrate temperature was varied from 50 to $400\,°C$ at an interval of $50\,°C$. Other deposition parameters are listed in Table 1.

In hot-wire method, substrates sometimes gets heated to higher temperature than the permissible level by thermal radiation from the hot filament. Therefore, accurate control of substrate temperature is crucial in thin film deposition by hot-wire method. It has been reported that the precise control of substrate temperature can be

achieved with the large spacing between the hot filament and substrate holder and with the reduction of thermal capacitance of the substrate holder on which the substrates were mounted [18]. Placing a substrate with large spacing from the hot filament leads to reduction of the effect of heat radiation and can lower the substrate temperature. However, disadvantage of this method is in the decreasing of deposition rate. To maintain the substrate temperature independent from the heat of hot wire, we have kept substrates at a distance of 6 cm from the filaments. In order to ensure the stable substrate temperature, prior to each deposition, the substrate holder and deposition chamber was baked for 2 h at $200\,°C$. This also helps to remove any water vapor absorbed on the substrate and to reduce the oxygen contamination in the film. After that, the substrate temperature was brought to desired value by appropriately setting the temperature controller. The substrate temperature was held constant during the deposition to the desired value accurately using a thermocouple and temperature controller with an error of $\pm 2\,°C$ by circulating chilled water at a speed of 2400 liters per hour (LPH) through stainless steel (SS) substrate holder. With this arrangement, small increase in substrate temperature ($\sim 5$–$10\,°C$) was still observed during the 10 min of deposition period for substrate temperature less than $150\,°C$.

The dark conductivity ($\sigma_{dark}$) and photoconductivity ($\sigma_{photo}$) were measured with a co-planar Al electrode. The FTIR spectra was recorded on Perkin-Elmer FTIR spectrophotometer. Bonded H contents ($C_H$) were calculated from Si–H wagging mode infrared absorption peaks using the method given by the Brodsky et al. [19]. The optical band gap was measured using conventional Tauc's plot [20]. Raman spectra were recorded with Spex third laser Raman Spectrometer [21]. The thickness of films was determined by Talystep profilometer (Taylor–Hobson). To investigate the degradation of optimized intrinsic a-Si:H films, films were directly exposed to the intense sunlight having average solar insolation $860\,mW\,cm^{-2}$ and the photocurrent was monitored.

## 3. Results and discussion

### 3.1. Variation in the deposition rate

Fig. 1 shows the variation of deposition rate ($r_d$) as a function of substrate temperature ($T_{sub}$) for a-Si:H films deposited by hot-wire method. As seen from the figure, the deposition rate of a-Si:H films decreases from 35 to $12.5\,Å\,s^{-1}$ as substrate temperature increase from 50 to $400\,°C$. The decrease in deposition rate with increase in substrate temperature can be attributed to the temperature dependence of the reaction probability or an increase of silane desorption from the film surface. At low substrate temperature, the radial surface mobility of ad-atom is low. As a result, numbers of ad-atoms are incorporated in the growing film. With an increase in substrate temperature, each ad-atom receives sufficient energy and its surface

Table 1
Deposition conditions for a-Si:H films

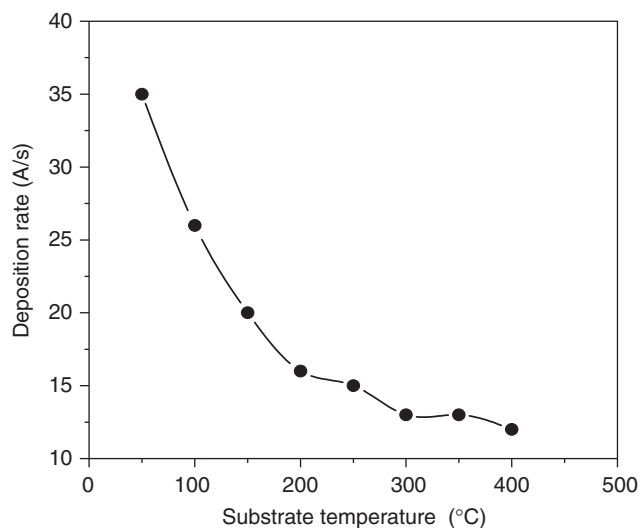| | |
|---|---|
| Substrate temperature, $T_{sub}$ (°C) | 50–400 |
| Filament temperature, $T_{fil}$ (°C) | $1900 \pm 25$ |
| Chamber pressure, $P$ (mTorr) | 20–21 |
| Silane flow rate, $F_{SiH_4}$ (sccm) | 5 |
| Substrate to filament distance, $d_{s-f}$ (cm) | 6 |
| Deposition time, $t$ (min) | 10 |

Fig. 1. Variation of deposition rate as a function of substrate temperature of a-Si:H films deposited by hot-wire method.

mobility increases. This increases their diffusion lengths allowing them to choose favorable low energy sites. As a result, the deposition rate decreases with increase in substrate temperature. The decrease in the deposition rate with increase in substrate temperature was reported previously by Klein et al. [22] for HW-CVD deposited microcrystalline silicon thin films.

### 3.2. FTIR spectroscopic analysis

In order to investigate the silicon hydrogen bonding configuration and to determine the hydrogen content in the a-Si:H films FTIR spectroscopy was used. The FTIR spectra (normalized for thickness) of hot-wire a-Si:H films deposited on c-Si at various substrate temperatures ($T_{sub}$) are shown in Fig. 2. Following observations have been made from the FTIR spectra for a-Si:H films deposited at various substrate temperatures:

1. All films have major IR absorption band centered near $618\,cm^{-1}$ which corresponds to rocking/wagging modes of local vibrations of mono-hydrogen (Si–H) bonded species [23,24]. Its absorption amplitude depends on the deposition temperature. It is clearly seen that the absorption amplitude of Si–H wagging mode decreases with increase in $T_{sub}$.
2. The absorption band at $1900–2250\,cm^{-1}$ has been observed for all films and can be assigned to stretching mode of local vibrations of Si–H, Si–H$_2$ and Si–H$_3$ bonded species [25]. As seen from the spectra, with increase in $T_{sub}$, the intensity of band at $1900–2250\,cm^{-1}$ also decreases.
3. An interesting feature of FTIR spectra is the absence of the $2090\,cm^{-1}$ absorption mode due to di-hydride (the isolated Si–H$_2$ units) or the clustered hydrogen bonding in (Si–H$_2$)$_n$ form (the coupled Si–H$_2$ units) over the entire range of $T_{sub}$ studied.
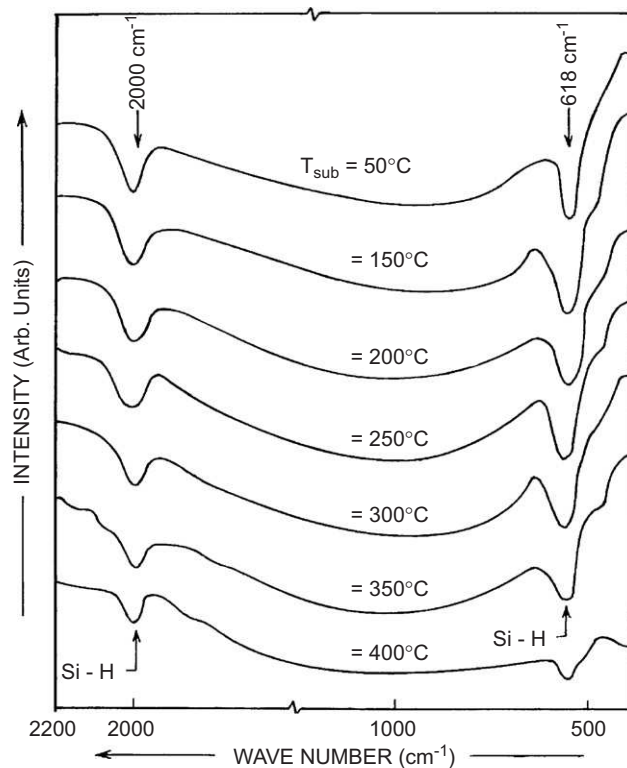


Fig. 2. H-related features of a-Si:H films deposited by hot-wire method at various substrate temperatures.

4. All bands are strongly broadened, indicating the amorphous nature of films.

These results indicate that the mono-hydride silicon (Si–H) bonding configuration is predominant in hot-wire deposited a-Si:H films. At the filament temperature $1900\pm25\,°C$, SiH$_4$ can be very effectively decomposed into atomic Si and atomic H. These radicals are primary radicals available for the film growth process. For the deposition of a-Si:H films by HW-CVD, we have employed low deposition pressure $20\pm1\,mTorr$ and filament to substrate distance 6 cm. Under these conditions, these primary radicals are expected to reach the substrate directly without any gas reaction [12,13]. Hence, the growth of a-Si:H in hot-wire method is expected from hot Si and H species. Therefore, only mono-hydride silicon (Si–H) bonding configuration is expected. The absence of the $2090\,cm^{-1}$ absorption mode due to Si–H$_2$ or SiH$_3$ units in the FTIR spectra over the entire range of substrate temperature studied further support this.

It is observed that bonded hydrogen content in the films estimated from different methods is quite different. However, it has been reported that the most reliable way to estimate the bonded hydrogen content is from peak at $\sim618\,cm^{-1}$ (rocking/wagging mode) [26]. Fig. 3 shows the variation of total hydrogen content ($C_H$) estimated using integrated intensity of $\sim618\,cm^{-1}$ band as a function of substrate temperature ($T_{sub}$). It is interesting to note that the total bonded hydrogen content in the films was found
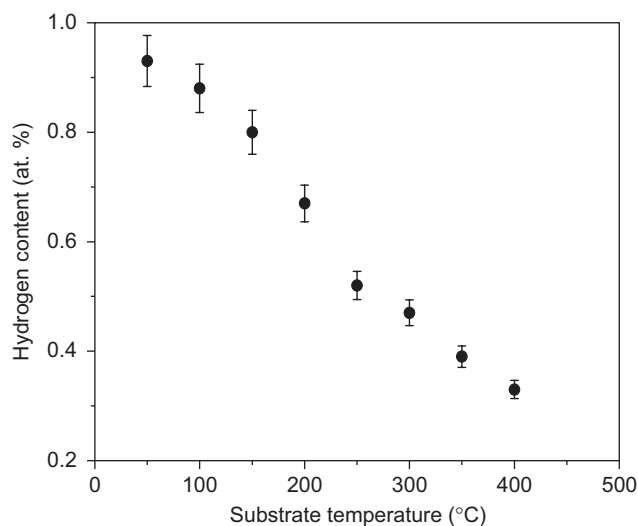
Fig. 3. Variation of hydrogen content of a-Si:H films deposited by hot-wire method as a function of substrate temperature.

to be less than 1 at% over the entire range of substrate temperature studied. The reproducibility of hydrogen content in the films was confirmed by depositing films under same set of parameters for three times. The values of hydrogen content were found the same within the experimental error ($\pm 5\%$). The error bars in Fig. 3 are derived from the differences obtained in hydrogen content from FTIR measurement by repeating deposition of a-Si:H films under the same set of parameters. However, a-Si:H films with very low hydrogen content (0.07 at%) as reported by Mahan et al. [12] could not be reproduced. We believe that the low hydrogen content in hot wire a-Si:H films is dominated by two factors viz. high density of atomic H during deposition and the absence of bombardment of the growing surface with high energetic ions. One $SiH_4$ molecule when decomposed on the hot filament it produces one Si and four H atoms. Therefore, even without hydrogen dilution, a significant amount of atomic H is present in the deposition chamber [27]. The atomic H plays a crucial role in the growth of a-Si:H by hot-wire method [28]. The hot filament thermalizes these H atoms. The fraction of these hot H atoms will impinge on the growing surface with sufficient energy to subtract hydrogen from the surface of growing films. Thus, the generation of higher silane, which causes to bring large amount of hydrogen on the growing surface, is likely suppressed. The atomic H stabilizes the film structure by breaking weak Si–Si bonds and promoting cross-linking [28]. This allows the deposition of device quality a-Si:H at low hydrogen content. Besides, the local heating of growing film due to hot primary radicals evaporated from the heated filament causes the evaporation of H from the growing surface. However, the temperature of such local heating is yet not known. Another probable reason for low hydrogen content in hot wire a-Si:H films may be the absence of bombardment of growing surface with high

energetic ions [29] which is present at different levels in P-CVD depending upon the deposition conditions.

### 3.3. Raman spectroscopic analysis

In order to investigate the effect of such low hydrogen content on structural order of a-Si:H films Raman spectroscopy have been employed. Fig. 4 shows the Raman spectra of a-Si:H films deposited on c-Si at different substrate temperatures ($T_{sub}$) by hot-wire method. The broad and smooth band nature of Raman Si–Si transverse optic (TO) mode indicates that all films are amorphous. Other noticeable effect is the increase of Rayleigh scattering with increase in substrate temperature, which is seen to overlap with transverse acoustic (TA) band positioned near $100$–$150\,cm^{-1}$. The variation of TO peak position ($\omega_{TO}$), its full width at half maximum ($\Gamma_{TO}$) and deviation in bond angle ($\Delta\theta$) determined from the model calculations of Beeman et al. [30] are plotted in Fig. 5. As seen from Fig. 5a, the TO peak which is located at $470.0\,cm^{-1}$ for the film prepared at $T_{sub} = 50\,°C$ shifts to $480.5\,cm^{-1}$ for the film prepared at $250\,°C$ and then saturates at $480.5\,cm^{-1}$ when $T_{sub}$ further increased to $400\,°C$. Furthermore, it can be seen from Fig. 5b and c that both $\Gamma_{TO}$ and $\Delta\theta$ decrease with increase in substrate temperature upto around $250\,°C$ beyond which both the quantities increase. The minimum of $\Gamma_{TO}$ and $\Delta\theta$ are $42\,cm^{-1}$ and $7.00°$, respectively. All these variations indicate that $T_{sub} = 250\,°C$ is an optimum substrate temperature for the synthesis of a-Si:H films by HW-CVD at which the structural disorder is minimum. We think that the surface mobility of ad-atom and hydrogen desorption play an important role during the growth of these films. We believe that below $250\,°C$, the lower surface mobility of ad-atom is important while above this the substrate temperature becomes more important due to increase of hydrogen desorption during growth process. An increase of hydrogen desorption during the growth process
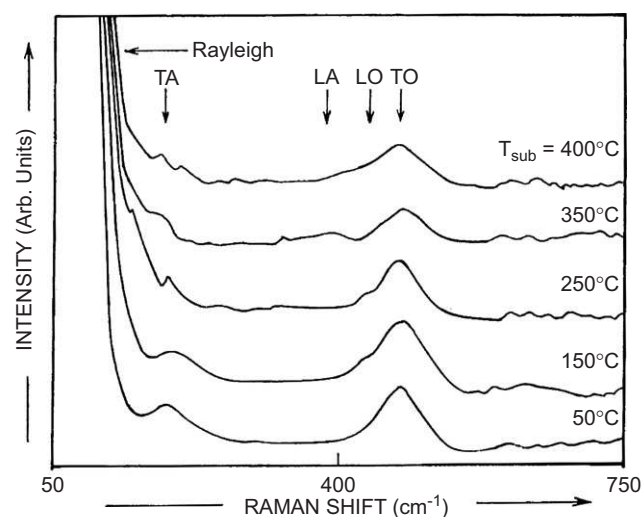


Fig. 4. Raman spectra of a-Si:H films deposited by hot-wire method at different substrate temperatures.
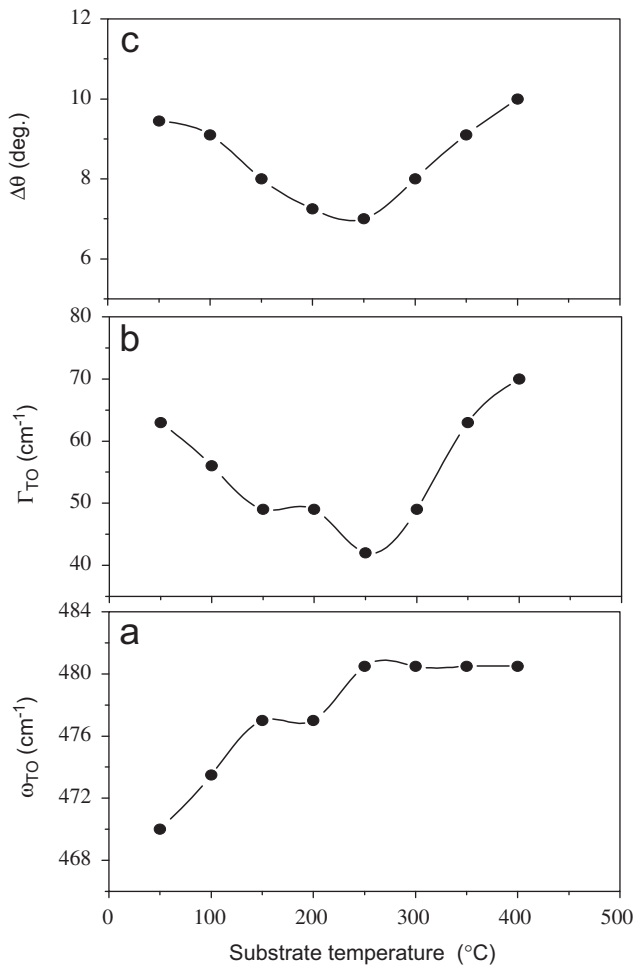
Fig. 5. Variation of (a) TO peak position ($\Gamma_{TO}$), (b) full width at half maximum of TO peak ($\omega_{TO}$) and (c) bond angle deviation ($\Delta\theta$) of a-Si:H films deposited by hot-wire method as a function of substrate temperature.

has been observed with increasing substrate temperature for HW-CVD films [31]. Over a small regime around $T_{sub} = 250\,^{\circ}\text{C}$ or there about the conditions are such that the beneficial and the adverse effects of increase in substrate temperature result in a device quality film structure. The above inference is further strengthened by the observed variation in dark conductivity and photoconductivity as a function of substrate temperature for the HW-CVD grown a-Si:H films (Fig. 7, next section). The film deposited at $T_{sub} = 250\,^{\circ}\text{C}$ shows the maximum photosensitivity of $\sim10^6$ due to minimum structural disorder. More recently, XRD experiments by Sahu et al. [32] have shown that the internal stress in HW-CVD material is minimum for the films deposited at $T_{sub} = 250\,^{\circ}\text{C}$ due to the better network in the films. Furthermore, the real time spectroscopic ellipsometry studies revealed that the best material properties are obtained when HW-CVD a-Si:H films deposited at $T_{sub}\sim250$–$350\,^{\circ}\text{C}$ [33]. Since $\omega_{TO}$, $\Gamma_{TO}$ and $\Delta\theta$ are related to topological disorder in the film other techniques such as TEM will assist in clarifying further structural changes and will be reported in future work. It is interesting to note that

the values of $\Gamma_{TO}$ and $\Delta\theta$ are smaller than those obtained for P-CVD deposited a-Si:H films in the same substrate temperature range [21]. Hence as suggested by Mahan and co-workers, hot-wire grown Si:H films have a different better Si–Si network structure compared with P-CVD films.

### 3.4. Optical properties

The optical band gap ($E_{opt}$) for a-Si:H films deposited by hot-wire method at different substrate temperatures ($T_{sub}$) is shown in Fig. 6. The optical band gap decreases almost linearly with increase in substrate temperature. It decreases from 1.86 to 1.70 eV as substrate temperature increases from 50 to 400 °C, typical of a-Si:H films. In P-CVD deposited a-Si:H films, the optical band gap decreases to 1.5 eV when hydrogen content reduce to 2 at% [34]. On the other hand, the optical band gap of hot-wire deposited a-Si:H films remain as high as 1.70 eV or much higher even when the hydrogen content in the films is less than 1 at%. Thus, only the number of Si–H bonds cannot account for high band gap. The high value of optical band gap in hot-wire deposited a-Si:H films may be due to the presence of microvoids in the films. The Raman spectra of a-Si:H films deposited at various substrate temperatures clearly indicate the increase in the Rayleigh scattering with increase in substrate temperature. This feature may be attributed to increase in the void fraction in the film [35]. The microvoids reduce the effective density of material and increase the average Si–Si distance. This lowers the absorption in the film and shifts the transmission curve towards higher photon energy. This produces higher optical band gap, which is estimated by extrapolation of absorption curve on the energy axis. Thus, the presence of microvoids in the films prevents the optical band gap from reducing its values less than 1.70 eV. In hot-wire method, atomic Si is one of the major species responsible for the growth of a-Si:H films
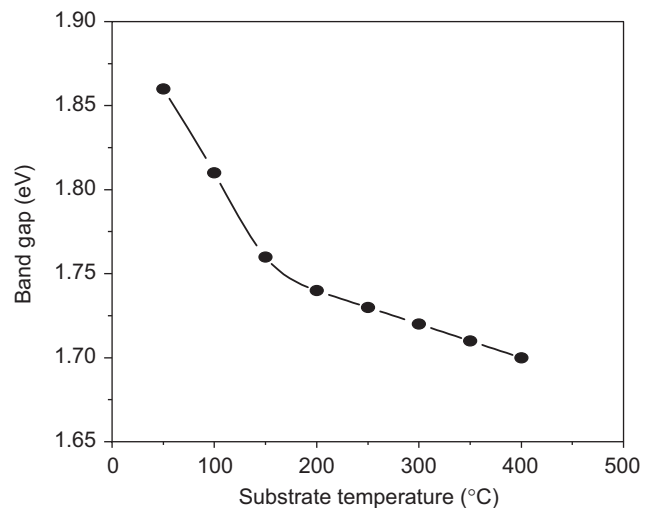


Fig. 6. Variation of band gap of a-Si:H films deposited by hot-wire method as a function of substrate temperature.

at low pressures. Si is believed to result in a void rich material with large structure factor $R^*$ [27,36]. The voids are created in hot-wire grown a-Si:H films mainly by the presence of shadowing effects and simultaneously by the absence of charge particle bombardment [37] which helps to increase the density of the films by impact induced compaction of the debris like pile ups due to shadowing effects. Moreover, with increasing substrate temperature, the surface passivation with hydrogen during the growth decreases. This enhances the probability of formation of voids in the films. Small-angle X-ray scattering (SAXS) measurements by Mahan et al. [38] and nuclear magnetic resonance (NMR) experiments by Wu et al. [39] already revealed that hot-wire grown material generally has higher void fraction than P-CVD material.

### 3.5. Electrical properties

The variation of dark conductivity ($\sigma_{dark}$) and photoconductivity ($\sigma_{photo}$) as a function of substrate temperature ($T_{sub}$) for the hot-wire grown a-Si:H films is shown in Fig. 7. As seen from the figure, $\sigma_{dark}$ for most of the films is found within the range $10^{-10}$ to $10^{-11}\,\mathrm{S\,cm^{-1}}$. These values are comparable with those optimized P-CVD deposited a-Si:H films. It is seen from the curve of $\sigma_{photo}$ that its value is maximum ($\sim 10^{-5}\,\mathrm{S\,cm^{-1}}$) for the film deposited at $T_{sub} = 250\,^\circ\mathrm{C}$ while it decreases both at low as well as at higher substrate temperatures. At low $T_{sub}$ ($> 200\,^\circ\mathrm{C}$), the bond angle deviation as seen from the Raman spectroscopy results is large (see Fig. 5). The density of gap states, which results from the deviation in bond angle and length, in the band tails, is also large. These localized energy levels act as

trapping centers and hampers the photoconductivity. When $T_{sub}$ is between 200 and 250 $^\circ$C, the bond angle deviation is minimum and so the density of states in the band tails. Films deposited at these substrate temperatures show high photoconductivities. At high substrate temperatures, the film becomes more disordered as suggested by higher values of bond angle deviation, and results in lower photoconductivities. Therefore, the photosensitivity
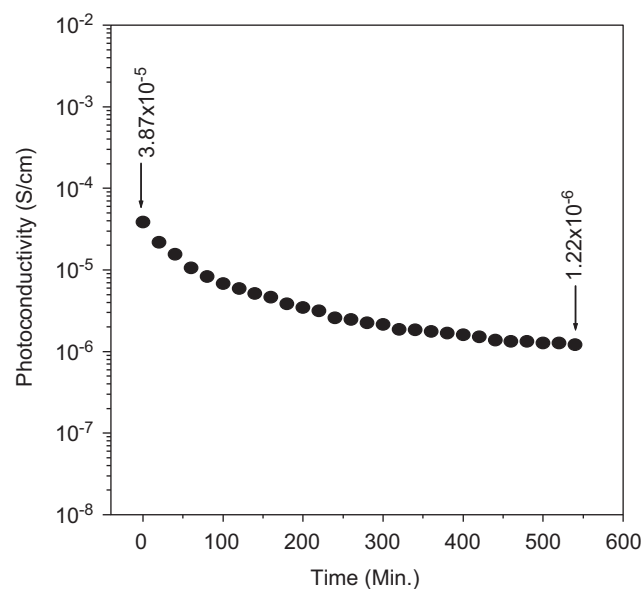


Fig. 8. Degradation behavior of photoconductivity of optimized intrinsic a-Si:H film deposited at substrate temperature 250 $^\circ$C by hot-wire method under the illumination of intense sunlight (average solar isolation 860 mW cm$^{-2}$).
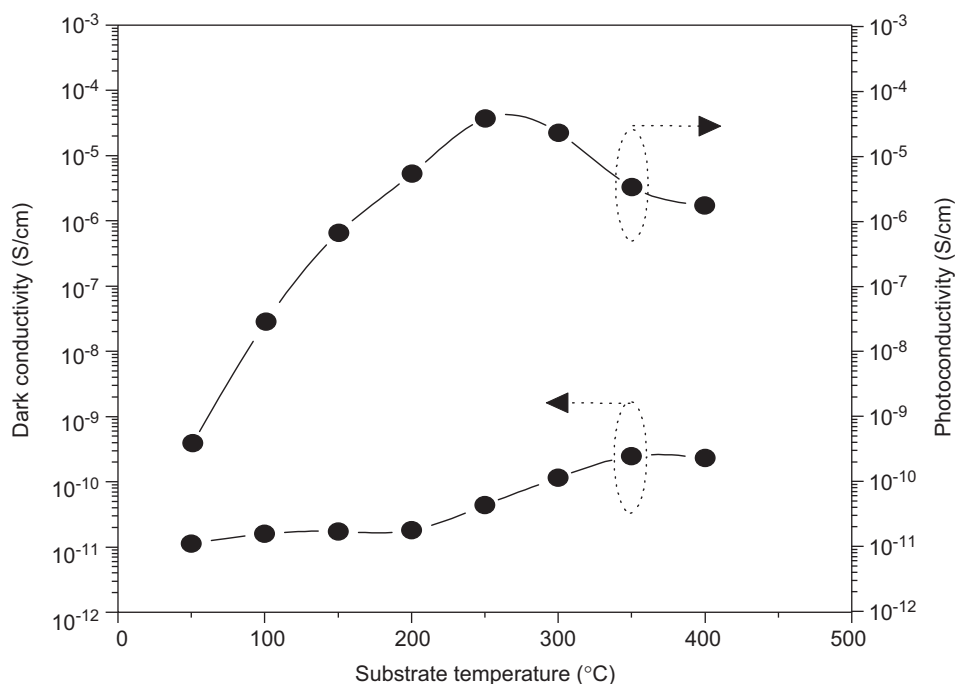


Fig. 7. Variation of dark conductivity and photoconductivity of a-Si:H films deposited by hot-wire method as a function of substrate temperature.

($\sigma_{photo}/\sigma_{dark}$) of the deposited film shows a maximum of $\sim 10^6$ at $T_{sub} = 250\,°C$.

Finally, the photoconductivity degradation behavior of optimum intrinsic a-Si:H film deposited by hot-wire method ($T_{sub} = 250\,°C$, thickness $= 9000\,\text{Å}$) have been investigated and is shown in Fig. 8. As seen from the figure, initially the photoconductivity degradation was quite high and then it decreased slowly. The low hydrogen content and more relaxed Si network of hot-wire grown a-Si:H film may be responsible for such photocuticivity degradation behavior [40].

## 4. Conclusions

An attempt has been made to ascertain the role of substrate temperature on the electrical, optical and structural properties of a-Si:H films deposited by hot-wire method. The hydrogen content in the films was found to be less than 1 at% over a range of substrate temperature studied. However, the optical band gap remains as high as 1.70 eV or much higher. Thus, only the number of Si–H bonds cannot account for high band gap. The high value of optical band gap at such low hydrogen content may be due to the presence of microvoids in the films. The values of $\Gamma_{TO}$ and $\Delta\theta$ of hot-wire deposited a-Si:H are narrower than the P-CVD deposited films. Hence, hot-wire grown a-Si:H films have a different Si–Si network structure as compared with P-CVD films. The maximum photosensitivity was obtained for film deposited at $T_{sub} = 250\,°C$ at which the structure have minimum disorder. The photoconductivity degradation result demonstrates the high potential of a-Si:H film deposited by hot-wire method for the photovoltaic applications. Therefore, employment of hot-wire grown a-Si:H film may lead to more stable solar cells.

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