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Study of Effect of HW-CVD Parameters for Synthesis of Intrinsic and Doped Microcrystalline Silicon Thin Film

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ABSTRACT: The paper provides a comprehensive study of the silicon thin film solar cells. It provides insight in the basic physics governing the behavior of solar cells, the different loss mechanisms and the various steps involved in the fabrication. Novel ideas were explored for increasing efficiency of cells. Emphasis was put into understanding the fabrication process, the effect of process parameters on film and eventually cell quality. Various experimental investigations carried out to explore (i) the losses in different layers and the available spectra for the lower cell in micromorph, and (ii) the effect of every parameter of HW-CVD on film characteristic, the conductivity – a rough measure of crystallinity and doping.

KEYWORDS: conductivity, HW-CVD, micromorph

I. INTRODUCTION

United Nations has predicted a steady annual growth of 0.77% [1] in world human population for the next 40 years. This tremendous increase in population is bound to put a lot of pressure on all the resources presently being used by mankind. Other than the risk of completely exhausting the valuable resources, there is also a great concern due to environmental degradation.

Energy without causing pollution has become an extremely hot topic for the world over the last decade. Due to worldwide increasing consumption and limited fossil fuels, it is clear that the solution to our energy problems is to transition from traditional energy sources to renewable energy sources like solar energy, wind energy and geothermal energy etc. Out of these solar energy is the best option, because the energy reaching the earth every year is more than ten thousand times the energy requirement of the entire world.

Solar energy technologies harness the energy received by the earth from the sun and convert it into thermal or electrical energy. Amongst the various methods to convert solar energy into electrical or thermal energy, Solar cell facilitates the direct conversion from solar to electrical energy, leaving no residues or exhaust gases; it is a clean and green energy process.

A solar cell is an optoelectronic device that converts the energy of light directly into electricity by the photoelectric effect. The operation of a solar cell requires the following basic attributes: the absorption of light, generating electron-hole pairs; the separation of charge carriers of opposite types; the separate extraction of those carriers to an external circuit. Conversion efficiency of solar cells depends on the structural, optical, electrical properties of the light absorber materials and device design.

Crystalline silicon (c-Si) solar cells are currently the most common solar cells in use mainly because c-Si is stable, it delivers efficiencies in the range of 15% to 25%, it relies on established process technologies with an enormous database, and, in general, it has proven to be reliable. There are two doped regions in c-Si solar cell. When a photon of energy higher than band gap of silicon falls on solar cell, it excites an electron from valence band to conduction band and leaves a hole in valence band. Mostly photo carriers are generated far away from electric field reign, so they move towards the electrodes by diffusion process and generate electric power in the external load. Thus the c-Si solar cell is a diffusion type solar device and optical current depends on the diffusion length between electrons and holes.

The major cost for the c-Si solar cells is due to the tremendous amount of semiconductor material that is needed to fabricate it. To reduce this material cost the concept of thin film Si cells came into picture. Thin film Si cells consume a lot less of semiconductor material and hence are cheaper, but since it degrades relatively rapidly and has lower

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efficiency, it has not yet been able to capture the markets. In this thesis I have focused my attention and efforts to make thin film Silicon solar cells more efficient by trying to capture broader solar spectra. Micromorph Solar cell is one exciting cell, in which two cells of silicon are stacked over one another. The two Silicon cells have different morphology, and that changes the effective band gap. Having multiple band-gap cells stacked over one another, helps in effectively capturing more solar spectra, and hence a potentially higher overall efficiency.

II. THEORY

A. *Device Physics*

Following are the key equations that are used to theoretically analyze the behavior of charge carriers in any semiconductor device.

$$\frac{d\varepsilon}{dx} = \frac{q}{\epsilon}(p - n + N_D - N_A)$$

$$J_e = q\mu_e n\varepsilon + qD_e\frac{dn}{dx}$$

$$J_h = q\mu_h p\varepsilon + qD_h\frac{dp}{dx}$$

$$\frac{1}{q}\frac{dJ_e}{dx} = U - G$$

$$\frac{1}{q}\frac{dJ_h}{dx} = -(U - G)$$

Where ε is the electric field, U is the rate of recombination, G the rate of generation, J charge current density, p is concentration of holes, n is concentration of electrons, μ the mobility and N_A and N_D are the acceptor and donor concentration respectively.

B. *Diode Equations*

Current density for p-n diode assuming no recombination or generation is given by:

$$J = J_0\left(e^{qV/kT} - 1\right)$$

J_0 is the reverse saturation current density, V the voltage, q the elementary charge, k the Boltzmann constant and T the temperature. The reverse current is due to the thermal generation of hole – electron pairs that reach junction. At equilibrium, the forward diffusion current equals the reverse saturation current. For an ideal diode, J_0 is given by:

$$J_0 = qn_i^2\left(\frac{D_n}{L_nN_A} + \frac{D_p}{L_pN_D}\right)$$

Where D_n and D_p are the diffusion constants of electrons and holes respectively, L_n and L_p the diffusion lengths of electrons and holes and N_A and N_D the acceptor and donor density respectively.

- *p-n junction*

When light is incident on the p-n junction, the incoming photons collide with the electrons in the valence band and excite them to the conduction band if carrying sufficient energy, in the process creating what is termed as electron and hole pair. The electrons and holes that are able to reach the barrier are swept across by the high electric field, generating an electric potential across the cell.

The value of V_0 is given by the equation: [2]

$$V_0 = \frac{kT}{q}\ln\left(\frac{N_aN_d}{n_i^2}\right)$$

Current generated due to incident light, when the g_{op} is the rate of generation at any place in the diode, is given by:

$$I_{op} = qAg_{op}(L_p + L_n + W)$$

Where L_p and L_n are the diffusion lengths of holes and electrons respectively. W is the depletion width.

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Therefore, total current for the cell under illumination is given by:

$$I = I_0\left(e^{qV/kT} - 1\right) - I_{op}$$

Where I_0 is the saturation current, I_{op} is the photocurrent and V the voltage developed. The open circuit potential is given by setting I=0 in the above equation.

$$V_{oc} = \frac{nkT}{q} ln\left(\frac{I_L}{I_0} + 1\right)$$

I_L is the short circuit current density.[3]

- *p-i-n junction*

The diffusion lengths in amorphous and polycrystalline thin film semiconductors are extremely low, and hence is the photocurrent. A novel concept of having an intrinsic layer was introduced to effectively increase the volume where generated electron hole pairs are captured. The intrinsic layer acts as the generation site as in electron hole pair generated in this region are swept apart by the high electric field. Since, i-layer throughout is the generating layer, its thickness is increased.[3]

- *Tandem cell*

The novel concept of tandem solar cells, which was introduced to cover a broader spectrum, and hence more efficiently use the solar spectrum, has one cell of larger band-gap stacked over the smaller band-gap cell. The upper cell generates power using the high energy photons and allows the lower energy photons to pass through, which get absorbed by the lower cell. Though the concept of tandem cell appears elementary, there are few aspects that need to be taken care of while analysing.[4]

The cells are in series, therefore though the voltages are going to add up; the operating current of both the cells is going to be controlled by the lower of the two. Hence there is a need to match currents. To match currents, the thickness of the cells is so adjusted that the number of electron hole pair generated in upper i-layer is same as that in the lower one.[5]

C. Efficiency Limits and Losses

Detailed Balance analysis by Shockely and Queisser in 1961 theoretically predicted the limit of single junction to be ~31% for a band gap of ~1.11 eV surprisingly close to Si band gap [6]. Later a lot of scientists tried to find the efficiency limit for tandem cells. The results varied a little bit but ~47% efficiency for band gaps of ~0.98 eV and 1.63 eV [7]again, coincidentally, almost the band of microcrystalline and amorphous silicon.

The efficiency calculated are in case of ideal conditions: all the photons with energy greater than the band gap are absorbed and generate an electron hole pair with energy that of band gap. Also, the photons with energy less than that of band gap pass through completely. It is also assumed that solar cell acts as black body and emits spectra characteristic of its temperature. No recombination takes place; and in case of tandem cells, perfect recombination takes place at the Tunnelling Recombination Junction.

There are various methods of efficiency losses, but to model them could be categorised into two types: series resistance and shunt resistance.

Series resistance in a solar cell has three causes: firstly, the movement of current through the emitter and base of the solar cell; secondly, the contact resistance between the metal contact and the silicon; and finally the resistance of the top and rear metal contacts. The main impact of series resistance is to reduce the fill factor, although excessively high values may also reduce the short-circuit current. Figure 7 shows a schematic of the model of effective circuit in solar cell with series and shunt resistance.

Significant power losses caused by the presence of a shunt resistance, RSH, are typically due to manufacturing defects, rather than poor solar cell design. Low shunt resistance causes power losses in solar cells by providing an alternate current path for the light-generated current. Such a diversion reduces the amount of current flowing through the solar cell junction and reduces the voltage from the solar cell. [8]

In the presence of both series and shunt resistances, the IV curve of the solar cell is given by:

$$I = I_L - I_0\left(e^{q(V+IR_S)/nkT} - 1\right) - \frac{V + IR_S}{R_{SH}}$$

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III. SYNTHESIS AND CHARECTERISATION EQUIPMENTS

A. Synthesis:

There are various fabrication techniques for thin film deposition. Hot Wire – Chemical Vapor Deposition (HW-CVD) and Plasma Enhanced – CVD (PE-CVD) are the most popular tools for silicon PV application. Both the techniques aid in depositing semiconductor thin films at relatively low substrate temperatures, and hence are the preferred option of deposition. HW-CVD uses high temperature to decompose precursor molecules whereas PE-CVD, as name suggests, uses plasma to decompose the precursor.[9] Since, the complete work is carried out using HW-CVD, we would restrict further discussion of fabrication to HW-CVD only.

Al/ Ag contacts are created by using thermal evaporation. The thermal evaporation chamber consists of a tungsten boat on which metal to be deposited is placed. Tungsten boat is resistively heated and the evaporated metal gets deposited on the sample, forming a contact. Generally sample is covered by a mask to be able to deposit fine grid lines of metals on the surface. [9]

In sputtering the thin film is deposited by the atoms that are ejected from the target material by Ar+ ion bombardment. Since the substrate is not conducting, RF sputtering is used. At the high frequencies RF Sputtering (~13.56 MHz) ions and electrons have vastly different mobility in the fluctuating field hence the electron which have high mobility ionise argon near the target increasing the etch rate. The signal being cyclic avoids charge to get accumulated on substrate. [9]

B . Charaterisation:

Two probe technique was used to characterise dark and illuminated conductivity values of semiconductor thin film. Conductivity of conducting oxide was measured by using four probe technique. Reliable voltage and current readings was taken using Keithley source meter or Advantest digital electrometer.

Conductivity measurement is necessary to be able to quantize and hence subsequently reduce the power loss due to resistance of solar cell itself. Material resistance of solar cell gets added up in the series resistance in the equivalent circuit, which as discussed earlier significantly reduces the efficiency of the cell.

a) UV – Visible Spectrophotometer:

The transmittance, reflectance and absorption coefficient of the film could be measured using UV – Visible Spectrophotometer. JASCO V-530 UVVIS is the spectrophotometer which would be used for characterisation in my study. The operating wavelength range is 200nm – 1200nm.

b) Stylus Profilometer:

The thickness of the film, one of the key fabrication parameter could be measured using Stylus Profilometer. The one going to be used in my study is Veeco, Dektat 150 with the step size measuring range being ~30 nm – 65 µm, resolution of 50 Å. This technique helps us in calculating the deposition rate, hence giving us control over the thickness of the film deposited.

c) Raman Spectroscopy:

One of the very few characterizing tools available to quantitative measure the percentage crystallinity is Raman Spectroscopy with high confidence level. Percentage crystallinity directly corresponds to the variation in band-gap, mobility of charge carriers and absorption coefficient. Though there are other characterising tools that directly does measure the above mentioned properties, lattice matching is one very important aspect in solar cell fabrication, information about which would be available through Raman spectroscopy.

d) Fourier Transform Infra-Red (FTIR) Spectroscopy:

FTIR helps us get information about the hydrogen content in the thin film. Information about defect density and hydrogen content could qualitatively help in determining the amount of hydrogen in interstitial positions, presence of which is supposed to have a detrimental effect on efficiency of solar cell over long exposure to light.

e) Hall Effect Measurement:

Hall Effect measurement helps in finding the mobility and charge carrier concentration. It is especially helpful while characterising and optimising the doping and thickness of doped layers

IV. EXPERIMENTAL INVESTIGATION

It was observed that the attempted microcrystalline cell had extremely high series resistance and hence generated almost negligible current. The possible reasons were:

- Electron and hole pair are not getting generated in the i-layer at all
- Doping is not enough because of which the electron hole pair are not getting separated

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- Defects at the interface are so high that all the generated current is getting recombined

Experiments were carried out to check for the three reasons. The parameters were optimized such that i-layer had a photogain of ~1.5 orders, making sure that electrons and holes are getting generated. The p-i and i-n interface has a huge number of defects since the percentage crystallinity varies across the interface. To reduce the recombination efforts were taken to make the microcrystalline doped layers. While optimizing parameters for microcrystalline cell, I have worked at two substrate temperatures regimes, 220°C (high temperature depositions) and 150°C (low temperature depositions).

A. Losses in Aluminium doped Zinc Oxide (AZO) layer and the amorphous top cell:

The section shows the study of losses due to AZO layer which acts as a Transparent Conducting Oxide (TCO) in cell fabrication (**Error! Reference source not found.**).

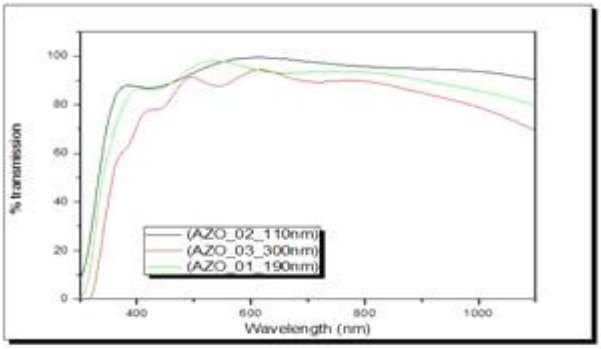

Figure 1 Transmission Vs Wavelength variation with thickness of AZO

Figure 1 shows the spectra available for the amorphous solar cell after various thicknesses of AZO layers. The percentage loss due to the AZO layer is also quantified.

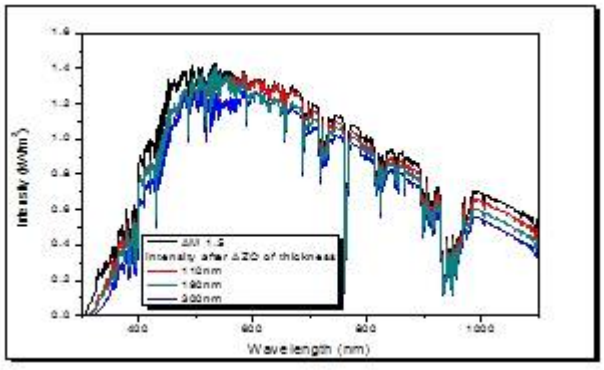

Figure 1 Solar spectra available after AZO of different thicknesses and
% losses due to the layers of varying thickness

The study of light absorption by amorphous top cell is shown in Figure 2 along with the absorption spectra of the microcrystalline film.

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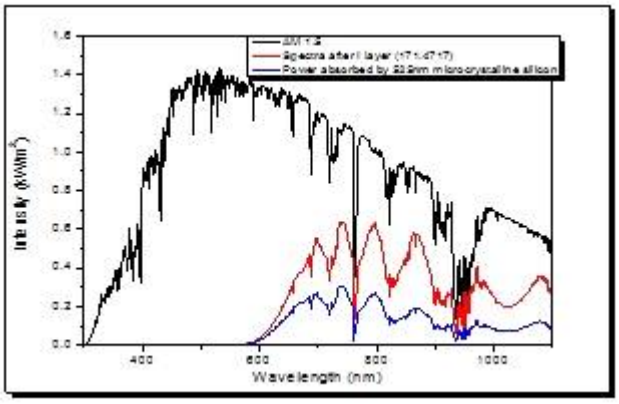

Figure 2 Spectra available for microcrystalline cell to use in power generation in micromorph

B. High Temperature Experiment:

High temperature experiments were carried out to improve the short range order of amorphous layers to check if there is any improvement in the cell, but since conductivity suffered as the parameters were varied (conductivity being the measure of doping), the field it could produce in cells reduced. No cell had any photo generated current. Though, in dark they showed brilliant diode characteristics with dark current in the order of 5-6 mA/cm^2.

- Study of i-layer crystallinity with hydrogen dilution

Hydrogen dilution defined as [SiH_4]/([SiH_4]+[H_2]) was varied in the range where it was expected to have a crystallinity of 50%, which is reported to show best results for photogain.

Figure 4 shows the Raman spectra of µc-Si:H films deposited at various silane concentrations at substrate temperatures of 220°C. It was observed that at hydrogen dilution of 0.9677 the % microcrystallinity was 50%, and hence the ratio, filament temperature, chamber pressure and filament substrate distance was fixed and varied when needed around these.

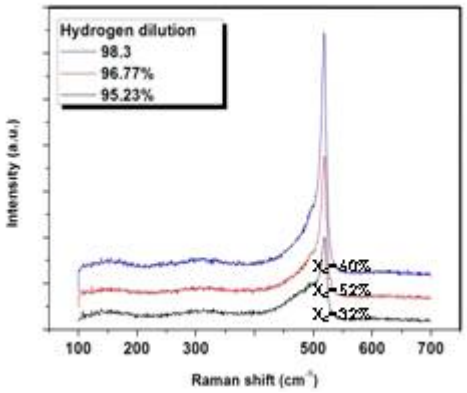

Figure 4: Raman spectra of µc-Si:H films deposited at various silane concentrations at high temperatures

C. Low Temperature Experiments:

Since the flexible substrate we were using was unstable above 150°C, substrate temperature of 150°C was used.

- Intrinsic microcrystalline silicon deposition

Prolonged exposure to filament temperature greater than 1600°C damaged the substrate; hence i-layer depositions at lower filament temperatures were carried out and checked for crystallinity.

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Figure 5 shows variation of crystallinity factor with hydrogen dilution.

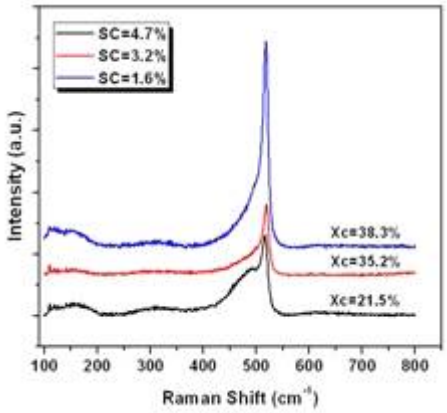

Figure 5: Raman spectra of µc-Si:H films deposited at various silane concentrations at low temperatures

- N-type microcrystalline silicon

Since, Si-P bond is stronger than Si-Si bond, the hydrogen dilution method used to synthesize microcrystalline silicon worked extremely well. The only concern was that, due to high crystallinity, there were high stresses developed in the film due to which it peeled off. It was found that the film was extremely sensitive to filament temperature. The average filament temperature was controlled and kept under 1625°C during the complete deposition. Extremely high conductivity of 25 S/cm obtained for n layer and the Raman plot shown in Figure 6 proves it to be crystalline and also highly doped.

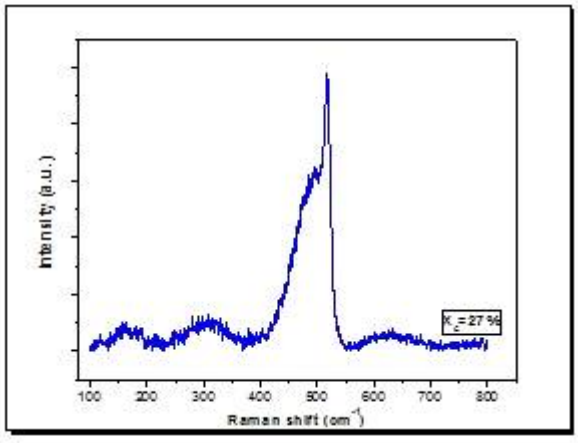

Figure 6 Raman spectra of n-type microcrystalline silicon

- p-type microcrystalline silicon deposition

The most challenging part was to fabricate the microcrystalline p-type silicon and required the study of effect of each control parameter on conductivity of the layer. It was observed that none of the films were microcrystalline; hence efforts possibly only improved the order keeping the doping constant.

a) Effect of Filament Temperature

Increasing the filament temperature increases the % decomposition of precursor and the purity of deposited film, but it also increases the power consumed. Increasing the filament temperature above a certain limit also reduces purity. The filament gradually starts evaporating and molecules start getting incorporated in the film. The filament temperatures used for deposition of amorphous and microcrystalline Silicon layers are in the range of 1550°C – 1900°C

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[9] [10]. High filament temperature meant more number of dissociated Boron species and it was observed that conductivity of deposited films improved with temperature, but damaged the substrate as shown in Figure 7.

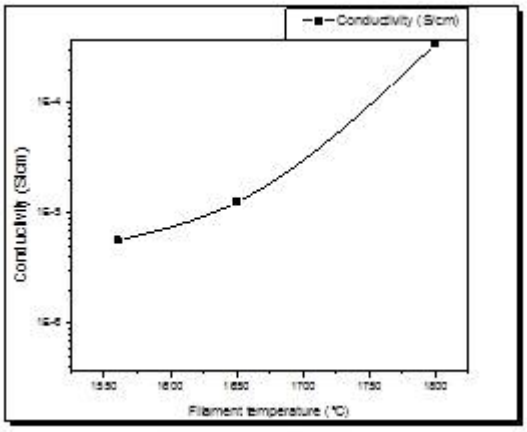

Figure 7 Plot of conductivity vs. Filament temperature

b) Effect of Flow Rates and Chamber Pressure

The precursor flow rate determines the residence time on the filament and hence affects the growth kinetics. Flow rates together with chamber pressure determine the trade-off between the rate of deposition and the quality of film in terms of density, porosity, hydrogen content and gas utilisation. The flow rates (FR) of precursors, their ratios and the substrate temperature, together determine the microstructure of the deposited film. A good quality amorphous film is found to be deposited at zero dilution and flow rate >10sccm whereas good microcrystalline film is found to be deposited at >95% dilution ([FR of H_2] x 100/ [FR of SiH_4 + FR of H_2]) and ~2sccm SiH_4 flow. The chamber pressure is varied in the range of 10-75 mTorr. [11] It is observed that the conductivity of the film improved as we lowered the pressure as shown in Figure 8.

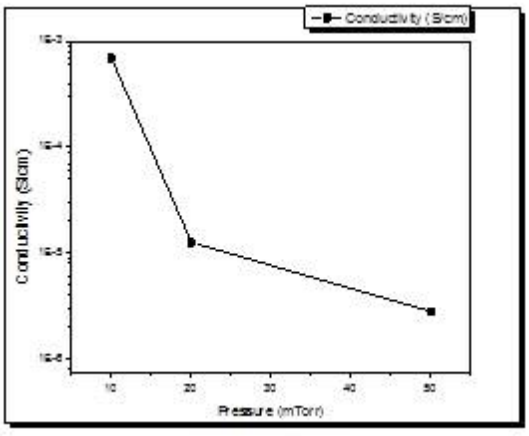

Figure 8 Plot of conductivity vs. Pressure

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Further, increasing boron-silicon ratio potentially increased the disorder in the film since for similar thicknesses the Raman peak intensity increased as shown in Figure 9.

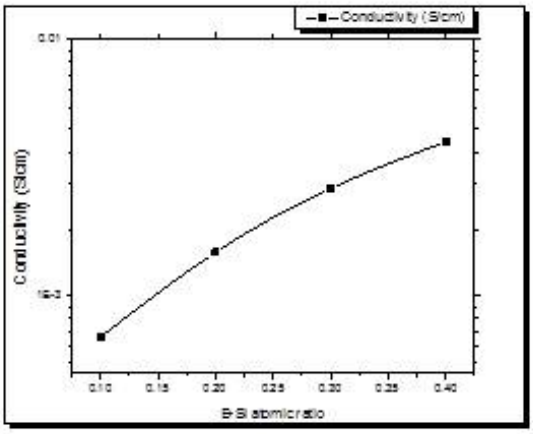

Figure 9 Plot of conductivity vs. B-Si atomic ratio

It is also observed from Figure 10 that the conductivities also increased with increasing hydrogen dilution.

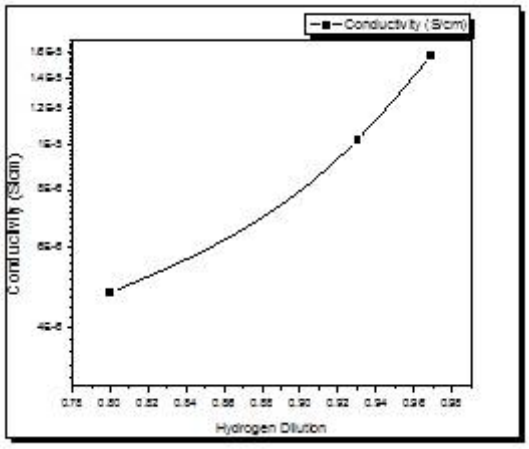

Figure 10 Plot of conductivity vs. Hydrogen dilution

c) Effect of Substrate-Filament Distance

The closer the filament to the substrate, higher will be the deposition rate. Molecules get less time to recombine with each other to form reactants in gaseous phase, hence deposition of desired material increases and impurity content decreases. Keeping filament too close heats the substrate possibly damaging it in the process. Also, determination of substrate temperature during deposition becomes extremely difficult. The distance is generally kept at ~5-10 cm. All the films deposited at 5cm distance showed extremely bad conductivities.

d) Effect of Substrate Temperature

Increasing the temperature of substrate plays a critical role in determining the defect density and uniformity of the deposited film. At high temperatures, atoms have higher surface diffusivity, aiding uniform, defect free deposition. But, higher temperature means higher power and limited substrates. A good amorphous film (defect density ~10^{16}/cm^3) has been obtained at 200°C. Also, a good microcrystalline film (% crystallinity ~50%) is obtained at 220°C substrate temperature. Lowering of substrate temperatures is found to have a some detrimental effect on the quality of film. The sensitivity of polymer made it imperative for us to keep the substrate temperature at no more than 150°C.

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e) Effect of Number of Filaments

A number of experiments have been carried out, and the number of filaments and the substrate – filament distance to maximise the rate of deposition. Increasing the number of filaments increases the rate of deposition but also increases the power consumed for the deposition (11). The number of filaments used during this research work was 4.

V. CONCLUSION

The work carried out was initially to quantify the losses due to various layers and then to reduce the losses at the interface. To reduce the losses at the interface, attempts were made to bring the crystallinity factor of the p-, i-, and n- type silicon films as close to each other as possible. It was observed that synthesis of microcrystalline p-type silicon thin film formation using hydrogen dilution method is difficult at low temperatures because of the close bond energies of amorphous Si-Si and Si-B bond. It could also be attributed to the fact that Boron is famously a glass former, i.e. increases the films amorphous nature. A systematic study of various parameters of HWCVD was carried out to check their effect on film crystallinity and conductivity. Finally, n-type microcrystalline silicon thin film was deposited with χc=27% and conductivity of 25S/cm at 150°C substrate temperature. The microcrystalline i-layer deposited showed a photogain of the order of 1. All the attempts to synthesize microcrystalline p-type silicon failed, since none of the deposited films showed any percentage of crystallinity. The best achieved conductivity of p-type layer was of the orders of -3. The efficiency of solar cell fabricated with the optimised parameters was 1.7%.

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BIOGRAPHY

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