Effect of Temperature on Electrical Properties of Undoped Amorphous Carbon Thin Films Deposited by Bias- Assisted Pyrolysis-CVD

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***Abstract*—The undoped amorphous carbon thin films were deposited on glass substrates by bias-assisted pyrolysis-CVD at various deposition temperatures in the range of 250°C-550°C. The electrical, optical and structural properties were characterized by using current voltage (I-V) measurement, UV-VIS/NIR spectrophotometer and Atomic Force Microscopy (AFM). The electrical conductivity of amorphous carbon thin films increased as the temperature increased. The highest and lowest photo responses were found at 350°C and 500°C, respectively. The highest absorption coefficient was found at the high temperature, 550°C. The AFM images showed that, density and uniformity have correlated with the resistivity and conductivity for undoped amorphous carbon thin films deposited at different temperatures.**

***Keywords*—chemical vapor deposition;amorphous carbon; palm oil; thin films**

I. INTRODUCTION

For a decade, photovoltaic solar cells are mainly fabricated using silicon material and compound semiconductor which dominated the market share [1-3]. Accordingly, allotropes carbons as reported will be promised as a potential candidate for an alternative material in the future due to the abundantly in nature, suitability as a precursor, excellent photoconductivity and high optical absorption of visible light, can be deposited on any inexpensive substrate. It is possible to form a very wide area of solar cell since it can be deposited directly from a kind of vapor phase growth onto non crystalline substrate [4].

Carbon is a good candidate for an alternatively materials in replacement the remarkable silicon in the future due to the abundantly in nature and suitability as a nature precursor. Carbon is found of having a wide band gap which is suitable for optical devices as well as for photovoltaic solar cells application. The band gap of some allotrope carbon such as amorphous carbon in the range of 0.1 eV to 5.5 eV could be tuned tailoring with energy band gap of photon by deposition process [5].

Meanwhile, the undoped amorphous carbon is reported as weakly p-type in nature with complex structure and high density of intrinsic defects [6,7].Until now, various standard deposition techniques are practically used such as thermal-CVD, plasma enhanced chemical vapor deposition (PECVD), ion beam sputtering (IBS), microwave surface-wave plasma (MW-SWP) CVD, etc [8,9]. It is reported that, the standard deposition was claimed to improve the deposition process by acceleration deposition rate, reduce contamination or maintain repeatable process [10,11].

In this work, bias-assisted pyrolysis-CVD is used to deposit a-C thin films and DC bias is not applied to support the deposition process. In order to deposit a-C thin films, carbon source precursor is required. Vaporized of palm oil is used as a precursor to deposit amorphous carbon thin films. Therefore, less energy is needed for break down its bonding. The aim of this work is to investigate the effect of temperature on the electrical properties, optical properties and structural properties of a-C thin films.

II. EXPERIMENTAL DETAILS

The glass substrates were cleaned with acetone (C_5H_6O) to remove the contaminated glass. The glass substrates were cleaned again with methanol (CH_3OH) to remove acetone and de-ionized water to remove methanol for 15 minutes in Ultrasonic Cleaner (power sonic 405), respectively. Finally, the glass substrates were exposing to the Nitrogen gas (N_2).

Figure 1 shows the schematic diagram of bias-assisted pyrolysis-CVD for deposition of amorphous carbon thin films.

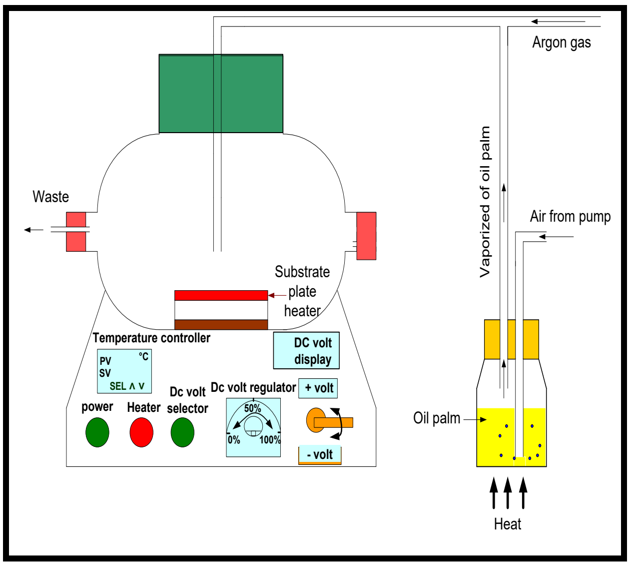


Figure 1: Schematic diagram of bias-assisted pyrolysis-CVD

The cleaned of glass substrates were then placed in the chamber at different temperatures for 1 hour deposition. The chamber was heated with temperature starting from 250ºC to 550ºC. The palm oil was used as a precursor heated at 150ºC with the hot platter (Stuart CB162). A vaporized of palm oil precursor was then pushed into the chamber by two air pumps and the amount of vaporized palm oil, argon pressured into the chamber were set to be constant at 114 mL/min, 186 mL/min, respectively . The argon gas was used for carrier the deposition particles onto the substrate and also to dispose contaminated particles outside the chamber.

Then, all the samples were characterized by using I-V measurement to measure electrical properties, UV-VIS/NIR spectrophotometer (JASCO/V-670 EX) to measure the optical properties and Atomic Force Microscope (XE-100 Park Systems) for measuring the structural properties.

III. RESULTS AND DISCUSSION

A. Electrical Properties

In this research, electrical properties are the important data in solving many issues related to the properties of amorphous carbon thin films [12]. The electrical properties of the undoped a-C thin films were characterized by using current-voltage (I-V) measurement whereas gold is used as a metal contact. In the I-V measurement, two probes were used as an interface for measuring a current voltage relationship between two metal contacts of a-C thin films. There are many forms of current voltage relationships obtained in the literature [15,16] such as linear (ohmic), slightly linear, and nonlinear forms. Besides the ohmic behavior, the low resistivity and high conductivity were also important as a semiconductor was required.

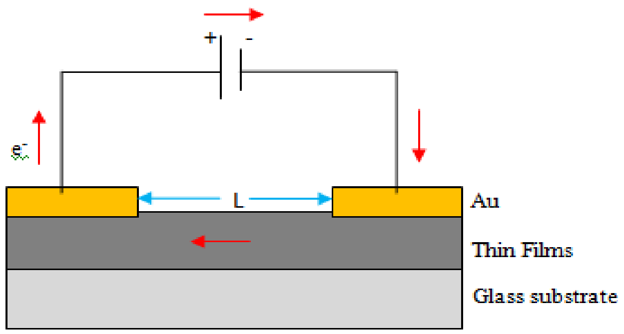


Figure 2: Cross sectional view of the a-C thin films deposited on glass substrate

Figure 2 shows a cross sectional view of the a-C thin films deposited on the glass substrate. Theoretically, the resistivity and conductivity of amorphous carbon thin films can be obtained directly from the equation (1) and (2) respectively [14].

$$\rho = \frac{RA}{L} \tag{1}$$

where ρ is resistivity between two measured metal contact, R is resistance measured from -10V to 10V, A is area of metal contact and L is distance between two metal contacts.

$$\sigma = \frac{1}{\rho} \tag{2}$$

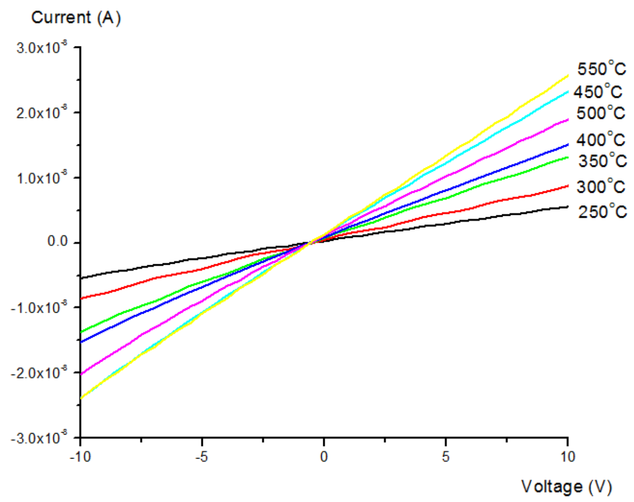


Figure 3: I-V curves of a-C thin films deposited at different temperatures

Figure 3 shows the amorphous carbon thin films deposited at different temperature in the range of 250ºC to 550ºC. The result showed that, amorphous carbon thin films were in ohmic contact behavior with different value of resistances. These show that gold is a good metal contact compared to other material. The highest slope was found for a-C thin films at 550ºC while the lowest slope contact was at 250ºC. The slopes contacts were then slowly decreased starting from 550ºC, 450ºC, 500ºC, 400ºC, 350ºC, 300ºC, and 250ºC, respectively. The slope contact became smaller might be due to the additional of high resistance which come from high density of intrinsic defects.

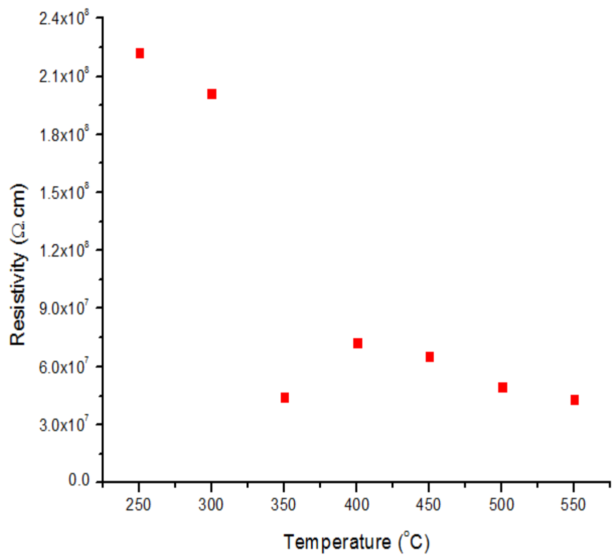


Figure 4: Resistivity curves of a-C thin films deposited at different temperatures

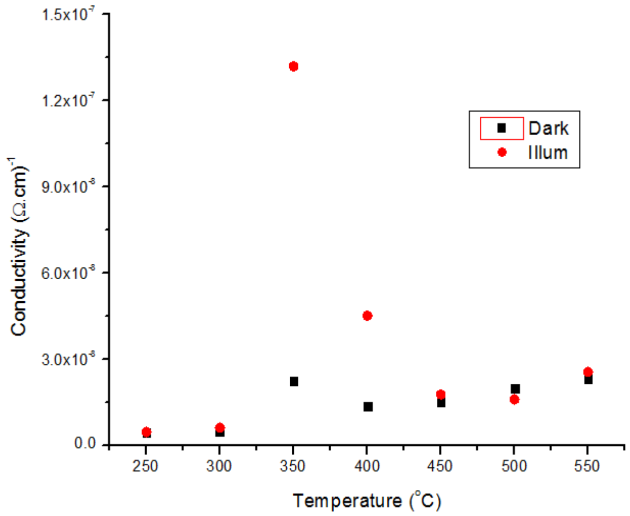


Figure 5: Conductivity curves of a-C thin films deposited at different temperatures

Figure 4 and Figure 5 show the effect of temperatures toward the resistivity and conductivity between two measured mask electrodes. From the Figure 4, it is observed that the resistivity is drastically decreased from 250ºC until 350ºC and increased starting from 350ºC to 400ºC. Then, the resistivity is slightly decreased again from 400ºC to 550ºC. In contrast, the conductivity is drastically increased starting from 250ºC until 350ºC and increased between 350ºC to 400ºC but slightly increased from 400ºC until 550ºC as shown in Figure 5.

It was believed that, the structure of amorphous carbon has transformed from semiconductor to semi-metallic materials. Besides that, the density and uniformity of amorphous carbon have contributed for the increasing of the conductivity as proved by Atomic Force Microscope as shown in Figure 9. According to the Figure 4, the resistivity is varied against various deposition temperatures. The resistivity for 400ºC is $7.26\times10^{7}\Omega$.cm and decreased gradually to $4.32\times10^{7}\Omega$.cm at 550ºC. The conductivity increases from 4.51×10^{-9}S.cm^{-1} to 2.32×10^{-8}S.cm^{-1} for undoped a-C thin films deposited from 250ºC to 550ºC. This trend for resistivity and conductivity variation resembles the trend reported by Hussin et al [13] and A. N. Fadzilah et al [14].

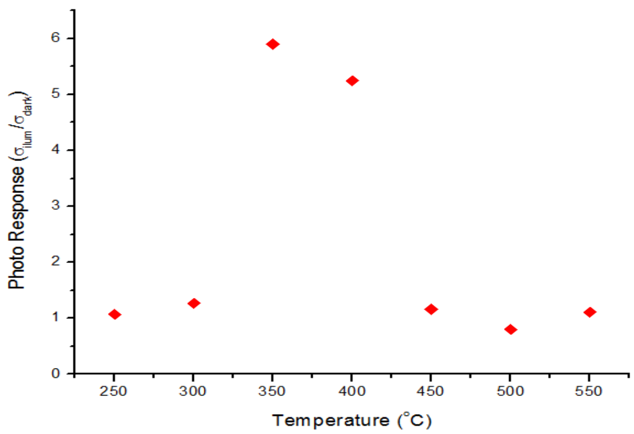


Figure 6: Photo response of a-C thin films deposited at different temperatures

Table 1: The photo response of a-C thin films deposited at different temperature.

Temperature (ºC)	Conductivity (S.cm^{-1})		
	Under Illumination	In Dark	Photo Response
250	4.830×10^{-9}	4.510×10^{-9}	1.0709
300	6.320×10^{-9}	4.980×10^{-9}	1.2691
350	1.330×10^{-9}	2.253×10^{-8}	5.9041
400	7.230×10^{-7}	1.378×10^{-8}	5.2477
450	1.780×10^{-8}	1.532×10^{-8}	1.1616
500	1.610×10^{-8}	2.010×10^{-8}	0.8009
550	2.570×10^{-8}	2.320×10^{-8}	1.1077

Figure 6 shows the effect of different temperatures toward the photo response. A photo response is defined as the ratio of conductivity under illumination to the conductivity under dark. From Table 1, the values of photo response from 250ºC to 550ºC were 1.0709, 1.2691, 5.9041, 5.2477, 1.1616, 0.8009, and 1.1077, respectively. The results showed the photo response was slightly increased from 250ºC to 300ºC and then dramatically increased from 300ºC to 350ºC. The photo response was slowly decreased until 400ºC and dramatically decreased from 400ºC to 500ºC. The highest and lowest of photo response was found at 350ºC and 500ºC which is 5.9041 and 0.8009, respectively. By comparing with the slope variations, resistivity, conductivity, and photo response results, it was believed that at 350ºC is directly interrelated with those data and concluded as the optimum temperature.

B. Optical Properties

To investigate the optical properties of amorphous carbon thin films, transmittance measurement were carried out in the range of 200-2000nm by using UV-VIS/NIR spectrophotometer (JASCO/V-670EX). Figure 7 shows the UV-VIS transmittance spectrum of a-C thin films deposited at different temperatures.

According to the Figure 7, it is observed that all samples have high transmittance (T>90%). From this result, the optical properties of a-C thin films vary with the composition and the thickness of the films. However, the thickness did not strongly influence the transmittance spectrum due to the smaller average thickness. It was found that in visible light, about 90% of light are transmitted to the thin films.

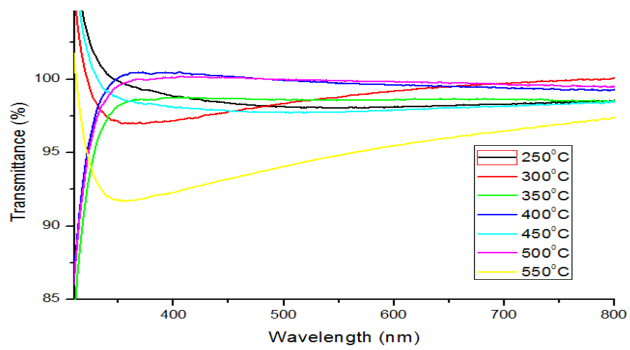


Figure 7: Transmittance spectrum of a-C thin films deposited at different temperatures

Moreover, the absorption coefficient (α) was calculated by the spectral reflectance, transmittance and the film thickness. Figure 8 shows the absorption coefficient of a-C thin films at different temperature. It was showed that thin films at 550ºC is the highest absorption coefficient followed by thin films at 350ºC, 450ºC, 300ºC, 400ºC, 250ºC, and 500ºC, respectively.

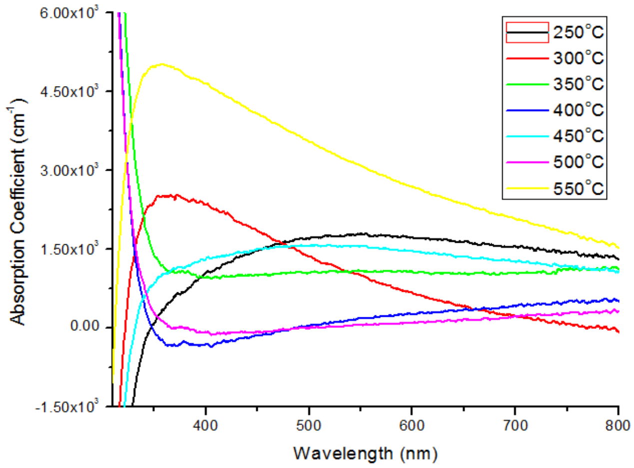
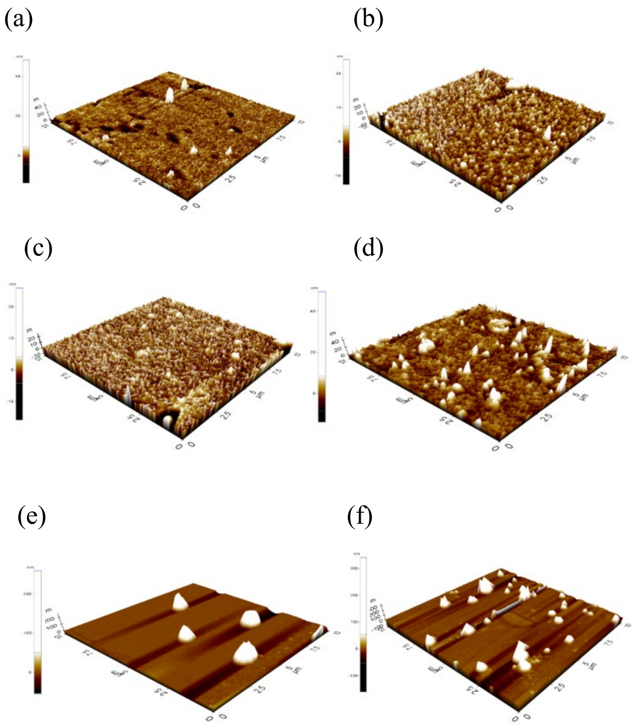


Figure 8: Absorption coefficient of a-C thin films deposited at different temperatures.

From the Figure 8, the low absorption coefficient were found at 400ºC and 500ºC. It shows that the low absorption coefficient might be related with the inconsistently of localized electron and bonds between the electron to any other allotrope of carbon.

C. Structural Properties



(g)

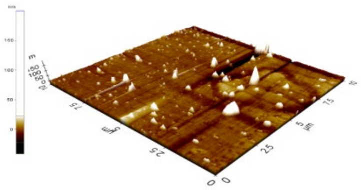


Figure 9: AFM 3-dimensional image of a-C thin films at different temperatures: (a) 250ºC, (b) 300ºC, (c) 350ºC, (d) 400ºC, (e) 450ºC, (f) 500ºC, (g) 550ºC.

Figure 9 shows 3-dimensional (3-D) of the a-C thin films deposited at different temperatures with a scan rate 1 Hz and scan size of 10 µm. Figure 9 (a) shows that a very less of the deposited thin film on glass substrate was present. After the temperature increased from 250ºC to 550ºC, the thin films was showing an increasing of density of thin film at the temperature from 250ºC to 350ºC but dropped from 350ºC to 400ºC. However, the density of thin film increased again at 450ºC to 500ºC. At temperature, 550ºC the density of thin film dropped again. As a conclusion, the results show that changing the temperature affects the uniformity and thickness of the amorphous carbon thin films.

Table 2: The surface roughness and average grain size of amorphous carbon thin films deposited at different temperature.

Deposition Temperatures (ºC)	Surface Roughness (nm)	Average Grain Size (µm²)
250	0.113	3.6330
300	0.116	1.5590
350	0.317	1.2330
400	0.194	0.8059
450	5.394	0.9606
500	6.419	0.8325
550	0.403	0.7556

Table 2 shows surface roughness properties of amorphous carbon thin film and characterized by using AFM (XE-100, Park System). The surface roughness average from 250ºC to 550ºC is 0.113nm, 0.116nm, 0.317nm, 0.194nm, 5.394nm, 6.419nm, and 0.403nm, respectively. At temperature 250ºC and 500ºC, the surface roughness has the lowest and highest value of 0.113nm and 6.419nm respectively. The surface roughness of thin film is increased from 250ºC until 350ºC and slowly dropped starting from 350ºC to 400ºC. The surface roughness is increased from 400ºC until 500ºC but drastically dropped to 550ºC. From this result, it shows that, at temperature of 500ºC, it has the optimum value of the surface roughness. From Table 2, the grain sizes are 3.633µm², 1.559µm², 1.233µm², 0.8059µm², 0.9606µm², 0.8325µm², and 0.7556µm², respectively. For the average grain size, 3.633µm² is the highest for the sample 250ºC while 0.7556µm² is the lowest average grain size for sample 550ºC. It was believed that the grain boundary and surface roughness are related with electrical properties especially carrier concentration and mobility of the electron.

IV. CONCLUSION

The undoped of amorphous carbon thin films were successfully deposited by bias-assisted pyrolysis–CVD. The temperatures were strongly influenced the electrical, optical and structural properties of a-C thin films. All the thin films showed the ohmic contact with the different slope of current voltage relationships. The resistivity of undoped a-C thin films from 250ºC to 550ºC is 22x10^{8}Ω.cm, 2.01x10^{8}Ω.cm, 4.44x10^{7}Ω.cm,7.26x10^{7}Ω.cm, 6.53x10^{7}Ω.cm, 4.97x10^{7}Ω.cm and 4.32x10^{7}Ω.cm, respectively. The resistivity of all thin films decreased as the temperature increased. The conductivity of undoped a-C thin films from 250ºC to 550ºC was 4.51x10^{-9}S.cm^{-1}, 4.98x10^{-9}S.cm^{-1}, 2.25x10^{-8}S.cm^{-1}, 1.38x10^{-8}S.cm^{-1}, 1.53x10^{-8}S.cm^{-1}, 2.01x10^{-8}S.cm^{-1} and 2.32x10^{-8}S.cm^{-1}, respectively. The electrical conductivity of amorphous carbon thin films increased as the deposition temperature increased. All thin films at temperature 350ºC and 500ºC showed the highest and the lowest of photo response, respectively. The optical properties revealed high transmittance which is above 90% for all thin films. The highest and lowest of transmittance were at 250ºC and 550ºC. However, the highest and lowest of absorption coefficient at visible wavelength were at 550ºC and 500ºC. It has the optimum value of the surface roughness at 550ºC. The average grain size was decreased as the temperature increased. The AFM images showed that, density and uniformity has correlated with the resistivity and conductivity of undoped a-C thin films at different temperatures. These results show that there is the possibility to improve the electrical for the application of solar cell.

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