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Structural Properties Of P-Type Boron Doped Carbon Film From Hydrocarbon Palm Oil For Photovoltaic Heterojunction Solar Cell Device

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***Abstract**: The complex-structured boron doped amorphous carbon were prepared from natural hydrocarbon palm oil precursor deposited by negative bias substrate voltage of 0 V and -20 V are presented. Field-emission scanning electron microscopy (FESEM) revealed the carbon films were very complex-structured. The open circuit voltage (Voc), current density (Jsc), fill factor (FF) and efficiency (η) of Au/a-C:B/n-Si/Au heterojunction solar cell device at 0 V were approximately 254 mV, 0.2324 mA/cm^2, 0.241, and 0.0141%, respectively. Meanwhile, the V_{oc}, J_{sc}, FF and η of Au/a-C:B/n-Si/Au heterojunction solar cell device at -20 V were 426 mV, 5.351mA/cm^2, 0.243, and 0.553%, respectively. The results showed the precursor palm oil and negative substrate DC bias of -20 V can be used as an alternative precursor and technique for fabricated heterojunction solar cell device.*
***Keywords**: Amorphous carbon, palm-oil, negative bias, boron, pyrolysis- CVD, solar cell.*

I. INTRODUCTION

Many types of carbon precursors have been discovered from renewable precursors [1-3] and non-renewable sources for producing allotrope carbon such as carbon nano tubes (CNT), graphene, amorphous carbon, etc. using various method of depositions [1-3]. Beside of those precursors, palm oil the other abundantly promising 'green' source was successfully synthesized the vertically aligned carbon nanotubes (VACNTs). Palm oil is scientifically known as hexaeconoic acid which was derived from fibrous exocorp and mesacarp of the fruits of palm tree. The palm oil is contained carbon (67), hydrogen (127) and oxygen (8) to form the chemical binding of $C_{67}H_{127}O_8$ (3). This compound has the highest carbon content among the known precursors. The synthesizing of a-C on the other hand, required less energy compared with other allotropes carbon for instant, the VACNTs need deposition temperature above 700^{o}C [1,2]. Nevertheless, the a-C films are weak *p*-type in nature and they possess complex structure and high density of defects, thereby restricting their doping capacity; this low doping efficiency is the main obstacle for their application in various electronic devices. Amorphous carbon (a-C) films have gained considerable attention because of their controllable optical gap, which allows for its wide application in the manufacture of semiconductors. In order to solve that problem, it was suggested the control of doping could reduce the existing of defect and at the same time modified the electronic properties [4-6].

Among deposition parameters, negative bias voltage applied to the substrates could significantly change film properties due to enhancement of adatom mobility and the effects of ion bombardment. The ion bombardment during coating deposition would play an important role in affecting the morphology, structure, composition and mechanical properties of coatings [7-9]. Many attempts were studied by others on the effect of negative bias for instant through the use of pure lubricant coatings (MoS_2) composite film. It was reported that, the increase of bias caused preferential re-sputtering of S resulting in a reduced S/Mo ratio, which can affect different properties of the film. A reported study on pure MoS_x films deposited by bipolar pulsed DC showed that even an S/Mo ratio of 0.8 was able to provide good lubricious property due to the strong basal plane orientation and application of a bias voltage was found to reduce the coefficient of friction [8-10]. Therefore, an understanding of substrate bias effects is necessary to improve the physical and mechanical properties of MoS_2-based coatings but also important for structural, electrical as well as electronic properties of any semiconductor film.

In this paper, we report the complex-structured boron doped amorphous carbon films deposited by using deposition temperature without bias (0 V) and deposition temperature with the help of a constant negative bias (-20 V). To the best of our knowledge, there is less report on natural bio-hydrocarbon of palm oil as a p-type of a-C film for heterojunction solar cell using negative bias technique.

II. METHODOLOGY

The Boron doped amorphous carbon (a-C:B) were deposited by using bias-assisted pyrolysis-CVD onto the corning glass substrates (thickness: 1mm) and n-Si (100) (thickness 325 $\pm$ 25 µm, resistivity 1-10 Ω cm). Substrates (glass and n-silicon) were together cleaned with acetone (C_5H_6O) followed by methanol (CH_3OH) for 15 min in ultrasonic cleaner (power Sonic 405), respectively and the glass substrates were then rinse with deionizer water (DI) water for 15 min.

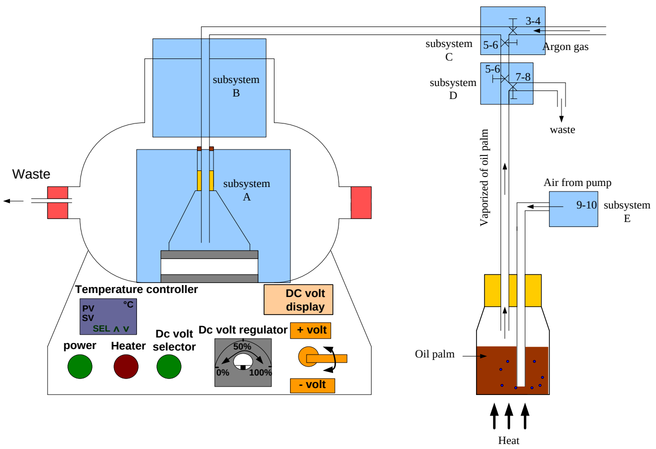


Figure 1 A schematic diagram of bias assisted pyrolysis-CVD

Moreover, excess oxide layers of n-type silicon substrates were continued by the etching process with diluted hydrofluoric acid (10%) solution for about 3 min before rinsing in DI water. Substrates were blown with nitrogen gas. The cleaned of glass and silicon substrate was attached inside the chamber as shown in Fig. 1. The deposition temperature was set at 325°C for 1h deposition. A liquid of palm oil precursor was heated in the bottle outside the chamber at around 150°C by using hot platter (Stuart CB162). The vaporized of palm oil was then pressured into the chamber using low cost aquarium air pumps (model GA8000) (not shown in Fig. 1). The amount of vaporized palm oil, carrier gas argon used into were set to be constant at 114 mL/min, 200 mL/min, respectively by using AALBORG flow meter. For doping process, 1g of boron was placed on the aluminium foil above the metal plate heater. The negative bias voltage is set at 0V and -20V, respectively.

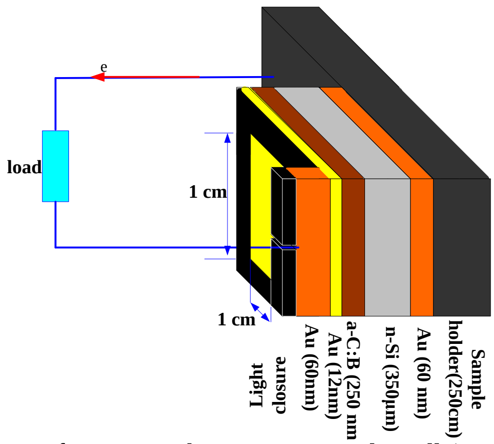
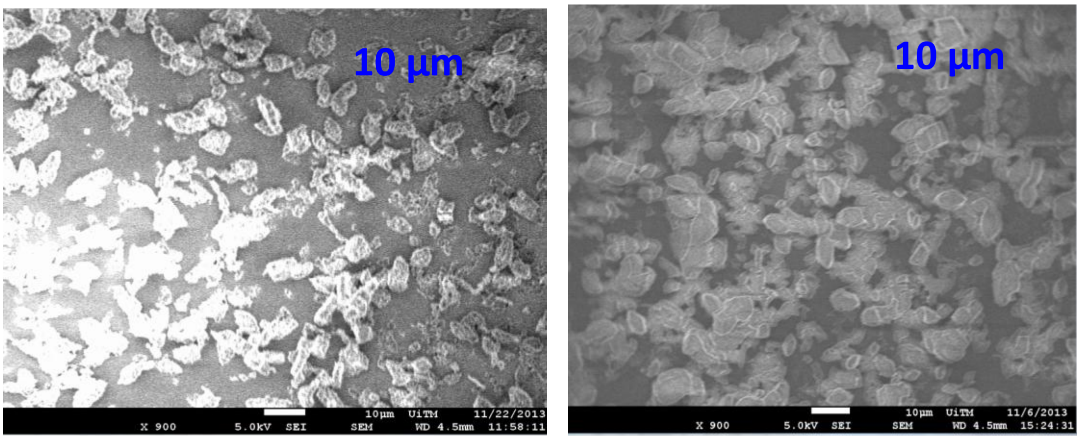


Figure 2. The technique for measure heterojunction solar cell (Au/a-C:B/n-Si/Au)

In the measurement of solar cell device, both sided of silicon (buttom and top) is deposited with approximately 60nm and 12nm of gold, respectively. Another gold with thickness of 60 nm deposited at the top surface of 12 nm gold for confirm the point of probe is properly contacted on the gold metal contact. The light closure is attached at the top of the device as shown in Fig. 2 (dark color) to ensure light strike only the area of 2 cm^2. For complete the whole circuit, the other probe is connected to the conductive metal holder. Solar simulator (Bukuh Keiki EP200), surface profiler (Veeco Dektak 150), JASCO UV-VIS/NIR Spectrophotometer (V-670 EX) and field emission scanning electron microscopic (FESEM, ZEISS Supra 40VP) to characterize the electronic properties, optical properties, and surface morphology, respectively.

III. RESULT AND DISCUSSION



(a) (b)

Figure 3 FESEM images of boron doped amorphous carbon with magnification of 900 at (a) 0 V and (b) -20 V

Fig. 3 (a) and (b) show the FESEM images of complex-structured a-C:B films. The images were taken with magnification of 900 and voltage of 5.0 kV. The images in Fig. 3 (a) shows the irregular pattern of structure synthesized from pure hydrocarbon precursor of palm oil with the micro-structured size. As can be observed from FESEM images, the miro-structured a-C:B film consists of irregularly scattered micro flower-like of agglomerated particles. In contrast, the complex-structured a-C:B film has more finer and denser. As bias voltage of -20 V, the flower-like particles becomes denser and finer. This difference in the surface morphology film can be attributed to the ion bombardment during the growth of the films which is controlled by the applied of negative bias onto the substrate. More specifically, during the deposition of a-C:B film with applied of -20 V, an intense positive-ion bombardment on the growing film surface is occurred. The flux and the energy of these species affect the mechanisms that govern the incorporation of boron in the a-C network and the formation of a-C:B bonding groups [10-12]. The negative bias of -20 V increases the energy of the bombarding ions, enhancing the chemical reactions between different species and their mobility at the growing film surface. As a result, the high-energy gas ions dissociate the deposition carbon clusters affecting the boron distribution, and the bonding structure of the a-C:B film. Accordingly, without bias voltage (0 V) applied to substrate, the ions energy is too weak to penetrate into the growing surface and most of the ions are only trapped on the growing surface, resulting in the formation of the loose cross-linking [13-15] resulting to the irregularly scattered micro flower-like of agglomerated particles.

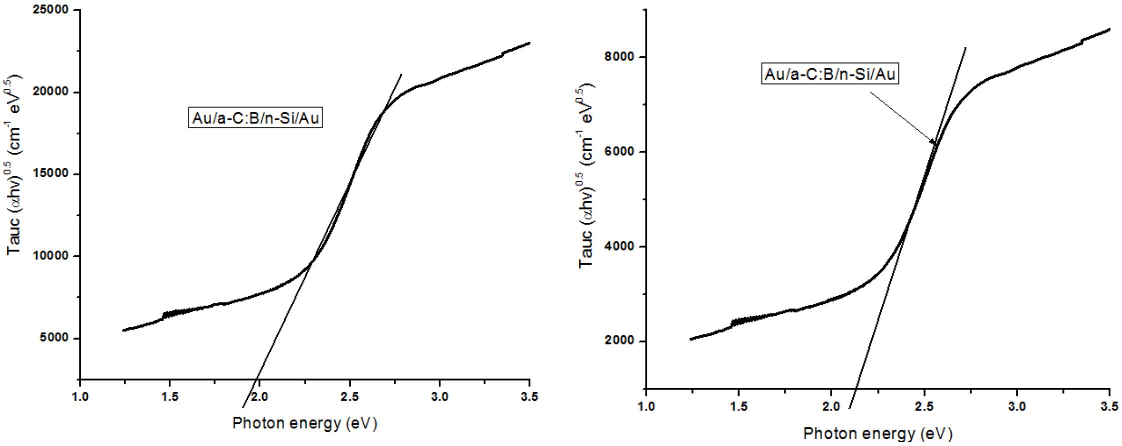


Figure 4 Optical band gaps of boron doped amorphous carbon at 0V and -20V

The optical properties of thin films are investigated by UV-visible spectroscopy measuremets on glass substrates in the range of 350-900 nm wavelength, to derive the Tauc estimated optical band gap (E_g) for amorphous semiconductors [16,17]. The E_g of the thin films is obtained from the extrapolation of the linear part of the curve at the α=0, using the Tauc relation, $(\alpha hv)^{1/2}=B_2(E_g\text{-}hv)$,where B_2 is the Tauc parameter [13,14].The estimated E_g of a-C:B at 0 V and -20 V are approximately 2.1 and 2.0 eV, respectively as shown in Fig. 4 (a) and (b). The optical band gap is decreased as the applied of negative bias voltage of -20 V. The extra energy by negative bias of -20V attributed to more interstitial doping boron (B) into a-C films which modify a-C:B bonding that related with E_g. Similar phenomena causing decreasing of E_g were also reported by using other parameters such as the percentage of dopants, (nitrogen and phosphorous), and deposition temperature [16-18]. The values of these E_g were a good agreement why the surface morphology at -20 V has finer and denser than 0 V.

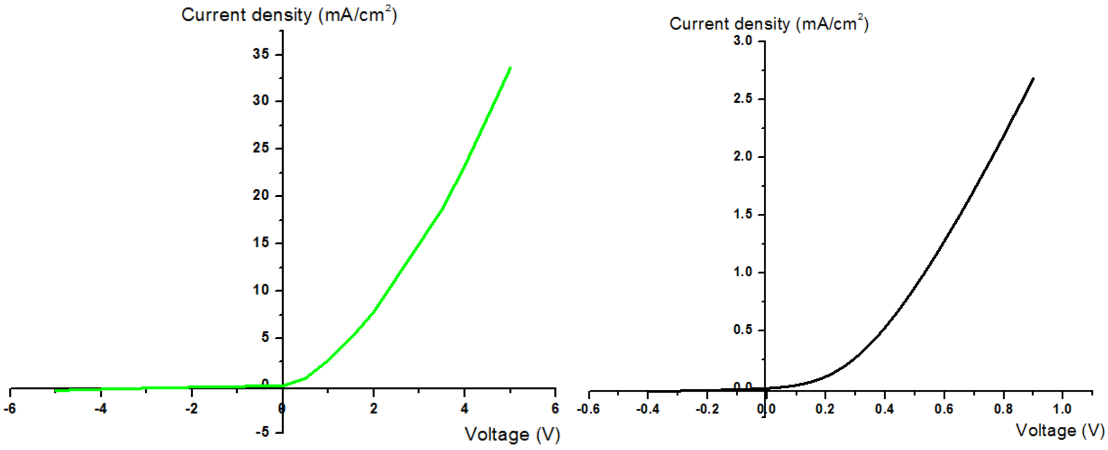


Figure 5 The current-voltage characteristics of Au/a-C:B/n-Si/Au heterojunction solar cell under dark measurement

The current density–voltage (*J–V*) characteristics of Au/a-C:B/*n*-Si/Au heterojunction solar cell device with and without applied Dc bias susbstrate in dark environment are shown in Fig. 5 (a) and (b). The Au/a-C:B/*n*-Si/Au heterojunction solar cells display rectifying curves, which indicate the formation of heterojunction between the a-C:B film and silicon. The a-C:B layers acted as a *p*-type semiconductor with respect to n-type silicon, thus forming the rectifying curve. The reverse saturation current, which is low as compared with the forward current, gradually increases with reverse bias (photocurrent increases). These behaviors can be attributed to the generation of minority carriers within the depletion region. At forward bias, the current increases exponetially, indicating a good quality of *p–n* junction. The ideality factor is approximatelty 2, indicating the dominance of the recombination current rather than the diffusion current. Many deviations from the ideal *p–n* characteristics are observed, which can be due to the high low doping efficiency and posses complex structure and high density of defects [19-21].

The *I–V* characteristics of Au/a-C:B/*n*-Si/Au devices under illumination at 100 mW/cm^2 are illustrated in Fig. 6 (a) and (b). A dissimilar trends in the form of curves are observed for micro-structured film for Au/a-C:B/*n*-Si/Au heterojunction solar cells. The obtained curve of Au/a-C:B/*n*-Si/Au solar cell at 0 V is less broad and small area as compared with the obtained curve of Au/a-C:B/*n*-Si/Au solar cell at -20 V. A slightly broad curve indicates reduced area (fill factor, FF) or maximum output power, and thus, the overall conversion efficiency is minimized. This phenomenon is attributed to series and shunt resistances, which are caused by metal contact and material defect, respectively. The open circuit voltage (V_{oc}), current density (J_{sc}), fill factor (FF) and efficiency (η) of Au/a-C:B/n-Si/Au at 0 V were 254 mV, 234 mA/cm^2, 0.241, and 0.0141 %, respectively. Meanwhile, the V_{oc}, J_{sc}, FF and η of Au/a-C:B/n-Si/Au at -20 V were 426 mV, 5.351 mA/cm^2, 0.243, and 0.553 %, respectively. It was observed that complex-strcutured a-C:B film deposited by -20 V use as the p-type in heterojunction solar cell give higher convension efficiency than complex-strcutured a-C:B film deposited without applied DC bias substrate (0 V).

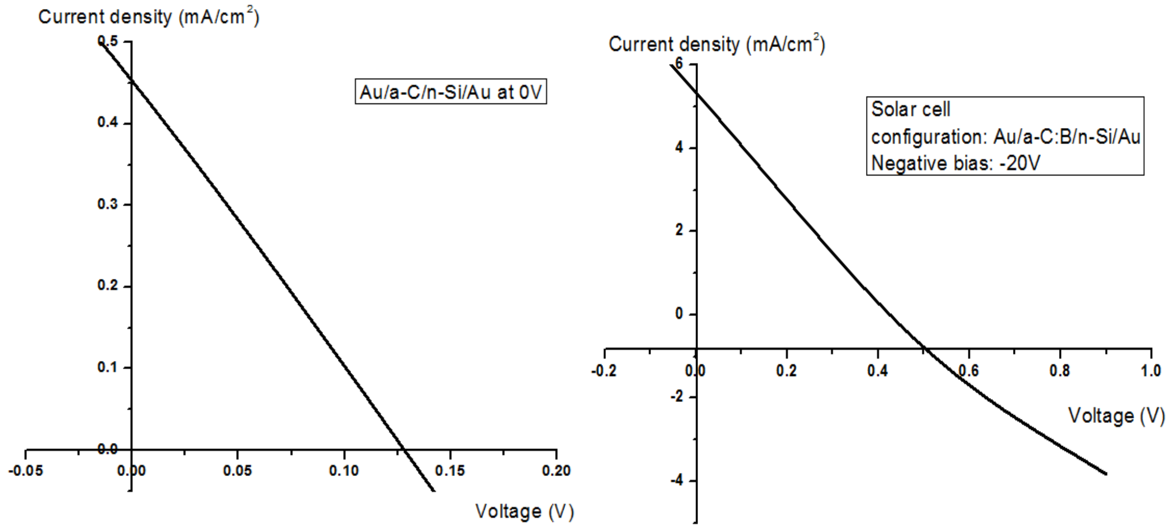


Figure 6 The current-voltage characteristics of Au/a-C:B/n-Si/Au heterojunction solar cell device under illumination

The series resistance results in voltage drop, thus preventing full photovoltaic voltage across the external load; likewise, series resistance affects open circuit voltage (V_{OC}) [8,11,15]. By contrast, internal resistance of material (shunt resistance) is due to the device edges and grain boundaries. Shunt resistance significantly contributes to the reduction of solar cell performance. In shunt resistance, a fraction of photo-generated carries are diverted away from the external load, thereby reducing current density (J_{SC}). Series resistance can be minimized by introducing metal contact based on grid metal arrangement in commercialized silicon solar cells. Although metal contact arrangement in the solar cell configuration in the present study is attributed to high series resistance, which reduces the overall solar cell efficiency, our objective is to prove the complex-structured a-C:B film from hydrocarbon source of natural palm oil can be applied in the fabrication of heterojunction solar cells.

The electronic properties of Au/a-C/*n*-Si/Au solar cell, including its open V_{oc}, J_{sc}, FF, and efficiency are presented in Fig. 6 (b). Low V_{oc} and *J*sc are found for complex-strcutured a-C:B film, which diretly indicate low FF and conversion efficiency. The low V_{oc} and *J*sc values are attributed to the low built-in voltage, which are caused by the high amount of defect in the micro-strcutured a-C film. The electronic properties of Au/a-C:B/*n*-Si/Au solar cells is remarakbly improved by the applied of DC bias substrate at -20 V. The Au/a-C:B/*n*-Si/Au solar cells fabricated through the deposition at -20 V shows the highest conversion efficiency. The improvement of convension efficiency can be attributed to the successful boron incorporation, which increases the number of excess carriers in the nano-strcutured a-C:B film. The bombardment of ions produced by -20 V is important in minimizing defects. The reduction of defects increases the built-in voltage, thereby prolonging the lifetime of excess carriers and providing wide difussion. Although the energy-conversion efficiencies of the fabricated solar cell devices are considerably low, the present study presents a viable alternative through the use of natural palm oil precursor in developing solar cells. Moreover, the energy-conversion efficiency achieved in this study (deposition at -20 V applied bias) is higher than that reported by Tian. (η = 0.3 %) [21] and Hayashi, (η =0.04 %) [22].

The current-voltage characteristics of both Au/a-C:B/n-Si/Au devices, under illumination are shown in Fig. 5 (a) and (b) and their electronic properties are summarized in Table 1. The open circuit voltage (V_{oc}), current density (J_{SC}), fill factor (FF) and efficiency (%) of Au/a-C:B/n-Si/Au at 0V were approximately 0.254 V, 0.234 mA/cm^2, 0.241, and 0.0141%, respectively. Meanwhile, the open circuit voltage (V_{OC}), current density (J_{SC}), fill factor and efficiency of Au/a-C:B/n-Si/Au at -20 V were 0.426 V, 5.351 mA/cm2, 0.243, and 0.553%, respectively. It was observed, convension efficiency of Au/a-C:B/n-Si/Au deposited using low negative bias voltage of -20V is higher than 0 V.The improvement of convension efficiency might be resulted from the number of boron incorporated with a-C to reduce the dangling bond in complex structured of films. The improvement on electronic properties showed negative bias voltage plays an important rule to reconfigure the structure of a-C and boron atoms to a more graphitic semiconductor.

Table I

The Electronic Properties of Au/a-C:B/n-Si/Au Solar cell Fabricated at 0 V and -20 V

Negative bias (V)	**Open-circuit voltage (V_{OC})**	**Current density (mA/cm^2)**	**Fill factor (FF)**	**Conversion efficiency (%)**
0	0.254	0.234	0.241	0.0141
-20	0.426	5.351	0.243	0.553

Fig. 7 shows spectral response of Au/a-C:B/n-Si//Au at applied DC bias voltage of 0 V and -20 V. The Au/a-C:B/n-Si/Au at -20 V shows higher spectral response as compared with the Au/a-C:B/n-Si/Au at 0 V. Spectral responses (0 V and -20 V) indicate two broad bands at the peak wavelength approximately 550 nm and 900 nm of wavelengths. It was reported that the wavelength above 700 nm is negligible since the photocurrent is mostly generated due to the silicon substrate [11,21,22]. In the region below 700 nm, boron doped layers act as carbon photon absorber, and quantum efficiency has a peak in short wavelength region. The photo current peak at approximately 550 nm are the good agreement with the optical band gap (Fig. 4 (a) and (b)) measurement and conversion effciecnt.

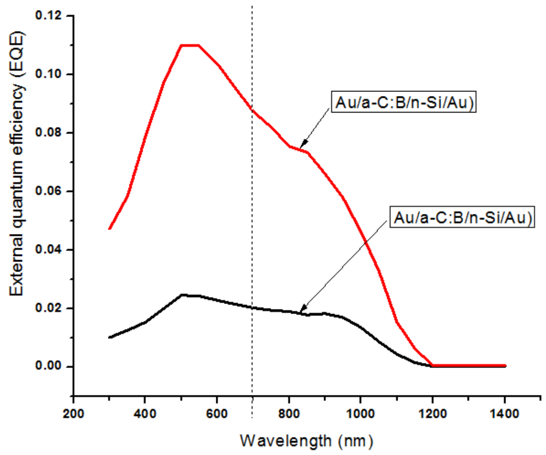


Fig. 7. Spectral response of heterojunction solar cell (Au/a-C:B/n-Si/Au) at different bias voltage

IV. CONCLUSION

The complex-structured of a-C:B films using novel precursor of palm oil for heterojunction solar cell applications was presented. FESEM revealed films deposited with negative bias of -20 V had finer and denser of flower-like particle. Negative DC bias improved the structural properties of a-C:B films and showed the significant increment of energy conversion efficiency by the ion bombardment effect of the negative bias (-20 V). Solar simulator analysis results showed an open circuit voltage (V_{oc}), current density (J_{SC}), fill factor (FF) and efficiency (%) of Au/a-C:B/n-Si/Au at 0V were approximately 254 mV, 0.234 mA/cm^2, 0.241, and 0.0141%, respectively. Meanwhile, the open circuit voltage (V_{OC}), current density (J_{SC}), fill factor and efficiency of Au/a-C:B/n-Si/Au at -20 V were 426 mV, 5.351 mA/cm^2, 0.243, and 0.553%, respectively The conversion efficiency was increased as constant negative bias of -20 V applied for doping boron into a-C film. Although the conversion efficiency of heterojunction solar cells are considerably low, it shows a good prospect of using palm oil as the carbon source with the help of negative bias substrate for fabricated a-C:B film as a p-type on n-silicon substrate in heterojunction solar cell device for the improvement of the energy conversion efficiency by optimizing the deposition conditions.

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