

The Optical Energy Gap of the Semiconducting Intrinsic Layer for Organic Solar Cell Application

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Abstract. The optical energy gap of the semiconducting intrinsic layer plays an important role in increasing efficiency of the material. The carbon-based biomass can be an alternative to the silicon used as material in the solar cell. Here, we investigate the best biomass that can be used as a semiconductor part in solar cell applications. Coconut shells as bio-waste and palmyra sap, which is available in most areas of Indonesia, can be the best candidates to be evaluated. The methodology that is being used here involves a carbonization process at high temperatures of around 300°C for 2 hours and 400°C for 5 hours to obtain high-carbon charcoal for palmyra sap and coconut shells, respectively. The XRD measurement showed both organic materials have an amorphous phase, and for coconut shells sample, has two broad peaks that are identical with graphene peaks, therefore, this material is called graphenic-like carbon (GC). Furthermore, from the UV-visible spectroscopy, it was shown that both materials have a high transmittance of more than 95%, which indicates that they have transparant properties. Also by the Tauc plot method, it gives information about the optical energy gap of coconut shell charcoal (GC) and palmyra sap (a-C) around 2.67 and 1.83 eV, respectively. From this result, both raw materials are in the range of semiconductors, and for palmyra sap material, it can be the best candidate to be applied as an intrinsic layer for semiconducting parts in solar cell applications.

Keywords: Amorphous phase, coconut shells charcoal, optical energy gap, palmyra sugar

Introduction

The transition to renewable energy is very necessary since fossil fuels produce a lot of harmful gases, such as carbon dioxide, which can be one of the factors contributing to climate change. There are many types of renewable energy; one of them is photovoltaic technology, which is considered a great alternative energy due to its ability to combat the negative effects produced by non-renewable fossil fuels [1], [2]. The most important indicator used to evaluate the performance of photovoltaic technology is conversion efficiency, which express by the ratio of electrical energy output to solar energy input [2]. The research that was done by Hamdani et al. through simulation revealed that the optical energy gap plays a crucial role instead of the thickness of every layer that forms the junction in order to increase the efficiency of solar cells [3]. It is also strengthened by the papers from Ishak et al., who said that the optimum optical energy gap for obtaining high-efficiency solar cells is in the range of 1 to 2 eV [4].



The research on semiconductor materials that can be applied to solar cell applications has been intensively studied by many researchers around the world. Silicon is the common material for solar cells, but silicon has several disadvantages, such as being easy to degrade, high cost, being difficult to fabricate, and sometimes needing such a harmful material for the environment while synthesizing silicon to be applied as a semiconductor layer [5]. Furthermore, the development of carbon-based materials to replace silicon has been conducted worldwide since they are cheaper for the raw material itself and easier for the fabrication process with excellent properties [6]. The amorphous carbon (a-C) that was developed by Priyanto et al. was successful, using palmyra sap as a source in order to make carbon-based material for a solar cell application. For the details, they made a homojunction solar cell, and they showed unique properties because they were more environmentally friendly, low cost, light weight, easily processed, and flexible in tuning their optical energy gap by the doping process. From their research, it showed the appearance of electricity that was conducted inside the biomass, as proven by the efficiency value of their homojunction solar cell, which is around 0.0037% for an active area of around 1x1 cm². Even though the efficiency value still seems lower than other organic solar cells, it was a groundbreaking experiment because the biomass was supposed to be an insulator, so no one paid attention to that material. The efficiency was lower due to their experiment, which was done without any optimization either in the thickness or optical energy gap value [7], [8]. Therefore, the investigation of the optimum optical energy gap is necessary before applying it to the homojunction solar cell.

The objective of our research is to investigate the semiconducting intrinsic material that is appropriate to be used for solar cell applications by investigating the optical energy gap. The raw materials that will be used here come from biomass, which is palmyra sap from Situbondo regency in East Java and coconut shells that were obtained from local markets as bio-waste. The carbonization process that is used to obtain high-carbon charcoal from palmyra sap is a little bit different from the previous study that was done by Priyanto et al. [7] in order to ensure the best way to result in optimum optical energy gap.

Experiments

We used two different kinds of raw materials: the first is palmyra sap that can be obtained from palmyra trees, and the second is coconut shell charcoal, which can be obtained from the burning process of coconut shells in the atmosphere. For the first raw material, the palmyra sap must first be cooked until it becomes thick using a hot plate atat 350 °C for around 15 minutes. As is already well known, palmyra sugar has a character that makes it easily melt even at room temperature; therefore, the cooking process is necessary here before the carbonization process to reduce the water content inside the sample. Then, the carbonization process can be applied for the next step using a furnace at a temperature of around 250 °C for 2.5 h until it becomes shiny black charcoal. After that, the sample was ground and sieved to obtain a homogeneous powder using a 400-mesh siever. After the siever process, the charcoal was washed with distilled water and filtered three times to remove the impurities, especially the KCl content that is usually found inside the palmyra sap after the heating process. The charcoal powder was dried using a hot plate at 70 °C. Afterwards, to obtain high-carbon charcoal, we have to do the second carbonization process at 300 °C for 2 h, resulting in the amorphous carbon powder from palmyra sap.

The second raw material is coconut shells that burned in the atmosphere, resulting in black charcoal, also called coconut shell charcoal. Here, we used old coconut shells because they contain many carbon atoms inside. It was collected by the local food market as biowaste. The raw



Computational and Experimental Research in Materials and Renewable Energy (CERiMRE Volume 6, Issue 1, page 14-21 eISSN : 2747-173X

Submitted : May 14, 2023 Accepted : May 29, 2023 Online : May 31, 2023 DOI : 10.19184/cerimre.v6i1.39254

material was cleaned and cleared of the husk before being dried for three days using solar thermal energy. Then, the clean shell was carbonized at 400 °C for 5 hours to produce high-carbon coconut shell charcoal. The sample was ground and sieved to obtain a homogeneous powder using a 200-mesh siever. Additionally, an ultrasonic cleaner was used to sonically exfoliate the distilled water in order to create a thinner graphitic layer. The wet powders of coconut shell charcoal were then dried at room temperature for 24 hours. The characterizations that are used in this work are XRD measurement and UV-Vis spectroscopy. The amorphous phase from both raw materials will be confirmed using XRD measurements with Cu-K α radiation as a source (λ = 0.154056 nm) in the range of 2 θ around 5° to 60°, and UV-Vis spectroscopy in the wavelength range of 200–1100 nm to analyze the absorbance and also obtain the optical energy gap using the Tauc Plot method from those samples.

Result and Discussion

The XRD measurement was used to confirm the phase of samples by analyzing the diffraction pattern in a specific range based on the properties of each material. In this case, we use the XRD in the range of $2\theta = 5.60^{\circ}$ for palmyra sap sample and $2\theta = 5.50^{\circ}$ for coconut shells charcoal sample, as shown in Figure 1. The XRD data are obtained by using CuK α radiation with(λ = 0.15406 Å, showing the broad peak for both samples, indicates that the phase is amorphous [9]. The differences in the range of 2θ value from both raw materials is not that necessary, since the broad peak from the biomass materials is commonly observed around 15-45° [9], [10], but we are using up to 60° in order to check if maybe the material is showing another peak due to the impurity compounds that remain after the carbonization process. According to the coconut shell charcoal diffaction patterns, it shows two broad peaks located at $2\theta \sim 24$ and 43° , respectively, and those peaks are identic with the graphene peak; therefore, we can say this material as graphene-like carbon (GC) [11]. Afterwards, the palmyra sap only showed one peak at $2\theta \sim 17^{\circ}$, therefore, we can just say this material is amorphous carbon (a-C). These peaks also seem similar to the previous study that was done by Risitiani et al. [11] and Priyanto et al. [7] using similar raw materials but different local markets, which means if we repeat the similar method even with a different place to obtain the raw materials, we will get the same result [12], [13].



Figure 1. XRD pattern of (a) GC sample, (b) a-C sample



UV-visible characterization was used to determine either absorbance or transmittance from all the samples. The absorbance and transmittance are given in Figure 2 (a) and (b), which show either the intensity of light absorbed or transmitted by the particle in the solution of carbon samples. The result of UV-Vis measurement showed that both samples have low absorbance and high transmittance in the range of UV light to visible light wavelengths. The absorbance of coconut shell charcoal and palmyra sap is 0.6 to 0.8%, respectively, which indicates that those materials are transparent. It also gives information that transmittance is the opposite of absorbance value, which means that when the absorbance rate is low, the transmittance of the samples is higher. The relation between absorbance and transmittance is also given by the mathematical equation shown in equation (1) below.

$$A = \log \frac{1}{T} \tag{1}$$

where *A* is absorbance data and *T* is transmittance data. From this equation, it is clearly seen that transmittance data is inversely proportional to absorbance data. The absorption peaks that appeared at 288 and 311 nm in the palmyra sap and coconut shells, respectively, relate to transitions of π to π^* orbital [9]. If we compared the absorbance and transmittance values in both samples, especially in the range of visible light wavelengths, it showed that the palmyra sap sample has a lower absorbance than the coconut shell charcoal sample, which indicates that palmyra sap has more potential to be applied in the solar cell application since the majority of light carried out by the sun is visible light. This result is also strengthened by the optical energy gap that can be obtained using the Tauc Plot method, which is for the equation shown in equation (2) below [14].

$$(\alpha hv)^n = K (hv - E_g) \tag{2}$$

where α is absorption coefficient, hv is the foton energy, n is determined from the DFT calculation which is for material that has direct optical energy gap, the n value is 2 whereas for indirect optical energy gap, the n value is $\frac{1}{2}$, K is the energy-independent constant, and E_g represent the optical energy gap. The value of hv can be obtained by Plack equation which is around 1240, whereas for α can be obtained using the Beer Lamberts' Law as long as the equation shown in equation (3) below [14].

$$\alpha = A \frac{1}{\log(e)L} \tag{3}$$

The value of log (e) is 0.4343, and L is the cuvet path length (usually around 1 cm). The value of the optical energy gap (E_g) can be obtained from the intercept of the linier line on the x-axes by fitting the Tauc Plot equation $(\alpha hv)^n$ toward the hv plot as shown in the dotted line in Figure 3 For the coconut shell charcoal (GC sample), due to the characteristic similar to that of graphene, the DFT calculation showed that this material has a direct optical energy gap type, whereas for the palmyra sap, the band structure of amorphous carbon shows an indirect optical energy gap type [15], [16]. According to Figure 3, the optical energy gaps of GC and *a*-C were estimated to be around 1.83 and 2.67 eV, respectively. The optical energy gap represents how much energy is needed for the electron in the valence band to move into the conduction band [17]. Since electricity can be generated by the amount of electrons in the conduction band, it is important to



know how easy it is for the electron to jump into the conduction band; therefore, it is desirable to know the optical energy gap by using the Tauc Plot method as a result of absorption data from UV-Vis spectroscopy. Besides, we also have to know the best optical energy gaps that are needed for solar cell applications in order to achieve the highest efficiency. The optical energy gap data showed that both materials are classified as semiconductor [18]. According to the prior study, it shows that the optimum optical energy gap for a semiconducting intrinsic layer to be applied as an i-layer in a solar cell application must be in the range of 1.5–1.8 eV; thus, based on the optical energy gap data of palmyra sap, it can be the best candidate to be applied as an i-layer in a solar cell application [4]. But remember, instead of the optical energy gap, there are many factors to investigate in order to determine the best i-layer before it can be applied in a solar cell, such as the thickness, the recombination process that might occur in the material, and the type of junction that will be made; therefore, this research can be one of many references to consider biomass as an i-layer based on the optical energy gap investigation [3], [19]. The investigation of the thickness was also done by previous researchers using a-Si:H as an i-layer, resulting that the thickness influences the efficiency of solar cells [3]. But in this case, there is no report yet about the thickness investigation using biomass as an amorphous carbon source in order to check the influences on the performance of solar cells. The simulation using biomass for organic solar cell application is needed for further research to obtain the optimum factors that contribute to the improvement of efficiency in solar cells.



Figure 2. UV-Vis result of (a) GC and (b) a-C samples



Computational and Experimental Research in Materials and Renewable Energy (CERiMRE Volume 6, Issue 1, page 14-21 eISSN : 2747-173X



Figure 3. Optical gap energy of GC and *a*-C samples using Tauc Plot method

Conclusion

This research to study the comparison between semiconducting intrinsic layers from two different raw materials, which are coconut shells and palmyra sap charcoal, has been completed. The XRD patterns exhibit a broad peak for palmyra sap powder and two broad peaks for coconut shell charcoal powder, which indicates that both materials have an amorphous phase, and for coconut shell charcoal, these broad peaks are identic with graphene; therefore, it is called graphenic-*like* carbon (GC). Afterwards, the UV-Vis result showed either low absorbance or high transmittance, which indicates that these materials are transparent. Furthermore, using the Tauc Plot method gives information about the optical energy gap, which is for *a*-C and GC was estimated to be 1.83 and 2.67 eV, respectively. From the optical energy gap value, it was shown that both samples are in the range of semiconductor materials. Then the *a*-C sample from palmyra sap can be the best candidate to be applied as an i-layer in solar cell applications.

ACKNOWLEDGMENT

This research was partially supported by National Competitive Research Grant, provided by The Ministry of Education, Culture, Research and Technology Indonesia 2021-2022 (*Hibah Penelitian Dasar Kompetitif Nasional 2021-2022, Kemdikbudristek, Indonesia*).

References

- D. S. da Silva, A. D. S. Côrtes, M. H. Oliveira, E. F. Motta, G. A. Viana, P. R. Mei, and F. C. Marques, "Application of Amorphous Carbon Based Materials as Antireflective Coatings on Crystalline Silicon Solar Cells," *Journal of Applied Physics*, vol. 110, 2011, doi: 10.1063/1.3622515.
- [2] J. Pastuszak and P. Węgierek, "Photovoltaic Cell Generations and Current Research Directions for Their Development," *Materials*, vol.15, no. 16, 2022, doi: 10.3390/ma15165542.



- [3] D. Hamdani, Y. Cahyono, G. Yudoyono, and D. Darminto, "Performances Analysis of Heterojunction Solar Cells through Integration of Hydrogenated Nanocrystalline Silicon Bilayer by Using Numerical Study," *Molecular Crystals and Liquid Crystals*, vol. 725, no. 1, pp. 1-20, 2021, doi: 10.1080/15421406.2021.1922226.
- [4] A. Ishak, A. N. Muhamad, and M. Rusop, "Optical Properties Of As-Deposited Amorphous Carbon Film Fromvarious Substrate Temperaturesvia Custom-Made-CVD," *International Journal of Scientific & Technology Research*, vol. 4, pp. 257-261, 2015.
- [5] A. N. Fadzilah, K. Dayana, and M. Rusop, "Carbon-Based Solar Cell from Amorphous Carbon with Nitrogen Incorporation," *AMR*, vol. 576, pp. 785-788, 2012, doi: 10.4028/www.scientific.net/AMR.576.785.
- [6] L. X. Chen, "Organic Solar Cells: Recent Progress and Challenges," *ACS Energy Lett*, vol. 4, no. 10, pp. 2537-2539, 2019, doi: 10.1021/acsenergylett.9b02071.
- B. Priyanto et al., "Hydrogenated Amorphous Carbon Films from Palmyra Sugar," *Journal of Renewable Materials*, vol. 9, no. 6, pp. 1087-1098, 2021, doi: 10.32604/jrm.2021.014466.
- [8] K. Nadiyyah, A. Z. Laila, I. S. Ardiani, B. Priyanto, and Darminto, "Electrical Characterization of N- and B- Doped Amorphous Carbon Film from Palmyra Sugar," *KEM*, vol. 860, pp. 196-201, 2020, doi: 10.4028/www.scientific.net/KEM.860.196.
- [9] D. I. Pamungkas, A. Haikal, M. A. Baqiya, Y. Cahyono, and Darminto, "Synthesis of Amorphous Carbon from Bio-Products by Drying Method," *AIP Conference Proceedings*, vol. 1945, 2018, doi: 10.1063/1.5030281.
- [10] F. Astuti, N. Sari, V. L. Maghfirohtuzzoimah, R. Asih, M. A. Baqiya, and D. Darminto, "Study of the Formation of Amorphous Carbon and RGO-like Phases from Palmyra Sugar by Variation of Calcination Temperature," *JFA*, vol. 16, no. 2, pp. 91-94, 2020, doi: 10.12962/j24604682.v16i2.6706.
- [11] D. Ristiani, R. Asih, F. Astuti, M. A. Baqiya, C. Kaewhan, S. Tunmee, H. Nakajima, S. Soontaranon, and Darminto, "Mesostructural Study on Graphenic-Based Carbon Prepared from Coconut Shells by Heat Treatment and Liquid Exfoliation," *Heliyon*, vol. 8, no. 3, 2022, doi: 10.1016/j.heliyon.2022.e09032.
- [12] B. Priyanto, R. Asih, I. S. Ardiani, A. Z. Laila, e, "Hydrogenated Amorphous Carbon Films from Palmyra Sugar," *Journal of Renewable Materials*, vol. 9, no. 6, pp. 1087-1098, 2021, doi: 10.32604/jrm.2021.014466.
- [13] D. Ristiani, P. Y. D. Maulida, A. A. Firdaus, R. Asih, F. Astuti, M. A. Baqiya, S. Tunmee, H. Nakajima, and D. Darmino, "Structural Investigation of Boron-and Nitrogen-Doped Reduced Graphene Oxide Prepared by Wet Mixing Method," *AIP Conference Proceedings*, 2023, doi: https://doi.org/10.1063/5.0115792.
- [14] E. H. Sujiono, Zurnansyah, D. Zabrian, M. Y. Dahlan, B. D. Amin, Samnur, and J. Agus, "Graphene Oxide Based Coconut Shell Waste: Synthesis by Modified Hummers Method and Characterization," *Heliyon.* vol. 6, no. 8, 2020, doi:. 10.1016/j.heliyon.2020.e04568.
- [15] *T. Soga, T. Nakagaki, S. Kat*o, and N. Kishi, "Effect of Sublimation Temperature on the Photovoltaic Properties of Amorphous Carbon Thin Films from Fullerene," *J. Solar Eneg. Res. Updat,* vol. 5, pp. 8-13, 2018.



- [16] P. N. O. Gillespie and N. Martsinovich, "Origin of Charge Trapping in TiO₂ /Reduced Graphene Oxide Photocatalytic Composites: Insights from Theory," ACS Appl. Mater. Interfaces, vol. 11, no. 35, pp. 31909-31922, 2019, doi: 10.1021/acsami.9b09235.
- [17] S. K. Tripathy and A. Pattanaik, "Optical and Electronic Properties of Some Semiconductors from Energy Gaps," *Optical Materials,* vol. 53, pp. 123-133, 2016, doi: 10.1016/j.optmat.2016.01.012.
- [18] S. M. Sze, Semiconductor Devices, 2022, *Physics and Technology, 2nd ed*, New York, Wiley.
- [19] P. Jelodarian and A. Kosarian, "Effect of P-Layer and i-Layer Properties on the Electrical Behaviour of Advanced a-Si:H/a-SiGe:H Thin Film Solar Cell from Numerical Modeling Prospect," *International Journal of Photoenergy*, vol. 2012, pp. 1-7, 2012, doi: 10.1155/2012/946024.