THE EFFECT OF SYNTHESIS STEP AND HEATING TEMPERATURE ON SYNTHESIS OF MATERIALS $CH_3NH_3PbI_3$

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Abstract

Research on the effeect of one-step and two-step methods on the electronic structure and character of perovskite material $CH_3NH_3PbI_3$ has been carried out, as well as the effect of temperature variations on the perovskite material $CH_3NH_3PbI_3$. Synthesis with one-step method was carried out by dissolving PbI_2 in DMF (taken by filtrate) and dissolving CH_3NH_3I in isopropanol then the filtrate and CH_3NH_3I solutions were mixed and stirring was carried out. After the two mixed solutions are then coated on a glass substrate that has been coated with compact-TiO_2 dan m-TiO_2, heating on the hotplate at 130°C for 15 minutes. The two-step method was carried out by dissolving PbI_2 in DMF (taken by filtrate), then coating it on a glass substrate, then dissolving CH_3NH_3I in isopropanol and overlaying PbI_2 then heated on a hotplate at 130°C for 15 minutes. Optimal temperature determination is carried out according to the one-step synthesis, heating temperature above the hotplate 100°C, 110°C, 130°C, and 150°C. The synthesis material was then characterized using XRD and UV-Vis.The XRD characterization results showed that $CH_3NH_3PbI_3$ material synthesized in one step and two stages had an effect on the crystal size of 29.0567 nm and 22.60172 nm, while for amorphous crystal structure temperature variations. The results of UV-Vis analysis result obtained absorbance data and band gap energy. One-step synthesis method produces a maximum absorbance of 745 nm, a synthesis method of two-step 733 nm, for the maximum temperature variation of absorbance data at 100°C, 110°C, 130°C, and 150°C is 585 nm, 585 nm, 481 nm, and 484 nm. Band gap energy has an effect on the synthesis of $CH_3NH_3PbI_3$ material with one-step and two-step. In the one-step synthesis method, the band gap energy is 1.543 eV and the two-step synthesis method obtained 1.614 eV, while for temperature variation produces 2.67 eV to 3.74eV.

Keywords: $CH_3NH_3PbI_3$, One-Step, Two-Step, Temperature Variation

INTRODUCTION

Energy needs are increasingly increasing, causing the availability of energy is increasingly depleting, especially energy derived from fossils. In 2014, proven oil reserves of 3.6 billion barrels, natural gas at 100.3 TCF and coal at 32.27 billion tons. If the energy is consumed continuously and there is no renewable energy reserves, based on the R / P (Reserve / Production) ratio, the energy derived from petroleum will run out in 12 years, natural gas 37 years, and coal 70 years (Sugiyono , et al, 2016).

Increasing energy needs, it is necessary to have alternative energy that can meet the demand for energy sources in the community. Alternative energy that can replace fossil energy is renewable energy, one of which is energy derived from the sun. Technology that can convert solar energy into electrical energy is a solar cell, which is a device that can convert solar energy directly into electrical energy that does not produce exhaust gases or pollution (Charr, Lamont, & Zein, 2011).

One type of solar cell that has recently attracted the attention of researchers is perovskite solar cells, in the last 5 years the development of perovskite solar cells has reached an efficiency of 20.1% (Gunther, 2015). Perovskite material is an organic-organic halide compound with ABX_3 structure where A is an inorganic cation, B is a metal cation and X is an anion (Graef., Et al, 2007). The structure of this perovskite compound produces a small band gap energy so that it can be used as an absorber. Solar cells from organic -organic halide perovskite material have a greater absorption coefficient than other solar cells so they can absorb more sunlight. Large diffusion lengths and low binding energy are also other advantages of perovskite solar cells (Gao. Et al. 2012).

$CH_3NH_3PbI_3$ material has produced a power conversion efficiency of 20% (Bi, et al., 2016). $CH_3NH_3PbI_3$ perovskite material can be synthesized by several methods, including one-stage and two-stage synthesis methods. Nida, F., et al (2016) has conducted a one-step spin coating research, which is by mixing perovskite material, then coating. Rahmanita, S., et al., (2015) have conducted perovskite material research with a two-step method (two step spin coating), namely by superimposing perovskite material on the glass and then coated with other perovskite material in the hope that the two layers can be converted into perovskite material has a higher energy conversion efficiency.

In this study synthesis of $CH_3NH_3PbI_3$ perovskite material was carried out with one-stage and two-stage synthesis methods, the most effective results of which would be to temperature variations to see the most optimal temperature in the synthesis of the material. The synthesis results were then characterized using X-Ray Diffraction (XRD) to determine lattice parameters, crystal size and crystal structure formed, UV-Vis to determine absorbance and band gap energy (Eg).

RESEARCH METHODS

The research subjects were perovskite $CH_3NH_3PbI_3$ material. The object of research is the character and crystal structure and the influence of the $CH_3NH_3PbI_3$ electrical character of the perovskite material. The materials used in this study are CH_3NH_3I, PbI_2, Isopropanol, DMF, compact-TiO_2, m-TiO_2, glass preparations, aluminum foil. The tools used include glassware, drop pipettes, measuring pipettes, tweezers, flakon bottles, analytical scales, spatulas, stirrers & heaters, XRD, and UV-Vis.

Research Procedure

a. $CH_3NH_3PbI_3$ Material Synthesis with One-Stage Method

The $CH_3NH_3PbI_3$ layer was prepared by dissolving 0.0013 grams of CH3NH3I dissolved into 0.2 mL of isopropanol, stirring until homogeneous. After that, 0.094 grams of PbI2 were dissolved into 0.2 mL of DMF by stirring until mixed, the filtrate from the solution was taken. Then the CH3NH3I solution and the filtrate were mixed and stirred until homogeneous, so that a $CH_3NH_3PbI_3$ solution was formed. The solution is then coated on a glass preparation that has been coated with compact-TiO_2 and m-TiO_2, then heated on a hotplate at 130°C for 15 minutes. After that, characterization with XRD and UV-Vis was carried out.

b. $CH_3NH_3PbI_3$ Material Synthesis with Two-Stage Method

The perovskite $CH_3NH_3PbI_3$ material was prepared by dissolving 0.0013 grams of CH_3NH_3I into 0.2 mL of isopropanol, stirring until homogeneous. 0.094 grams of PbI_2 were dissolved in 0.2 mL DMF, stirring was carried out until mixed, then the filtrate from the solution was taken. Synthesis is carried out on glass coated with compact TiO_2 and m-TiO_2. PbI_2 solution is coated on the glass, then CH_3NH_3I solution is coated over the PbI_2 layer. The substrate obtained was then heated on a hotplate at 130°C for 15 minutes. Substrate was then characterized by X-Ray Difraction (XRD) and UV-Vis.

c. $CH_3NH_3PbI_3$ Material Synthesis with Heating Temperature Variations

Material synthesis was carried out according to the $CH_3NH_3PbI_3$ material synthesis procedure with a one-step method with heating temperature variations above the hotplate with variations in temperatures of 100°C, 110°C, 130°C, and 150°C for 15 minutes, respectively. After that, characterization with XRD and UV-Vis was carried out.

RESULTS AND DISCUSSION

This research was conducted to obtain perovskite material $CH_3NH_3PbI_3$. $CH_3NH_3PbI_3$ material uses CH_3NH_3I and PbI_2 precursors. $CH3NH^{2+}$ as an organic cation, Pb^{2+} as a metal cation and I^- as an anion. In the synthesis process of $CH_3NH_3PbI_3$ material, the preparation glass used for synthesis has been coated by a layer of compact-TiO_2 and m-TiO_2. The m-TiO_2 layer functions to carry electrons, due to the breaking of the electron bond with the hole towards the negative electrode and compact-TiO_2 functions to pull the electron trapped in the m-TiO_2 layer.

The synthesis results of $CH_3NH_3PbI_3$ perovskite material were characterized by X-ray Diffractometer (XRD), using a radiation source of Cu-Kα, at a range of 2θ that is 2° to 80° with intervals of 0.02 and rate 2. The results of the characterization of this diffractogram can be used to determine lattice parameters, crystal structure and crystal size of $CH_3NH_3PbI_3$ material. XRD results that show the presence of narrow and sharp peaks, indicate that the material obtained has a crystal structure.

The XRD characterization results with one-stage and two-stage synthesis methods are presented in Figure 1.

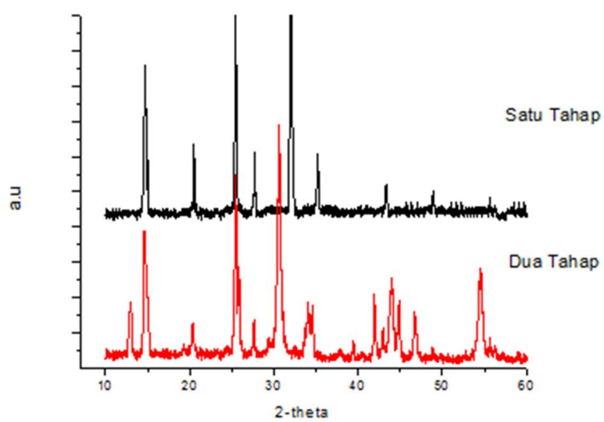

Figure 1. Difraktogram $CH_3NH_3PbI_3$ with one-step and two-step methods.

From the diffractogram above, it can be seen that in the synthesis method one stage there is a diffraction peak at 2θ in the area of 14.5°; 20.5°; 25°; 28°; 32.5°; 35.5°; 44.5° which is the peak of perovskite material (COD no. 8000234). In the two-stage method, 2aksi diffraction peaks appear in the area of 12.8°; 14.5°; 20.5°; 25°; 28°; 32.5°; 34.5°; 35.5°; 39.5°; 42.6°; 43.8°; 44.5°; 45.2°; 54.1° which is at a diffraction peak of 12.8°; 39.5°; 42.6°; 43.8°; 45.2°; and 54.1° is the peak of PbI_2 (COD 9009140). This indicates that the synthesis with two stages still contains PbI_2 which has not reacted completely with the CH_3NH_3I compound. The presence of PbI_2 in perovskite causes obstacles for electrons and holes to move, because PbI_2 is an insulating material, resulting in low conversion efficiency of solar cells (Rahmanita, 2015). From the two coatings, one method for synthesis is taken with heating temperature variations on the hotplate, the method used is one stage of coating.

The results of the XRD analysis were also analyzed using the Rietica program. The results obtained in the form of orthorhombic Crystal system, with Gof 0.340 for one stage and 0.252 for two stages. The crystal size produced from this study is 29.0567 nm in the one-stage method, and 22.60172 in the two-stage method.

The results of XRD analysis on the synthesis of $CH_3NH_3PbI_3$ material with variations in heating temperature produced an amorphous crystal structure, because there were no peaks in the results of the analysis. The results of the analysis are presented in Figure 2.

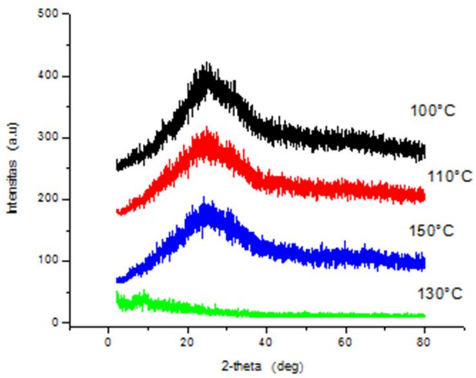

Figure 2. Difractogram $CH_3NH_3PbI_3$ material with heating temperature variations

From the XRD results, Figure 2 shows that the crystalline structure of the synthesis results in an amorphous structure because $CH_3NH_3PbI_3$ material has hygroscopic properties that affect the formation of crystal structures. In one-step and two-step synthesis crystalline structures are formed because the characterization process is direct, while for indirect temperature variations are characterized so that it is possible to interact with air.

Organic-inorganic perovskite material is strongly influenced by moisture. Based on research conducted by Zhou., Et al. Perovskite synthesis and fabrication processes must be at low humidity (relative humidity) RH ≤ 30% to produce perovskite with good structure and ability to conduct electricity. In high humidity, very young perovskite material is degraded because the organic parts contained in CH_3NH_3 material are hygroscopic. This research was carried out synthesis of material in open space so that air contact with high humidity occurs, H_2O in the air binds to NH_3 very strongly which prevents the formation of bonds between CH_3NH_3I and PbI_2.

The synthesized material was also characterized by UV-Vis spectroscopy. Analysis of the data obtained in the form of absorbance data and transmittance. The absorbance data of CH3NH3PbI3 material with one-stage and two-stage synthesis methods are presented in Figure 3.

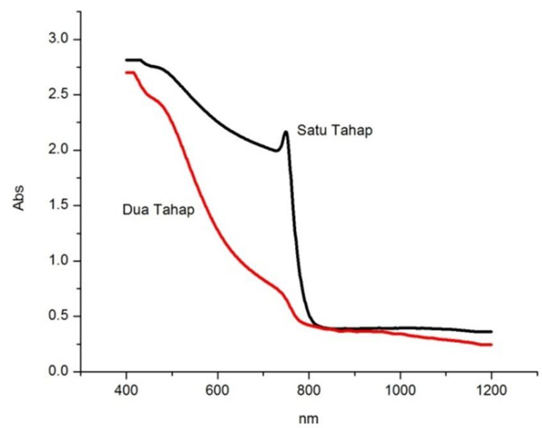

Figure 3. spectra UV-Vis with one-step and two-step methods.

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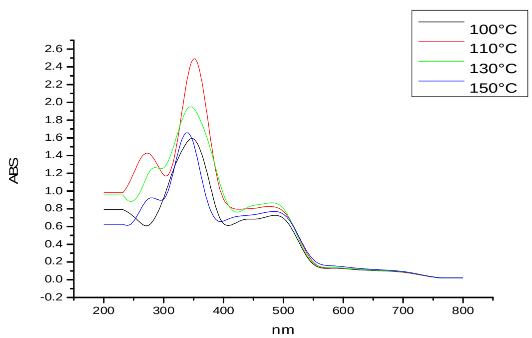

Figure 4. Spectra UV-Vis material $CH_3NH_3PbI_3$ with temperature variations

Figure 4 shows the maximum absorbance of the active synthesis of CH3NH3PbI3 material in the Visible Ray area (Visible). It is proven that the absorbance of the synthesized material has a wavelength between 481 nm-585 nm. Absorbance data from synthesis results with heating temperature variations are presented in table 1.

Variasi suhu pemanasan material $CH_3NH_3PbI_3$	λ (nm)
100°C	585
110°C	585
130°C	481
150°C	484

Table 1. Data Absorbance of heating temperature variations of $CH_3NH_3PbI_3$ material

The results of the analysis with UV-Vis are also used to calculate band gap energy. The results of band gap energy calculation on the synthesis of CH3NH3PbI3 material with variations in heating temperature of 100°C, 110°C, 130°C, and 150°C are presented in Table 2.

Variasi Suhu	Band Gap
100°C	3,65 eV
110°C	3,74 eV
130°C	2,67 eV
150°C	2,82 eV

Table 2. Band gap energy material $CH_3NH_3PbI_3$ with heating temperature variations.

Based on the theory, the band gap energy for $CH_3NH_3PbI_3$ material is 1.5 eV. The results of the analysis with variations in heating temperature obtained band gap energy above 2.5 eV, then the synthesis results are not suitable from the existing theory. This can happen because the material is hygroscopic so it affects the crystal formation.

CONCLUSIONS

Based on the results of the research carried out, the one-step and two-stage methods for the synthesis of perovskite $CH_3NH_3PbI_3$ material affect the band gap energy. From these two methods the results of the band gap energy calculation approach the theory and the previous research was a one-stage synthesis method. The crystal structure obtained from the research results is amoft, the effect of temperature can only be seen from the band gap energy obtained from the synthesis of perovskite material $CH_3NH_3PbI_3$. If the temperature is too low or too high the bandgap energy is produced. So band gap energy is close to the theory and previous research at 130°C.

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