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Processing Temperature Effect on Optical and Morphological Parameters of Organic Perovskite $CH_3NH_3PbI_3$ Prepared Using Spray Pyrolysis Method

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Renewable energy is actually in exponential evolution to find out a natural, low price, secured and safe alternative for fuel energy resources. Recently, different layer deposition methods of perovskite materials are investigated in photovoltaic applications in order to understand the effect of ambient parameters on solar cell characteristics such as short circuit current density (J_{sc}), open circuit voltage (V_{oc}), fill factor (*FF*) and power conversion efficiency (*PCE*). In this paper, the spray pyrolysis deposition method with a moving nozzle has been used to fabricate the organic-inorganic perovskite material layer: methylammonium lead triiodide $CH_3NH_3PbI_3$ (MAPbI3). In this context, three samples of organic perovskite $CH_3NH_3PbI_3$ material have been deposited under various processing temperatures; 70 °C, 80 °C and 100 °C at ambient conditions in order to deduce the effect of temperature on different optical and morphological parameters, namely, layer form, band gap and Auerbach energy. Therefore, each sample's band gap was calculated using optical transmittance spectrum (UV-visible spectrophotometer Shimadzu, Model 1800) and each optical characteristic was made through scanning electron microscope (SEM:FEI Quanta 250 with a tungsten filament). It is found that Auerbach energy decreases with increasing processing temperature giving the lower value of 0.66 eV at 100 °C, which also corresponds to the better band gap value of 1.49 eV. In addition, it is to note that from SEM analysis, the layer morphology of perovskite $CH_3NH_3PbI_3$ is better with high processing temperature value in comparison with lower ones. The obtained results are encouraging for use the 100 °C processed perovskite $CH_3NH_3PbI_3$ for new generation high efficiency solar cell applications.

Keywords: $CH_3NH_3PbI_3$, Spray pyrolysis, Band gap, Auerbach energy, Processing temperature.

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1. INTRODUCTION

Solar energy conversion into electricity using low processing fees and low-cost materials [1, 2] is an important field of research in last years. Perovskite materials are abundant in the nature and have electrical and optical characteristics that attract researcher's attention to investigate its application in solar cell technology. Perovskite-based solar cells are subject of experimental and simulation studies [3-5] to enhance electrical, optical and morphological parameters in order to maximize the power conversion efficiency. Therefore, preparing an organic-inorganic $CH_3NH_3PbI_3$ perovskite-based solar cell has a lot of methods from which we will study the spray pyrolysis one that uses low processing temperatures (less than 150 °C) and hence economical material processing.

Spray pyrolysis method [6] is a low cost processing method because it uses low temperature. In this work, a deposition of a $CH_3NH_3PbI_3$ perovskite layer using spray pyrolysis method has been investigated. Therefore, different processing temperatures have been used in layer spray pyrolysis phase in order to find out their effects on optical parameters and morphological characteristics of the resulting $CH_3NH_3PbI_3$ organic/inorganic perovskite material.

2. EXPERIMENTAL DETAILS

Fig. 1 shows the flow diagram of material processing steps, where the first step focuses on preparing perovskite $CH_3NH_3PbI_3$ material. This can be done by dissolving two substrates: lead iodide (PbI_2) and methylammonium iodide (CH_3NH_3I) in dimethylformamide (DMF). The second step consists in agitating the hole solution for 3 h at constant temperature of 70 °C to obtain a homogenous liquid. Next step, a spray pyrolysis processing is applied using 70 °C, 80 °C and 100 °C on obtained solution. Finally, the resulting layer is air heated at a temperature value of 100 °C for 10 min.

3. RESULTS AND DISCUSSION

Fig. 2, Fig. 3 and Fig. 4 present the morphology of layers deposited at various temperatures 70 °C, 80 °C and 100 °C, respectively. From the figures, it is clear that more compact morphology is obtained at 100 °C processing temperature which means that there is a large area for electron mobility. Therefore, the result gives a homogeneous structure that facilitates electron transport between the ETM and HTM layers. However, the film, which has been prepared at 70 °C, has many holes and cracks, which could cause a sharp leak. The film that is set at 80 °C contains a less number of holes but it has a few clusters.

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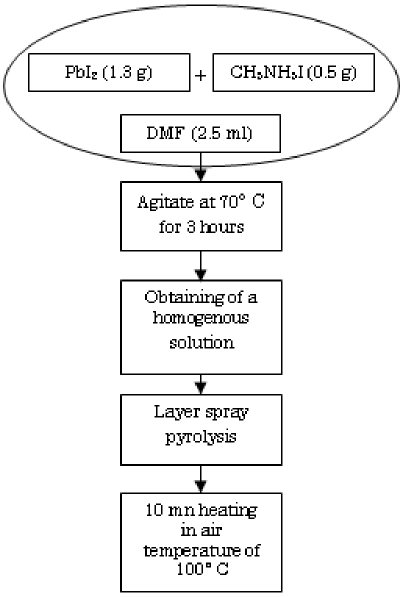


Fig. 1 – Material processing steps

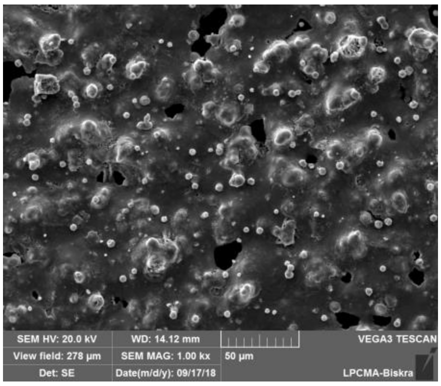


Fig. 2 – SEM image obtained under a processing temperature of 70 °C

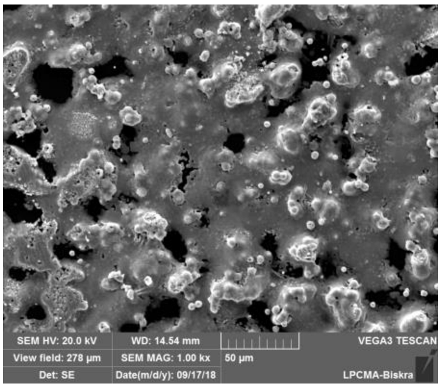


Fig. 3 – SEM image obtained under a processing temperature of 80 °C

Fig. 5 plots transmittance versus wavelength of perovskite layers deposited at various processing temperatures. From the figure, the $CH_3NH_3PbI_3$ thin film shows high absorbance for the wavelength range of 700-800 nm at different temperatures.

According to the absorption spectrometry, it is noticed that the layer deposited at high temperature (100 °C) has an optical absorbance greater than layers deposited at lower temperatures (70 °C and 80 °C).

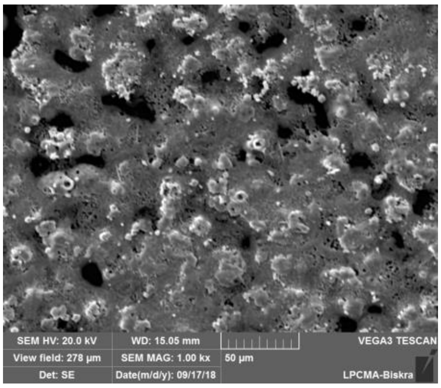


Fig. 4 – SEM image obtained under a processing temperature of 100 °C

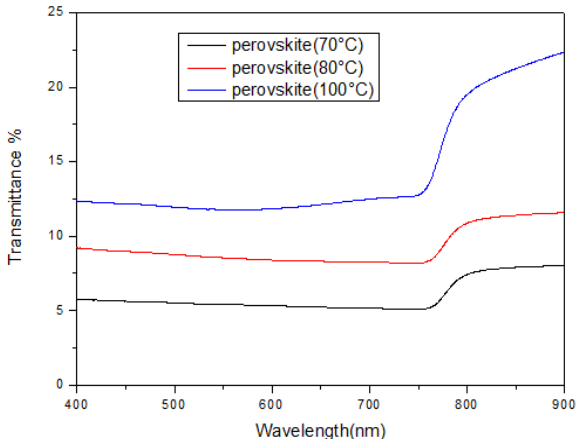


Fig. 5 – Variation of transmittance as a function of wavelength of perovskite layers deposited at different processing temperatures

This characteristic affects the layer band gap.

The absorption coefficient α is given as:

$$\alpha = \frac{1}{d}\ln\frac{1}{T}, \qquad (1)$$

where d is the layer thickness of thin films and T is its transmittance. The film thickness d is obtained from the following equation:

$$d = \frac{M}{\rho a}, \qquad (2)$$

where M is the mass of the thin film, $\rho = 4.15$ g/cm^2 is $CH_3NH_3PbI_3$ material density and a represents the surface area. The mass M is obtained using the following formula:

$$M = M_f - M_i, \qquad (3)$$

where M_i and M_f are the mass of the substrate before and after $CH_3NH_3PbI_3$ deposition, respectively.

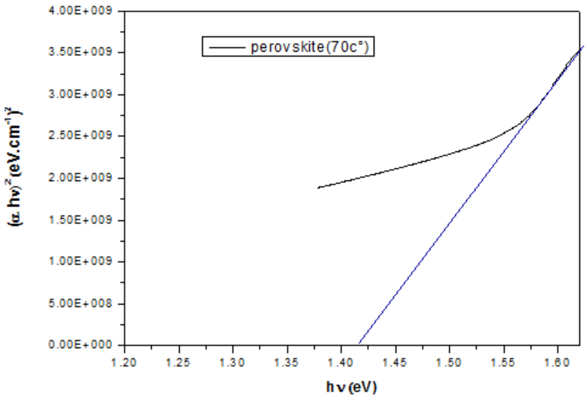
The band gap of the realized $CH_3NH_3PbI_3$ perovskite layer is written as:

$$\alpha h\nu = B\left(h\nu - E_g\right)^{\frac{1}{2}}, \qquad (4)$$

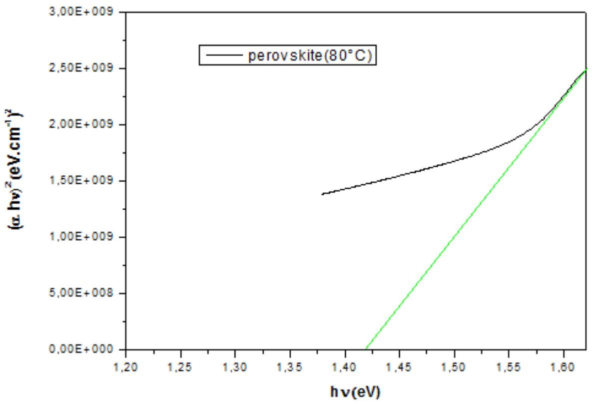
Fig. 6 plots $(\alpha h\nu)^2$ as a function of $h\nu$ for different processing temperatures of perovskite material. From the figure, the band gap can be obtained from the intersection between the straight line with the abscise line. Therefore, the deduced direct band gap can be found between 1.41 and 1.49 eV. Thus, the obtained results are summarized in Table 1.

Table 1 – Band gap and Auerbach energy values for different processing temperatures

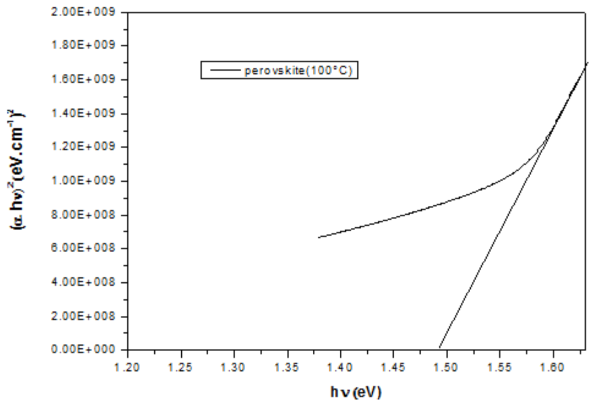
T, °C	70	80	100
E_g, eV	1.41	1.42	1.49
E_u, eV	0.78	0.74	0.66



a



b



c

Fig. 6 – Variation of $(\alpha h\nu)^2$ as a function of $h\nu$ for different processing temperatures a) 70 °C, b) 80 °C and c) 100 °C

It is clear that the band gap value increases with increasing processing temperature.

Auerbach energy E_u is a principal optical characteristic of a thin film layer; it is calculated using the following expression:

$$\ln(\alpha) = \ln(\alpha_0) + \frac{h\nu}{E_u}. \quad (5)$$

Fig. 7 shows the variation of ln(α) against $h\nu$ for different processing temperatures of perovskite material. From the figure, we can obtain the reverse Auerbach energy using the value found from the intersection between the straight line with the abscise line. Therefore, it is noticeable that Auerbach energy decreases with increasing processing temperature which means that high temperature processing gives a crystalline structure with fewer defects.

Table 1 also summarizes the Auerbach energy values obtained for different processing temperatures of perovskite material.

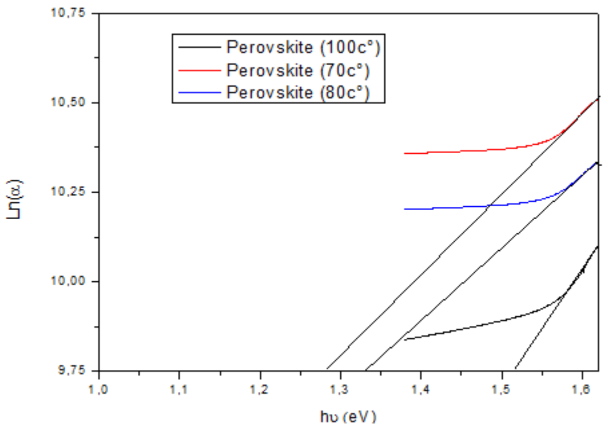


Fig. 7 – Variation of *ln(α)* as a function of *hυ* for different processing temperatures of perovskite material

4. CONCLUSIONS

In this work, the effect of processing temperatures on different optical and morphological parameters using spray pyrolysis deposition of $CH_3NH_3PbI_3$ perovskite has been presented. It is found that morphology structure, band gap and Auerbach energy are better with processing temperature of 100 °C in comparison with lower ones. Therefore, the obtained results provide guidance for use the 100 °C processed perovskite $CH_3NH_3PbI_3$ for new generation high efficiency solar cell applications.

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Вплив температури обробки на оптичні та морфологічні параметри органічного перовскіту $CH_3NH_3PbI_3$, отриманого методом спрей-піролізу

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Поновлювані джерела енергії фактично знаходяться в експоненційній еволюції, щоб з'ясувати природну, дешеву, захищену та безпечну альтернативу для енергетичних ресурсів палива. Останнім часом досліджено різні способи осадження шарів перовскітних матеріалів у фотоелектричних застосуваннях з метою розуміння впливу зовнішніх параметрів на характеристики сонячних елементів, такі як щільність струму короткого замикання (J_{sc}), напруга розімкнутого ланцюга (V_{oc}), коефіцієнт заповнення (*FF*) і ефективність перетворення енергії (*PCE*). У даній роботі метод осадження спрей-піролізом з рухомим соплом був використаний для виготовлення шару матеріалу органічно-неорганічного перовскіту. У цьому контексті три зразка матеріалу органічного перовскіту $CH_3NH_3PbI_3$ були осаджені при різних температурах обробки 70 °C, 80 °C та 100 °C при умовах навколишнього середовища, щоб з'ясувати вплив температури на різні оптичні та морфологічні параметри, а саме: форму шару, ширину забороненої зони та енергію Ауербаха. Тому ширина забороненої зони кожного зразка розраховувалася за допомогою спектру оптичного пропускання (УФ-видимий спектрофотометр Shimadzu, Модель 1800), і кожна оптична характеристика отримувалася скануючим електронним мікроскопом (SEM:FEI Quanta 250 з вольфрамовою ниткою). Було встановлено, що енергія Ауербаха зменшується зі збільшенням температури обробки, даючи мінімальне значення 0.66 еВ при 100 °C, що також відповідає кращому значенню ширини забороненої зони 1.49 еВ. Крім того, слід зазначити, що з аналізу SEM морфологія шару перовскіту $CH_3NH_3PbI_3$ краща при високому значенні температури обробки у порівнянні з нижчими температурами. Отримані результати заохочують використання перовскіту $CH_3NH_3PbI_3$ обробленого за температури 100 °C для високоефективних сонячних елементів нового покоління.

Ключові слова: $CH_3NH_3PbI_3$, Спрей-піроліз, Заборонена зона, Енергія Ауербаха, Температура обробки.