

Organometallic Perovskite Solar Cell

Mohammed Lakhdar Ayachi^{1*}, Atman Benhaoua¹, Ali Tliba², Belgacem Souyei³

¹Department of Physics, Faculty of Exact Science, University of El Oued, ALGERIA

²Lab. VTRS, Faculty of Exact Science, University of El Oued, ALGERIA

³Department of Chemistry, Faculty of Exact Science, University of El Oued, ALGERIA

Email*: ayachi-mohammedlakhdar@univ-eloued.dz

Abstract –Halide organometallic perovskite has an important role in the efficiency increase of the solar cell. Thus in this work, we formed the basic nucleus of the organic perovskite, and we studied its morphological properties. The X-ray diffraction result shows that this compound is consistent, homogeneous, and has preferential orientation growth be at (100) plane, which means that the experimental conditions which we worked on were optimal. After adding both tin iodide and methylamine chloride in organic solvents (DMF and DMSO). Deposited this mixture by spray pyrolysis method at Specific temperature 120C°, on the glass substrate, a thin black layer formed; the result of X-ray diffraction on this latter layer showed that it is a spectrum of perovskite compound, which has preferential orientation growth be at (110) plane. Via optical proprieties, it found that has low gap energy of 1.78 eV, and transmittance of 1,6% furthermore it has a high absorption coefficient of 8.10^4 cm^{-1} , in the visible domain. But it has a relatively high value of Auerbach energy 0.6 eV due to the crystal defects. So this compound could be an active layer in the solar cell.

Keywords: Auerbach energy, Optical and morphological properties, X-ray diffraction, Crystal defects.

Received: 05/10/2021 – Accepted: 20/12/2021

I. Introduction

Hybrid organic perovskite has the same structure as mineral perovskite (ABX₃) [1]. But A is organic cations such as methylammonium, formamidinium, or rare-earth elements as Cs. B is ions of heavy metal as (Pb⁺⁴, Sn⁺⁴) [2,3]. As for the X can be (Cl-Br-I-F) [4]. Hybrid organic perovskite has a wide range of applications in electrical engineering such as gas sensors and light-emitting diode (LEDs) [5], but the most important applications are in solar cells [6]. The solar energy conversion efficiency of perovskite solar cells is over high 25%. The preparation methods of these compounds in the laboratories have a major impact on the energy conversion efficiency of these cells [7]. The major obstacle facing relying on these compounds to convert solar energy is instability [8], this due to several factors including humidity [9] and

asymmetry between fundamental compounds. Halide perovskite has several kinds, among them we find (CH₃NH₃PbI₃) which has black color and many applications but contains lead which is hazardous on human and deteriorate rapidly [10, 11]. And there is another kind has formula (CH₃NH₃PbCl₃) also contains lead with white color but doesn't have greater applications because of the big gap of energy 3.1 eV [12], this latter type has good propriety that it doesn't deteriorate quickly because the basic nucleus is methylammonium chloride (CH₃NH₃Cl). another type of compound less hazardous on humans and has high efficiency conversion of solar energy, is (CH₃NH₃SnI₃) has black color with small gap energy of 1.3 eV but it deteriorates very quickly [13]. There are great researches efforts about the instability of active compounds, without forgetting the simulation studies by several software specialized in the field [14, 15].

The novelty of our work is combining between two properties structure stability and high efficiency conversion of energy, by preparing ($CH_3NH_3SnI_2Cl$) and depositing on glass layer by using spray pyrolysis technique where we prepared methylammonium chloride (CH_3NH_3Cl) in our lab and we studied the morphological structure and add tin iodide SnI_2 to methylammonium chloride and we study some optical and structural parameters of a new compound.

II. Method and experience

In the first step, CH_3NH_3Cl was synthesized by mixing 30 ml of methylamine solution (40% in methanol) and 32.3 ml of hydrochloric acid solution (57% by weight in water) in a flask at $0^\circ C$ in an ice medium for 2 hours with constant stirring. Then put the solution overnight at $60^\circ C$, take the solution, and put in the rotavaporator we got white precipitate. Finally, we wash with diethyl ether two or three times and then let this white precipitate at $70^\circ C$ until dry.

In the second step, we take 0.5g of CH_3NH_3Cl and 1.2 g of SnI_2 were mixed in 15ml DMSO because methylammonium chloride does not easily solute in DMF, then put the mixture at $60^\circ C$ overnight with constant stirring. Afterward, the small beaker was sealed and kept dark at room temperature to avoid reaction with sunlight. After this work we deposit the second compound over the glass layer by using the spray pyrolysis technique at $120^\circ C$, we note a brownish-red precipitate on the substrate, afterward heat treating for this second compound at $180^\circ C$, until the brownish-red color turns to black. Finally, we get organic perovskite thin film as shown in Figure 1 [16, 17].

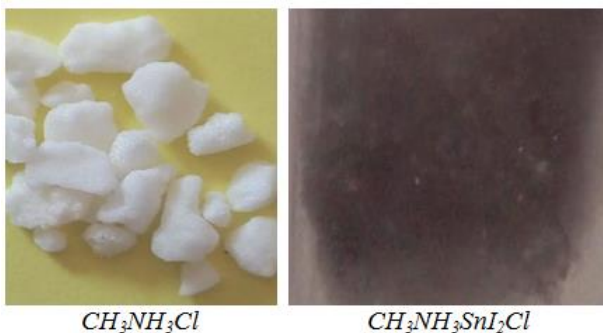


Figure 1. Organic perovskite thin film

III. Result and discussion

III.1 Structural proprieties

Figure 2 Shows the X-ray diffraction spectra of methylaminechloride CH_3NH_3Cl use PROTO AXRD device estimated wavelength $K\alpha$ of Cu ($0.15405nm$) scans angular interval between $10 - 80$ degree. The peaks which are appeared in CH_3NH_3Cl at 2θ : 17.75° , 27.4° , 29.7° , 35.15° , 42.05° , 56.3° , 58.29° , compatible to crystal levels of (100) , (111) , (310) , (210) , (224) , (320) , (321) . Based on previous studies prove the formation of first compound (CH_3NH_3Cl) has tetragonal form. So, the obtained curve demonstrates that the compound has polycrystalline structure in form tetragonal the sharp peak correspond with plan (100) indicates to best crystallization means that preferential orientation be at (100) plan compared with other plans.

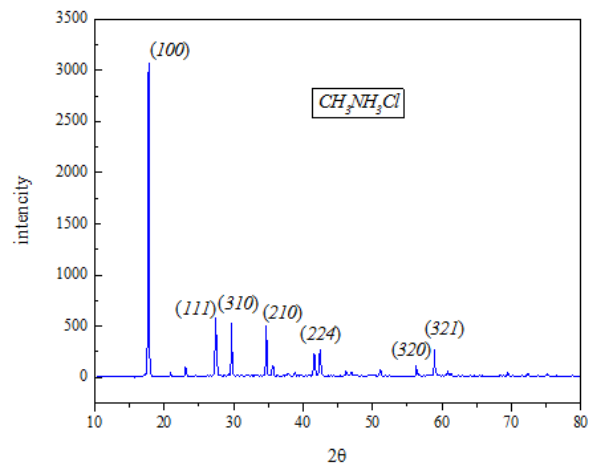


Figure 2. X-ray Diffraction spectrum of CH_3NH_3Cl powder

The peaks which are appeared in the second compound as shown in Figure 3 ($CH_3NH_3SnI_2Cl$) at 2θ : 14.01 , 21.964 , 31.4 , 34.61 , 41.3 , compatible with crystal levels (110) , (014) , (112) , (200) , (224) . Based on previous studies prove the formation of the second compound ($CH_3NH_3SnI_2Cl$) has a tetragonal form. Thus, the obtained curve demonstrates that the compound has a polycrystalline structure in tetragonal form, the sharp peak corresponding with a plan (110) indicates to best crystallization means that compared with other plans. The assigned (110) and (220) peaks confirm the perovskite phase. Diffraction peaks have a good corresponding with the previous articles. Both perovskite and methylamine chloride powder have tetragonal forms. For selecting inter-reticular distance d_{hkl} and grid constants a and c , we used the following relations [18, 19].

Table 1 summarizes some parameters among them d_{hkl} , 2θ , grid parameters, and Miller indexes.

$$2d_{hkl} \sin(\theta) = n\lambda$$

$$\frac{1}{d_{hkl}^2} = \frac{1}{a^2}(h^2 + k^2) + \frac{l^2}{c^2}$$
(1)

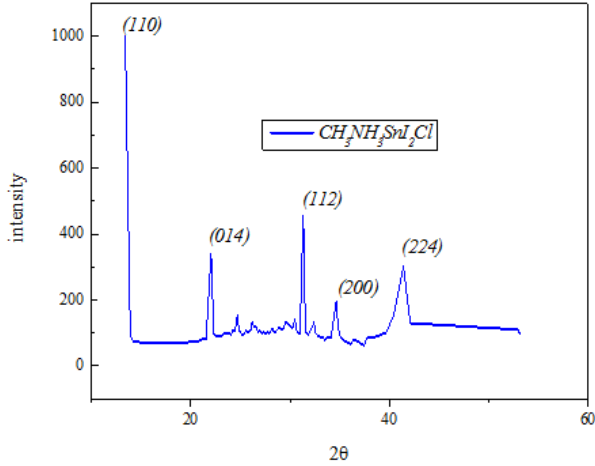


Figure 3. X-ray Diffraction spectrum of $CH_3NH_3SnI_2Cl$ thin films

Table 1. $CH_3NH_3SnI_2Cl$ and CH_3NH_3Cl parameters: d_{hkl} , grid parameters, Miller indexes

Compound	(hkl)	2θ	$d_{hkl}(\text{Å})$	$a(\text{Å}) = b(\text{Å})$	$c(\text{Å})$
CH_3NH_3Cl	100	17,75	4,995	9,9901	12,8972
	111	27,4	3,2538	6,5076	8,4013
	310	29,7	3,00688	6,0137	7,7637
	210	35,15	2,5521	5,1042	6,5895
	224	42,05	2,1479	4,2958	5,5459
	321	58,29	1,5822	3,1645	4,0854
$CH_3NH_3SnI_2Cl$	110	14,01	6,31899	12,6379	16,3155
	014	21,964	4,0453	8,0906	10,4449
	112	31,4	2,8478	5,6995	7,73531
	200	34,61	2,5907	5,1814	6,6891
	224	41,3	2,1851	4,3703	5,6421

III.2. Optical proprieties

Figure 4 shows the variation of the transmittance versus of the wavelength of $CH_3NH_3SnI_2Cl$ film. It is shown that the transmittance spectrum of 300-900 nm, Splitted to two intervals : the first interval between 600 and 900 nm, the transmittance of deposited film is in the order of 1,6%, low permeability value is caused by the black color of compound, thickness of layer and some crystal defects. Second interval between 300 and 600 nm, the transmittance of deposited film is in the order of 0.05% is very low permeability value is caused by electronic transition between valence band to conduction band so

we used Tauc's relation in determining direct gap energy[20, 21] :

$$(\alpha h\nu)^2 = A(h\nu - E_g)$$
(2)

Where:

A is proportional constant, E_g gap energy, α is absorption coefficient, h is Planck constant and ν is wave frequency.

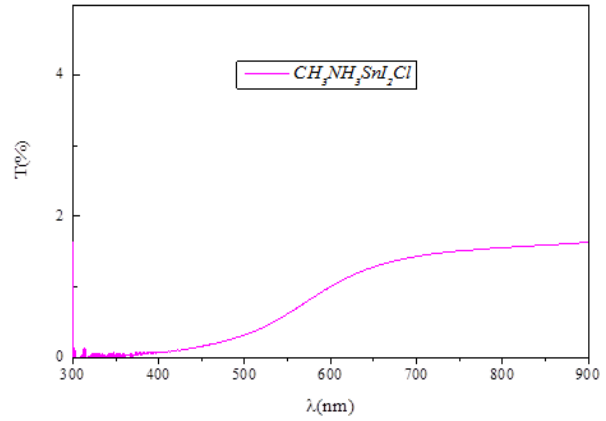


Figure 4. Transmittance versus wavelength

Figure 5 shows the variation of Tauc quantity of organometallic perovskite, where noticed slow variation at weak energy, after them, we noticed sharp increasing in this quantity, so the intercept between tangent line and energy axis giving a value of gap energy.

Auerbach energy E_u is a very important optical quantity in thin films, so we used the following relation for its determination [22].

$$\ln(\alpha) = \ln(\alpha_0) + \frac{h\nu}{E_u}$$
(3)

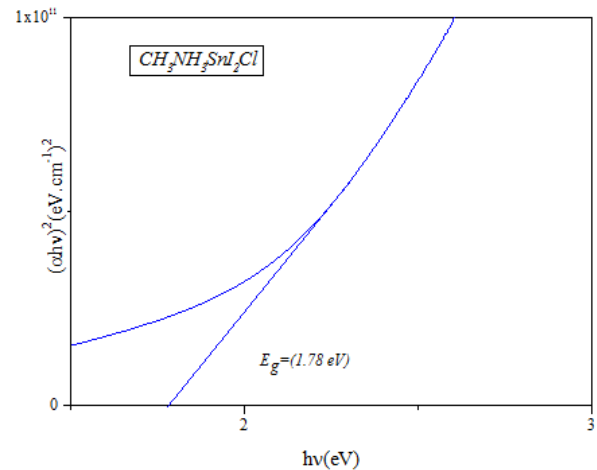


Figure 5. Curve shows the variation of $(\alpha h\nu)^2$ versus photon energy

Through the spectrum drawing of the variation of $\ln(\alpha)$ versus $h\nu$ as shown in Figure 6, we can find the Auerbach energy using the inverted value of the slop

tangent line with the curve. Auerbach energy value is relatively great due to crystal defects.

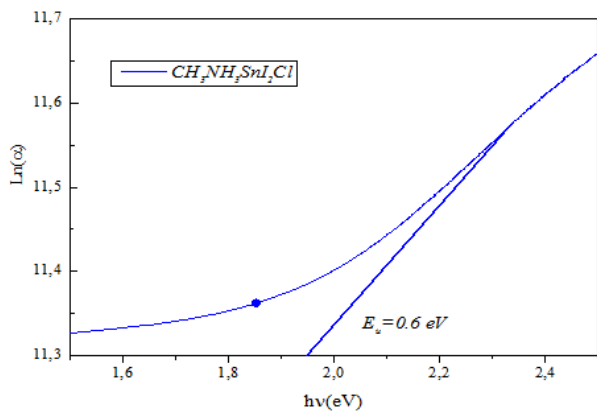


Figure 6. Curve shows the variation of $\ln(\alpha)$ versus photon energy

Figure 7 shows the variation of the absorption coefficient. Where found high value in visible and near infrared ranges within 10^5 cm^{-1} due to black color of films, but in near ultraviolet range noticed increasing in absorption coefficient due to principle transitions between valance band to the conduction band.

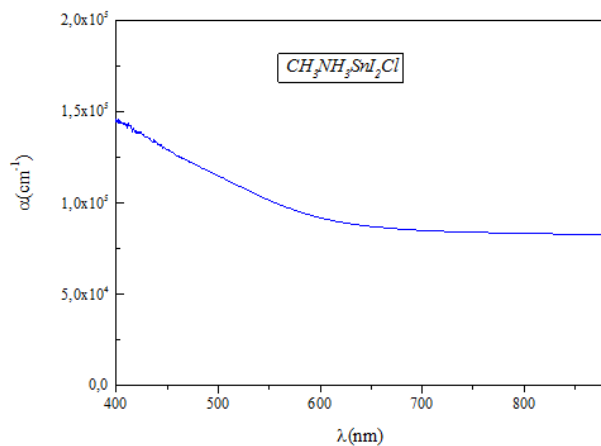


Figure 7. Curve presented a variation of absorption coefficient (α) versus wavelength

IV. Conclusion

Through this work, methylamine chloride powder is obtained, which is considered the basic nucleus of organic perovskite where it has the best crystallization and preferential orientation be at (100) level as shown via XRD spectrum. Meanwhile used this compound for producing perovskite thin film by the spray pyrolysis with moving nozzle deposition device at determining temperature (120C°). XRD spectrum shows the best

crystallization and preferential orientation at the (100) level. This film has a low bandgap (1.78 eV) and black color, furthermore has high Auerbuch energy (0.6 eV), and very high absorption coefficient in the visible range that's mean can be an active layer in solar cells.

References

- [1] [A. K. Abass, "Synthesis of CdTiO₃ thin films and study the impact of annealing temperature on their optical, morphological and structural properties," Eurasian J. Anal. Chem., vol. 13, no. 5, 2018.
- [2] Y. Takahashi, H. Hasegawa, Y. Takahashi, and T. Inabe, "Hall mobility in tin iodide perovskite CH₃NH₃SnI₃: Evidence for a doped semiconductor," J. Solid State Chem., vol. 205, 2013, pp. 39–43.
- [3] X. Li, L. Li, Z. Ma, J. Huang, and F. Ren, "Low-cost synthesis, fluorescent properties, growth mechanism and structure of CH₃NH₃PbI₃ with millimeter grains," Optik (Stuttg.), vol. 142, 2017, pp. 293–300.
- [4] S. A. Moyez and S. Roy, "Thermal engineering of lead-free nanostructured CH₃NH₃SnCl₃ perovskite material for thin-film solar cell," J. Nanoparticle Res., vol. 20, no. 1, 2018,
- [5] Chengxi Zhang, Lyudmila Turyanska, Haicheng Cao, Lixia Zhao, Michael W. Fay, Robert Temperton, James O'Shea, Neil R. Thomas, Kaiyou Wang, Weiling Luan, Amalia Patanè, "Hybrid light emitting diodes based on stable, high brightness all-inorganic CsPbI₃ perovskite nanocrystals and InGaN," Nanoscale, vol. 11, no. 28, 2019, pp. 13450–13457.
- [6] D. Li, J. Shi, Y. Xu, Y. Luo, H. Wu, and Q. Meng, "Inorganic-organic halide perovskites for new photovoltaic technology," Natl. Sci. Rev., vol. 5, no. 4, 2018, pp. 559–576.
- [7] Ganesh Alwarappan and Md Raiyan Alam and Walid M. I. Hassan and Mohamed F. Shibl and Sherin Alfalah and Sunil Patil and Reza Nekovei and Amit Verma, "Role of organic cation in modern lead-based perovskites," Sol. Energy, vol. 189, 2019, pp. 86–93.
- [8] C. L. Chen et al., "Improved open-circuit voltage and ambient stability of CsPbI₂Br perovskite solar cells by incorporating CH₃NH₃Cl," Rare Met., vol. 39, no. 2, 2020, pp. 131–138.
- [9] X. Dong et al., "Improvement of the humidity stability of organic-inorganic perovskite solar cells using ultrathin Al₂O₃ layers prepared by atomic layer deposition," J. Mater. Chem. A, vol. 3, no. 10, 2015, pp. 5360–5367.
- [10] J. Bisquert and E. J. Juarez-Perez, "The causes of degradation of perovskite solar cells." ACS Publications, 2019.
- [11] Su-Yong Bae, Su Young Lee, Ji-wan Kim, Ha Nee Umh, Jaeseong Jeong, Seongjun Bae, Jongheop Yi, Younghun Kim & Jinhee Choi, "Hazard potential of

- perovskite solar cell technology for potential implementation of ‘safe-by-design’ approach,” *Sci. Rep.*, vol. 9, no. 1, 2019, pp. 1–9.
- [12] F. Xu, T. Zhang, G. Li, and Y. Zhao, “Synergetic Effect of Chloride Doping and CH₃NH₃PbCl₃ on CH₃NH₃PbI₃-xCl_x Perovskite-Based Solar Cells,” *ChemSusChem*, vol. 10, no. 11, 2017, pp. 2365–2369.
- [13] H. Yao, F. Zhou, Z. Li, Z. Ci, L. Ding, and Z. Jin, “Strategies for Improving the Stability of Tin-Based Perovskite (ASnX₃) Solar Cells,” *Adv. Sci.*, vol. 7, no. 2020, pp.10.
- [14] Hima Abdelkader, Ahmed Khalil Le Khouimes, Abdallah Rezzoug, Mouslem Ben Yahkem, Abderrahmane Khechekhouche, Imad Kemerchou. Simulation and optimization of CH₃NH₃PbI₃ based inverted planar heterojunction solar cell using SCAPS software, *International Journal of Energetica*, Vol 4, n°1, 2019. pp. 56-59.
- [15] Abdelkader Hima, Abderrahmane Khechekhouche, Imad Kemerchou, “Enhancing of CH₃NH₃SnI₃ based solar cell efficiency by ETL engineering”, *International Journal of Energetica*, vol 5, no 1, 2020.
- [16] K. Deng, Z. Liu, Y. Xin, and L. Li, “PbI₂/CH₃NH₃Cl Mixed Precursor-Induced Micrometer-Scale Grain Perovskite Film and Room-Temperature Film Encapsulation toward High Efficiency and Stability of Planar Perovskite Solar Cells,” *Adv. Mater. Interfaces*, vol. 5, no. 15, 2018, pp. 1–8.
- [17] I. Kemerchou, F. Rogti, B. Benhaoua, N. Lakhdar, A. Hima, O. Benhaoua, A. Khechekhouche, “Processing temperature effect on optical and morphological parameters of organic perovskite CH₃NH₃PbI₃ prepared using spray pyrolysis method,” *J. nano-and Electron. Phys.*, no. 11, 3, 2019, pp. 3011.
- [18] I. Kemerchou, A. Khechekhouche, A. Timoumi, F. Rogti, A. Hima, A. Sadoun, A. Tliba, M. S. Aida, “Study of the chemical structure of CH₃NH₃PbI₃ perovskite films deposited on different substrates,” *J. Mater. Sci. Mater. Electron.*, vol. 32, no. 3, 2021, pp. 3303–3312.
- [19] M. Mahmoudi, A. Benhaoua, B. Benhaoua, L. Segueni, R. Gheriani, and A. Rahal, “Study of structural, optical and electrical properties of fluorine-cobalt co-doped tin dioxide SnO₂,” *Dig. J. Nanomater. BIOSTRUCTURES*, vol. 14, no. 4, 2019, pp. 1079–1086.
- [20] A. S. Hassanien and A. A. Akl, “Optical characteristics of iron oxide thin films prepared by spray pyrolysis technique at different substrate temperatures,” *Appl. Phys. A*, vol. 124, no. 11, 2018, pp. 1–16.
- [21] I. M. El Radaf, T. A. Hameed, G. M. El komy, and T. M. Dahy, “Synthesis, structural, linear and nonlinear optical properties of chromium doped SnO₂ thin films,” *Ceram. Int.*, vol. 45, no. 3, 2019, pp. 3072–3080.
- [22] F. N. C. Anyaegbunam and C. Augustine, “Study of optical band gap and associated Urbach energy tail of chemically deposited metal oxides binary thin films,” *Dig. J. Nanomater. Biostructures*, vol. 13, 2018, pp. 847–856.