Fabrication and Characterization of Organolead Halide Perovskite Solar Cell  

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ABSTRACT  
This research presents the description of the procedure for the manufacturing the Organolead halide Perovskite Solar Cells (OPSC). We described the synthesis organo-lead halide perovskite materials, MAPbIBr₂ and MAPbBrCl₃, which employed as absorption layer in OPSCs. Optical properties for the perovskite materials which used to prepare OPSCs is tested by the measurements the absorbance spectrum in order to calculate the energy gap. X-ray diffraction, AFM microscope and SEM microscope are used in our work to study the structure of the samples. Finally, the tests OPSCs by Light I-V Measurement System are implemented in order to obtained the parameters of the manufacturing devices. The Power Conversion Efficiencies (PCE) of OPSCs are (0.12%) and (0.07%) to the devices have MAPbBr₂ and MAPbBrCl₃ as sensitized absorption layer, respectively. Measurements are tested at AM1.5 global sunlight (100 mW cm⁻²).

Introduction  
Global energy consumption has been continually increasing with population growth and fast-paced industrial development in recent decades, which demands renewable energy sources in view of long-term sustainable development. Generating cost-effective and environmentally benign renewable energy remains a major challenge for both technological and scientific development. Solar cells based on the photovoltaic with the advantages of decentralization and sustainability have attracted great attention in the past 50 years. Currently, the photovoltaics markets is dominated by crystalline silicon-based solar cells with a share of 89%; however, a cost-effective and high-throughput material named perovskite has proven to be capable of Power Conversion Efficiency (PCE) 20%[1], compared to 3.8%[2] of PCE that was obtained only four years ago. Organic-inorganic halide perovskite solar cells have been the most significant development in the field of photovoltaic’s in the present decade and are the best bet at satisfying the need for high efficiencies while allowing for low cost solution based manufacturing. Low cost, stability and high efficiency are research reason in the development of organic-inorganic perovskite solar cells. Organolead halide perovskite solar cells has attracted researchers attention as a light harvester for perovskite solar cells because of its tenable band gap, large absorption coefficient, high charge carrier mobility, and long electron-hole diffusion [3,4].

Basic Theory  
Perovskites materials are described by the formula ABX₃, where X is an anion and A and B are cations of different sizes (A being larger than B). The “A” cation is divalent and the “B” cation is tetravalent. In the cubic unit cell, the A-cation resides at the eight corners of the cube, while the B-cation is located at the body center that is surrounded by 6 X-anions (located at the face centers) in an octahedral [BX₆]₄⁻ cluster [5]. The crystal structure of perovskites is depicted in Fig.1[6].

![Fig 1. Inset depicts schematic to the perovskite structure.](image)

The PCE of solar cell device is estimated by the equation (1)[6-8]:

\[
PCE = \frac{FF \times V_{oc} \times I_{sc}}{P_{in} \times A_{ac}}
\]

Where FF, Vₜ, Iₙ, Pₖ and Aₙ are full factor, open voltage circuit, short current circuit, input power to the solar cell estimated to one solar illumination which equate to (100 mW/cm²) and active area of solar cell, respectively.

The optical band gap of the thin film have been investigated for the allowed direct transition from the equation (2)[6,9]:

\[
\alpha \nu = B \left( \nu - E_g \right)^{1/2}
\]
Where, $h$ is the Planck constant, $(\alpha)$ is the absorption coefficient, $\nu$ is the light frequency, $E_g$ is the optical energy gap and $B$ is empirical constant.

**Experimental**

Composition and crystal structure studied by X-Ray Diffraction (XRD Shimadzu 6000, Cu-Kα) for USP coating. The surface morphology of sample is studied by investigation of Atomic Force Microscopy (AFM) (AA3000 Scanning Probe microscope, Angstrom Advanced Inc.). The difference in the shape of the perovskite crystals studied by images of Scanning Electron Microscope (SEM) (Bruker Nano GmbH, Germany), analysis is performed by using a magnification of 1kx and 5kx, high voltage 5kV and SEM in secondary electron mode.

Light I-V Measurement Test Reports are recorded by Photovoltaic measurements system, composed of Oriel I–V test station using an Oriel Solar simulator. The solar simulator is class AAA for spectral performance, uniformity of irradiance, and temporal stability. The solar simulator is equipped with a450 W xenon lamp. The output power is adjusted to match AM1.5 global sunlight (100 mW cm$^{-2}$).

I–V curves are obtained by applying an external bias to the cell and measuring the generated photocurrent with a Keithley model 2400 digital source meter. The transmittance of coated films is measured in the wavelength range of (400 - 800) nm using a (SPECTRO UV/Vis Double Beam (UVD-3500) Labomed Inc). A blank sample of substrate is used as a reference in the measurement of optical transmittance.

Synthesis of Organic Perovskite Materials (OPM) reported in Ref [6,8]. Methylamine Iodide (CH$_3$NH$_3$I) is prepared by reacting Methylamine, 33 wt % in Ethanol (BDH-LTD), with Hydro-Iodic acid (HI) 57 wt % in water (BDH-LTD) under ice bath stirring for 2 h. Typical quantities employed are 24 ml of Methylamine, 10 ml of HI, and 100 mL of Ethanol. Upon drying at 100 °C, a white powder is formed, which is placed overnight in a vacuum oven before use. Methylamine Bromide is prepared at the same method and previous quantities. To obtain the perovskite solution precursor of MAPbBr$_3$, we dissolved both the CH$_3$NH$_3$I and the PbBr$_2$ (BDH-LTD), in anhydrous N, N-Di Methyl Formamidine (DMF) (Sigma Aldrich) at a 3:1 molar ratio. To obtain the perovskite solution precursor of MAPbCl$_3$, we dissolved both the CH$_3$NH$_3$Br and the PbCl$_2$ (BDH-LTD) in anhydrous N, N-Di Methyl Formamidine (DMF) (Sigma Aldrich) at a 3:1 molar ratio, with final concentrations of 40 wt %. 10 ml of solutions preparation added to 0.15 g of Nano particle> 30 nm of Al$_2$O$_3$ powder (China of origin) to obtain scaffold perovskite precursor solution of CH$_3$NH$_3$I$\cdot$PbBr$_2$ and scaffold perovskite precursor solution of CH$_3$NH$_3$Br$\cdot$PbCl$_3$.

TiO$_2$ compact layer preparation is a first fabricated applying Aerosol Assisted Chemical Vapor Deposition (AACVD) technique [6,8,9] by using Ultrasonic Atomizer (402AI) with ultrasonic frequency (1.5MHz). The precursor solution, which preparation of Nano particle> 20 nm of TiO$_2$ powder (China of origin) dispersed in ethanol solvent, is sprayed on per-heated Transparent Conductive Oxide (TCO) glass substrates of Fluorine-doped Tin Oxide (FTO) (Coated Sodalime float glass of Visiontek, sheet Resistance 8 Ω/□) at 450°C. Deposited time is 1h and the substrates are left to cold at room temperature, followed by, depositing scaffold perovskite precursor solution of CH$_3$NH$_3$I$\cdot$PbBr$_2$ or CH$_3$NH$_3$Br$\cdot$PbCl$_3$, by spin coated at speed 2000 rpm and annealing at 150°C to obtain 2 samples of (FTO/Compact TiO)$_2$/Scaffold CH$_3$NH$_3$I$\cdot$PbBr$_2$ or CH$_3$NH$_3$Br$\cdot$PbCl$_3$, as shown in figure 2. The temperature of the (FTO/Compact TiO$_2$) coated substrates is monitored by an infrared temperature indicator through the all experimental runs.

![Fig (2)](image-url)
Fig. 3. (a) XRD patterns of $\text{CH}_3\text{NH}_3\text{IPbBr}_2$ layer on sheet glass substrate sample comparative with $\text{CH}_3\text{NH}_3\text{PbBr}_3$ Ref [10]. (b) XRD patterns of $\text{CH}_3\text{NH}_3\text{BrPbCl}_2$ layer on sheet glass substrate samples comparative with $\text{CH}_3\text{NH}_3\text{PbBr}_3$ Ref [12].

Fig. 4 (a and b) are illustrated top SEM images of $\text{CH}_3\text{NH}_3\text{IPbBr}_2$ layer. (c and d) are top SEM images of $\text{CH}_3\text{NH}_3\text{PbBrCl}_2$ layer. Scale bars of the (a and c) images are 10$\mu$m and the (b and d) images are 5$\mu$m, scanning with high voltage 5kV and magnification (5 and 10) kx, respectively. Samples are prepared by spin coated at speed 2000 rpm deposition of perovskite precursor solution on preheated sheet glass substrate at 150°C.

Fig. 4(e and f) depicts SEM images of compact layer of TiO$_2$ on FTO electrode deposited by (AACVD) technique on preheated sheet glass substrate at 450°C temperature and deposited at 1h time. The particle size dimensions of TiO$_2$ compact layer are appeared in SEM image as shown in the Fig. 4(e), (X=93.75nm, Y=93.32nm, D=132.3nm).

Fig. 4a & b. Top SEM images of $\text{CH}_3\text{NH}_3\text{IPbBr}_2$ layer. c&d Top SEM images of $\text{CH}_3\text{NH}_3\text{BrPbCl}_2$ layer, scale bars of left perovskite images are 10$\mu$m and right images are 5$\mu$m. Top SEM images of compact layer of TiO$_2$ on FTO electrode, Scale bar of left image is 500nm and right image is 2$\mu$m.

Fig. 5a is explained the absorption of halide perovskite films and appeared more absorption to $\text{CH}_3\text{NH}_3\text{IPbBr}_2$ than $\text{CH}_3\text{NH}_3\text{BrPbCl}_2$ sample that which are measured in the wavelength range (400-800) nm using a (SPECTRO UV/VIS Double Beam (UVD-3500) Labomed, Inc.) and glass substrates as reference samples. Plots inset of $(\alpha h\nu)^2$ versus photon energy $(h\nu)$ for perovskite layers on glass.
substrates are illustrated in Fig. 9b. A direct optical band gap energy (Eg) for halide perovskites materials \( \text{CH}_3\text{NH}_X \text{X}_3 \) (\( X = \text{Cl, Br, I} \)) is reported by Simon et al [11,12]. (Eg) of samples is determined by fitting the absorption data to the direct transition equation (2). The optical band gap value is obtained by extrapolating the linear part of the curve \((\alpha h\nu)^2\) as a function of photon energy, \( h\nu \), intercept the \((h\nu)\) axis at \( \alpha = 0\). The values estimated of Eg are (1.8 and 1.92) eV for \( \text{CH}_3\text{NH}_3\text{IPbBr}_2\) and \( \text{CH}_3\text{NH}_3\text{BrPbCl}_2\) samples, respectively. We observed that the energy gap is decreased and the absorption is increased of the mixed halide, \( X = \text{Br and I} \), perovskite structure of \( \text{CH}_3\text{NH}_3\text{IPbBr}_2\) sample, when insertion Chlorine instead of Iodine in mixed of halide, \( X = \text{Br and Cl} \), of \( \text{CH}_3\text{NH}_3\text{BrPbCl}_2\) sample then the energy gap is increased and the absorption is decreased, as shown in figure 5.

Fig. 5a. depicts absorption of Visible light of the \( \text{CH}_3\text{NH}_3\text{IPbBr}_2\) and \( \text{CH}_3\text{NH}_3\text{BrPbCl}_2\) films. (b) explain energy gap of these films.

I-V curve of OPSC have best PCE to the device employed \( \text{CH}_3\text{NH}_3\text{IPbBr}_2\) as absorption layer depicted in test report as shown in fig.6(a). Measurement is carried out under 1 sun illumination (AM1.5G, 100 mW/cm\(^2\)), active area of solar cell is 0.1 cm\(^2\) and sweeping voltages in the scan-direction with a scan rate of \( s = 50\) mV/s. Fig.6 (b) is the test report which depicted I-V curve of device employed \( \text{CH}_3\text{NH}_3\text{BrPbCl}_2\) as absorption layer. Measurement is tested at the same conditions as the fig.6(a). We observed increased the short current circuit to the device employed mixed halide (\( X = \text{Br and I} \)) \( \text{CH}_3\text{NH}_3\text{IPbBr}_2\) as absorption layer and increased open voltage circuit to the device employed mixed halide (\( X = \text{Br and Cl} \)) \( \text{CH}_3\text{NH}_3\text{BrPbCl}_2\) as absorption layer as shown as in the figure (6).
Conclusions

In summary, the OPSCs which employed the perovskite materials of CH$_3$NH$_3$IPbBr$_2$ or CH$_3$NH$_3$BrPbCl$_2$ with Al$_2$O$_3$ scaffold as harvester light and absorption layer have been successfully fabricated. We obtained the best efficiency is (PCE = 0.12 %) for (FTO/TiO$_2$/CH$_3$NH$_3$IPbBr$_2$ perovskite with Al$_2$O$_3$ scaffold /CuI/Al electrode) device . We observed that PCE is increased when increased the absorption and decreased band gap energy of pervskeite layer of OPSCs. We observed increased the short current circuit to the device employed mixed halide of ( X = Br and I ) as absorption layer and increased open voltage circuit to the device employed mixed halide of ( X = Br and Cl) as absorption layer of devices.

References