

Synthesis and Characterization of flexible Al-doped SnO₂-based paper Electrode for Perovskite Solar Cell Application

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ABSTRACT: Primarily, Perovskite solar cells (PSCs) were disclosed merely in 2012 but their tremendously fast development has reached an efficiency of confirmed 25% presently. PSCs are more low-cost and easier to develop than Silicon-based solar cells. They can respond to many wavelengths of the electromagnetic spectrum which helps them transform more light into electricity. The performance and stability of PSCs are mainly reliant on the Electron transport layer (ETL) material. Lignocellulose (LC) is an abundant, environmentally friendly, and biodegradable fiber. In this research work, LC fiber is used as a substrate instead of a glass substrate due to its flexibility and better sustainability. For Electron transport layer materials in PSCs, several semiconductors can be utilized. In this research article, SnO₂ is used as an Electron transport layer material because of its low-temperature fabrication and wider band gap. Overall, SnO₂ exhibits great optical transparency, chemical stability, conductivity, and electron mobility which display its good photovoltaic properties. Aluminium was added into SnO₂ as a dopant for thin film enhancement and improved Power conversion efficiency (PCE). Al-doped SnO₂ proposes a fine surface coverage of thin films and enhances the conductivity of ETLs which results in the better performance of the PSCs. Different Characterization techniques such as X-Ray Diffraction (XRD), Scanning Electron Microscopy (SEM), and Ultraviolet-Visible Spectroscopy (UV-Vis) were used to characterize the synthesized material. The results display that the conductivity of Al-SnO₂ increased as the band gap reduced due to Al-doping and the inclusion of LC fiber. Therefore Al-doped SnO₂ constituting ETL prepared at the low temperature displays an enhanced charge transport than that of undoped-SnO₂. Hence, Al-doped SnO₂ is a favorable contender for ETLs used in PSCs which gives high Power conversion efficiency and stabilization.

Keywords: Perovskite solar cells, optical transparency, chemical stability, conductivity etc.

INTRODUCTION

Since the start of humanity, non-renewable energy sources are employed for the harvesting of energy. These non-renewable energy resources majorly consist of fossil fuels which affect the biosphere because of the constant release of carbon dioxide footprints [1]. The reserves of these fossil fuels are also depleting rapidly due to the rapid increase in population [2]. Therefore these non-renewable energy resources must become an element of the past as they do not deliver a sustainable solution to our needs for energy. So to fulfill the increasing energy demands and to handle environmental issues, renewable energy is an excellent alternative because it can lead to steady energy rates as these sources are not very much affected by geopolitical disasters, supply chain disturbances, and rate spikes [3].

Currently, solar energy is one of the most used forms of renewable energy because of its simple energy harvesting process. Perovskite solar cells (PSCs) are third generation solar cells that contain a perovskite-structured compound which is typically a mixture of organic or inorganic (tin or lead) halide established elements, as light collecting active layer [4]. PSCs are more viable and easy to manufacture than Silicon-based solar cells.

In solar cells, the glass substrates have sustainability issues as they are not flexible and face cracks after a certain period. Therefore natural fiber-based resources called flexible electrodes have extracted massive attention because of their flexibility, abundance, and eco-friendly attributes [5].

Metal oxides are in demand as they display high specific capacitance than carbon-based materials because of their higher mobility [6]. Tin Oxide (SnO₂) is a pleasing prospect as electron transport layer (ETL) material in PSCs because of its excellent attributes like low-temperature fabrication, wide band gap (3.6eV), high electron mobility (100 to 200 cm²V⁻¹ s⁻¹), better power conversion efficiency, and high photovoltaic performance [7].

In literature, Saleem et al. reported that metal oxide-based composite paper electrodes show better photovoltaic performance than glass electrodes as the surface area and absorbency of the working electrode are amplified due to lignocellulose [8].

Moreover, with the addition of lignocellulose, the conductivity of the paper electrode increases as the band gap decreases. So overall, the efficiency of solar cells increases due to all these aspects. Hao Chen et al. reported that the doping of aluminum on Tin oxide improves the transportation of charges and current density in the electron transport layer (ETL) of perovskite solar cells (PSCs) compared to the un-doped Tin oxide [9].

Chemical precipitation is a simple, low cost and environmentally friendly technique for the synthesis of nanoparticles [10] so it is used for the synthesis of SnO₂ nanoparticles. The presented study firstly includes the fabrication of SnO₂-based paper electrodes and then the deposition of an aluminium layer on a paper electrode.

This study is an effort to regulate the conductive properties of SnO₂-based paper electrodes by the deposition of an aluminium layer on a natural fiber-based substrate. The presented SnO₂-based paper electrode is a favorable contender as electron transport layer (ETL) in perovskite solar cells (PSCs) because of its flexibility, high mobility, and good conductivity.

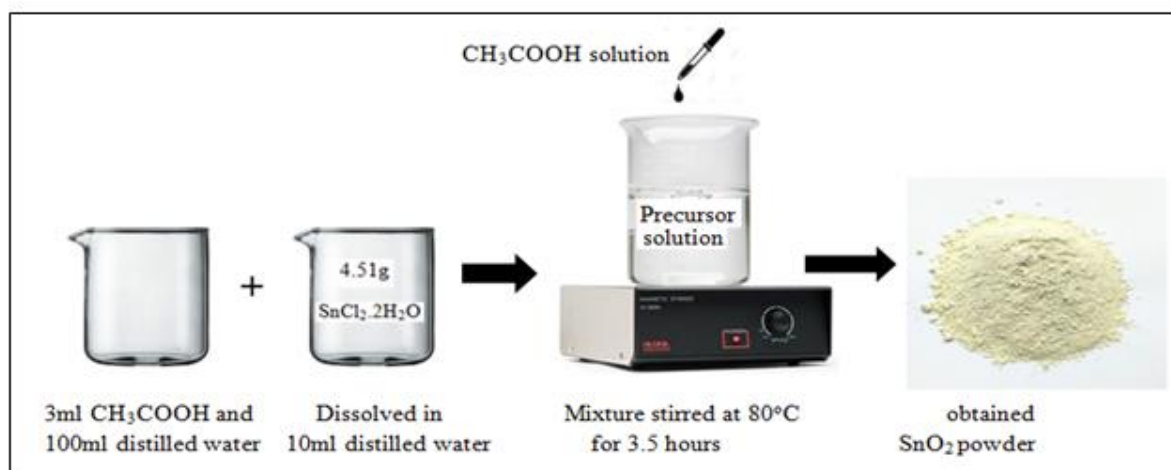


Figure 1: Synthesis of SnO₂ nanoparticles

Experimental

Reagents

Lignocellulose fibers were extracted from a plant named *Daucus carota*. Tin (II) chloride dihydrate (SnCl₂·2H₂O) and Acetic acid (CH₃COOH) were obtained from Sigma-Aldrich.

Synthesis of SnO₂ nanoparticles

3ml CH₃COOH and 100ml distilled water were mixed in a beaker under moderate stirring. 4.51g of SnCl₂·2H₂O precursor was dissolved in 10ml distilled water. Then both solutions were mixed using a magnetic stirrer at around 80°C for about three and half hours until the solution was dried and SnO₂ nanoparticles were acquired. The excellent obtained yield of SnO₂ powder is 3.407g.

Fabrication of Lignocellulose sheets

Daucus carota was used as the source for lignocellulose (LC) fibers. Firstly, 3 to 4 carrots were washed and crushed in a blender along with 50 ml distilled water until a paste was obtained. Then, the paste was converted into circular flexible LC sheets with the help of a vacuum pump and Buchner funnel. Lastly, the sheets were pressed between filter papers and dried at room temperature for about 48 hours. Hence, circular flexible LC sheets were obtained.

Fabrication of Lignocellulose LC/SnO₂ composite sheets

Firstly, 1g of SnO₂ powder and 0.5g of flexible LC sheet was weighed. The ratio of LC to SnO₂ powder is maintained at 1:2. Secondly, these two were crushed using a mortar and pestle along with some distilled water until a paste was formed. Thirdly, the paste was converted into circular flexible sheets using a vacuum pump and Buchner funnel. Lastly, the sheets were pressed between filter papers and dried at room temperature for about 48 hours. Thus, circular flexible LC-composite SnO₂ sheets were obtained.

Deposition of Aluminium on LC/SnO₂ composite Sheet

The deposition of Aluminium on the LC/SnO₂ sheet was done by using a vacuum coating unit. 0.1g of aluminium powder was measured and placed on the tungsten boat. Rotary and diffusion pumps were used to create the pressure inside of coating unit. Moreover, a transformer supplied electric current through the tungsten boat due to which the powder evaporated and deposited on LC/SnO₂ sheet

Characterization

X-Ray Diffraction (XRD)

XRD is used for examining the crystal structure of the material. XRD of SnO₂ nanoparticles is performed at room temperature using X-ray diffractometer for the structural analysis [11]. It can be seen visually from fig 1 that all the diffraction peaks are in decent contract with the data taken from JCPDS Card No: 41-1445 thus showing tetragonal crystal structure [12].

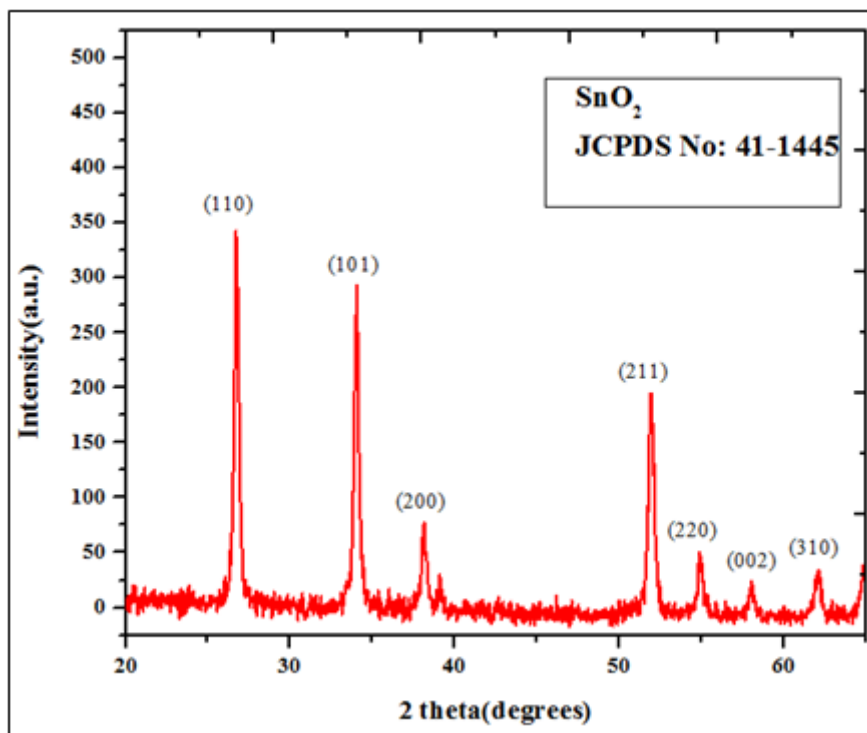


Figure 2: XRD spectra of SnO₂

The prominent XRD peaks in fig 2 are indexed as (110), (101), (200), (211), (220), (002) and (310) while the diffraction planes appeared at 26.71°, 33.42°, 37.15°, 52.12°, 55.05°, 57.52° and 62.45°. The highest intensity peak appeared at 26.71° which is indexed to plane (110). Moreover, the sharp peaks in fig 2 display that the material is highly crystalline [13]. The Crystallite size is determined by Scherer formula $D=0.9\lambda/\beta\cos\theta$ where 0.9 is shape factor, λ is the wavelength of x-rays, β is FWHM and θ is the angle of diffraction. The calculated crystallite size is 26.58nm. Furthermore, the interplanar spacing is determined by Bragg's law $n\lambda=2d\sin\theta$ where n is order of diffraction, λ is the wavelength of x-rays, θ is Bragg angle and d is interplanar spacing. The calculated interplanar spacing is 0.17nm.

Scanning Electron Microscopy (SEM)

SEM is an electron microscope which produces high resolution and magnified images of a sample surface. SEM images are 3-D in appearance and are beneficial for adjudging the surface morphology of sample's surface [14]. The average particle size and surface morphology were analyzed by scanning electron microscope.

From Fig 3, it is observed that the fibers are interconnected to bestow the strength which is required in contradiction of an applied strain to use as a flexible substrate. Moreover, the average particle size of LC/Al-SnO₂ is approximately 33.42nm which is calculated by ImageJ software.

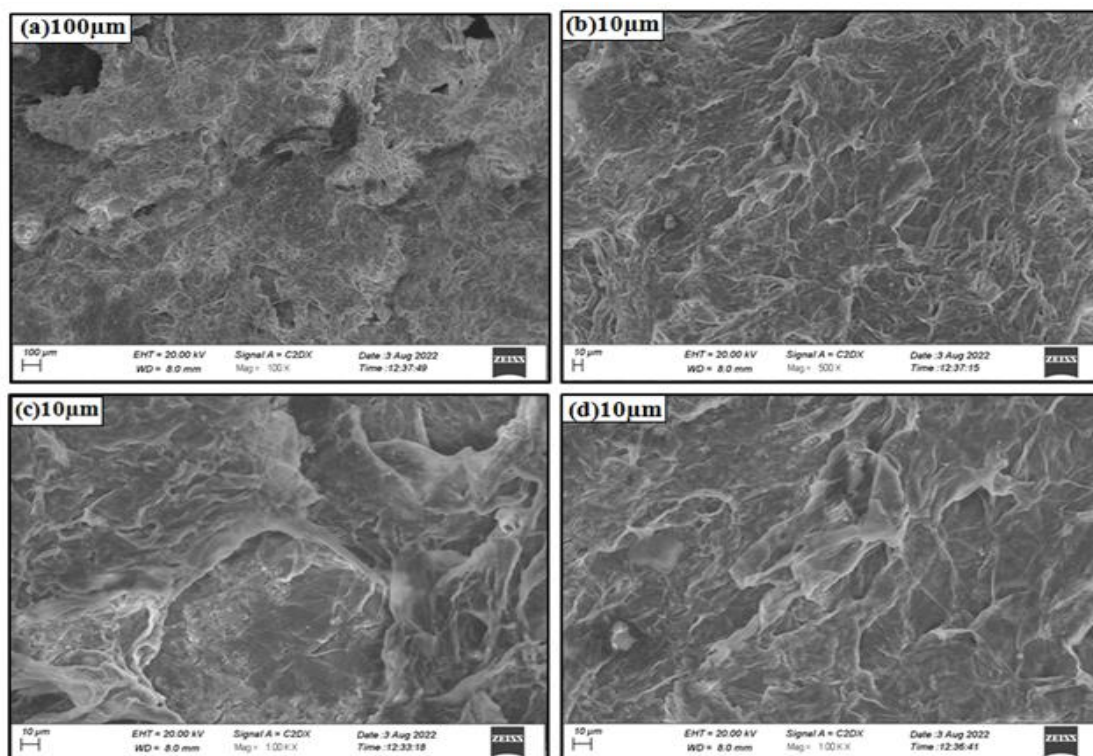


Figure 3: SEM image of LC/Al-SnO₂ at different magnifications (a) 100X (b) 500X (c, d) 1.00KX

As for the surface morphology of LC/Al-SnO₂, it can be seen that the surface is irregular and porous. For an ETL material, porosity is essential for better efficiency of electron transport since the porous surface improves the oxidation-reduction reaction [15]. Furthermore, the porosity gives flexibility to the composite sheets to work as flexible electrodes.

Ultraviolet-Visible (UV-Vis) Spectroscopy

UV-Vis Spectroscopy is a common characterization technique which displays the optical properties of a compound. In this characterization technique, light of the ultraviolet-visible region is absorbed by chemical compounds and spectrophotometer gives different spectra correspondingly [16]. In this study, the optical absorbance spectra used for experimentation was in the wavelength range of 350-700 nm.

From optical absorbance spectra, the energy band gap is calculated by using Tauc plot [17].

$$(\alpha h\nu)^n = K (h\nu - E_g)$$

Where α is coefficient of absorbance, $h\nu$ is photon energy, n is power factor of the transition mode whose value is 2 for direct band gap, K is an optical constant for direct transmission of light whose value is 1 and E_g is band gap energy.

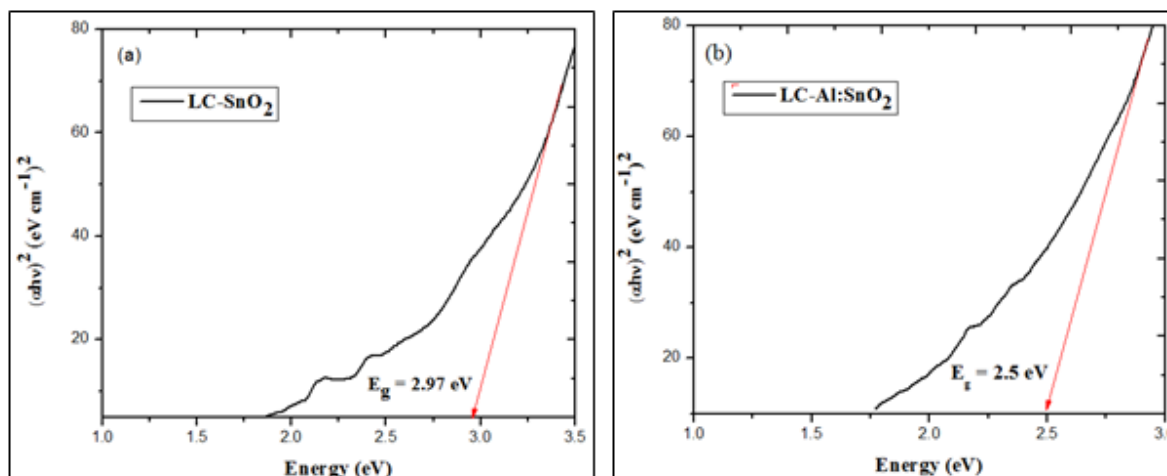


Figure 4: the UV-Vis graph of (a) LC/SnO₂ and (b) LC/Al-SnO₂ composite sheet

A direct fit was executed for the attained curve and the band gap energy was calculated by ignoring phonon's energy. The band gap energy of LC/SnO₂ is calculated as 2.97eV while the band gap energy of LC/Al-SnO₂ is calculated as 2.5eV. These

band gap energies signify that with the addition of LC and Al-doping, band gap decreases and conductivity increases which results in the better performance of the PSCs

CONCLUSION

In this research work, Tin Oxide (SnO₂) nanoparticles were synthesized by the chemical precipitation method. These nanostructures were used as an Electron transport layer material in perovskite solar cells because of their wider band gap and low-temperature fabrication process. For sustainability and flexibility, carrot sheets were prepared and used as substrates instead of ITO or FTO. Furthermore, Aluminium was deposited on LC/SnO₂ sheets for thin film enhancement and for improved conductivity which ultimately results in better Power conversion efficiency. The synthesized material was later characterized by XRD, SEM, and UV-Vis characterization techniques. By using XRD results, the calculated crystallite size of SnO₂ is 26.58nm and it has a tetragonal crystal structure. By using SEM results, the particle size of LC/Al-SnO₂ is calculated to be 33.42nm and has a porous and uneven surface morphology. The UV-Vis analysis displayed the maximum peak of LC/SnO₂ at 576nm and of LC/Al-SnO₂ at 573nm. Moreover, energy band gaps are calculated by using the Tauc plot method which shows the band gap of LC/SnO₂ as 2.97eV and of LC/Al-SnO₂ as 2.5eV. These band gap energies suggest that with the addition of LC and Al-doping, band gap decreases and conductivity increases which results in the better performance of the PSCs.

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