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Semiconducting Materials for Emerging Electronics and Technological Applications

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Abstract

Semiconducting materials are important for emerging electronic and photovoltaic technologies because their structural, optical and electrical properties can be tailored for specific device applications. In this study, a TiO_2-based composite photoanode is considered as a polymer-assisted semiconducting thin-film system in which inorganic TiO_2 nanoparticles are processed with organic film-forming components to produce a porous layer for dye-sensitised solar cell application. Nanocrystalline TiO_2 nanoparticles were synthesised from titanium isopropoxide using a hydrothermal method in an ethanol/deionised water medium. The nanoparticles were converted into a paste using ethyl cellulose and α-terpineol and coated onto fluorine-doped tin oxide glass substrates by the doctor-blade technique. Dye-sensitised solar cells were fabricated using TiO_2 films with thicknesses of 10, 12, 14 and 18 µm, N-719 dye, a platinum counter electrode and an iodide-based electrolyte. X-ray diffraction confirmed the formation of anatase TiO_2, while scanning electron microscopy and transmission electron microscopy showed nanoscale particles with porous and aggregated morphology. Photovoltaic measurements under simulated sunlight showed that film thickness strongly influenced dye adsorption, light harvesting, charge transport and recombination. The highest efficiency of 2.55% was obtained for the 14 µm TiO_2 film. The 18 µm film showed lower efficiency because of reduced transparency and increased recombination. These findings demonstrate that thickness optimisation of polymer-assisted TiO_2 composite photoanodes can improve DSSC performance and support the development of semiconductor composite films for photovoltaic and emerging electronic applications.

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INTRODUCTION

Semiconducting materials are at the heart of developments in electronic, optoelectronic and energy conversion technologies as their electrical, optical and structural properties can be tuned to suit diverse applications. Semiconductors are essential for devices that exploit the control of charge carrier motion in response to the application of tailored electrical and optical stimuli, such as solar cells, photodetectors, light emitting devices and sensors. As Chen and Mao [1] have noted, titanium dioxide-based nanomaterials have been extensively studied due to their tunable chemical composition, morphology, and surface properties for a wide range of applications. Likewise, Wang et al. [9] and Radisavljevic et al. [10] write that nanoscale

semiconductors are important for modern electronics and optoelectronics, as reduced dimensionality can alter charge transport, optical response and device behaviour.

Two-dimensional semiconductor materials are atomically thin or few-layer materials in which charge transport and electronic behaviour are mainly confined within a two-dimensional plane. These materials possess a very high surface-to-volume ratio, tunable band gap, strong light-matter interaction and excellent mechanical flexibility. Common examples include transition metal dichalcogenides such as MoS_2, WS_2, WSe_2, and other layered materials such as black phosphorus [3]. Because their electronic and optical properties can be controlled by changing the number of layers, strain, doping or heterostructure formation, 2D semiconductor materials are considered highly useful for next-generation transistors, photodetectors, sensors, flexible electronics and optoelectronic devices.

Graphene is considered promising for future electronics because it is a single atomic layer of carbon atoms arranged in a hexagonal honeycomb structure and shows excellent electrical conductivity, high carrier mobility, mechanical strength, thermal conductivity, optical transparency and flexibility. These properties make graphene suitable for high-speed electronic devices, transparent conducting electrodes, flexible displays, sensors, energy storage devices and advanced composite electronic materials [5]. However, pristine graphene has a zero band gap, so its direct use in switching devices is limited. This limitation can be addressed through approaches such as graphene nanoribbons, chemical functionalisation, doping and heterostructure design. Therefore, graphene remains an important material platform for future electronic and technological applications.

Solar energy is one of the most meaningful forms of renewable energy and can be converted into electricity with photovoltaic cells. Dye-sensitized solar cells (DSSCs) are considered highly desirable photovoltaics due to their relative ease of manufacture, low cost of materials and potential to be constructed into thin, flexible and lightweight structures. The first low-cost dye-sensitized solar cells were reported by O'Regan and Grätzel [2] based on colloidal Polymer-assisted TiO_2 thin films. Grätzel and Hagfeldt et al. [3, 4] later described a light-conversion mechanism in which the coupling between the dye molecules, the TiO_2 semiconducting composite photoanode, an electrolyte and a counter electrode determines the performance of DSSCs. In comparison to silicon-based solar cells, DSSCs decouple light harvesting and charge carrier transport. Here, the incident light is absorbed by the dye molecules and the electrons generated are extracted and transported through a wide band gap semiconducting material [3, 4].

A typical dye-sensitized solar cell consists of a dye-loaded nanocrystalline semiconductor film coated on a transparent conducting glass substrate, an electrolyte solution and a counter electrode. Titanium dioxide is the most widely used photoanode material for the dye-sensitized solar cells, because of its wide band gap, chemical stability, optical properties, cost and compatibility with nanostructured thin film [1, 4]. Under light excitation, these dye molecules promote electrons from the ground state to the excited state and inject these electrons into the conduction band of the TiO_2. These electrons then move through the TiO_2 network to the transparent conducting substrate and further via the external circuit to the counter electrode, generating a photocurrent and completing solar energy conversion [2–4]. Likewise Yella et al. [5] stated that the high performance of DSSC devices depends on the optimal design of sensitizer, redox mediator and semiconductor-based device architecture.

Semiconductors play a central role in renewable energy technologies because they can absorb photons, generate charge carriers and transport electrons and holes for energy conversion. In photovoltaic devices, semiconductors such as silicon, TiO_2, ZnO, CdTe, CIGS, perovskites and organic semiconductors are used to convert sunlight directly into electrical energy. In dye-sensitised solar cells, TiO_2 acts as a wide band gap semiconductor photoanode that accepts electrons injected from the excited dye molecules and transports them to the conducting substrate. Semiconductors are also used in photoelectrochemical water splitting, photocatalysis, solar fuel production, light-emitting devices and

power electronic systems used for renewable energy management [12]. Thus, semiconductor materials are essential for improving the efficiency, stability and scalability of clean energy technologies.

DSSC performance is influenced by a number of factors, including the particle size and morphology, porosity, crystallinity of the Polymer-assisted TiO_2 thin film, Polymer-assisted TiO_2 thin film texture, Polymer-assisted TiO_2 thin film thickness, dye loading, electrolyte and counter electrode properties [1, 4, 6]. Of these factors, the film thickness has a dominating influence on the performance of the cells because it affects the optical absorption, dye adsorption, electron generation and charge transport. Very thin Polymer-assisted TiO_2 thin films lack the surface area for efficient dye adsorption and result in reduced light absorption and photocurrent. Thick Polymer-assisted TiO_2 thin films can lead to poor electron transport through the film and increased recombination losses, which may reduce the photocurrent. Thus, the Polymer-assisted TiO_2 thin film thickness should be optimised to improve the photovoltaic performance. The TiO_2-based composite photoanode architecture can increase the DSSC performance through improving the light harvesting and charge transport, as indicated by Wang et al. [6].

TiO_2 can be synthesised into nanoparticles, nanotubes, nanorods, nanowires, and thin films. These different nanostructures provide different pathways for electron transport and surface areas for dye adsorption [1]. Nanocrystalline Polymer-assisted TiO_2 thin films can act as nanostructured electrodes for dye-sensitized solar cells (DSSCs). This is helpful because the pores are large enough to accommodate a high density of dye molecules, and the high surface area of TiO_2 nanoparticles increases the contact area between the dye and the semiconductor [1, 4]. At the same time, the connection between TiO_2 particles also influences the electron transfer, recombination, the current density and conversion efficiency of the DSSCs [3, 4, 6]. TiO_2 nanomaterials can be synthesised and deposited by different methods, such as sol-gel synthesis, hydrothermal synthesis, physical vapour deposition, chemical vapour deposition, spin coating, screen printing and doctor blading. The hydrothermal method can produce TiO_2 nanoparticles with controllable crystal structure and morphology. Furthermore, doctor blading is a simple and efficient coating method for the preparation of the TiO_2 paste on the transparent conducting glass (TCG) substrates [1, 6]. Doctor blade coating has then been employed for the TiO_2 paste as an approach to control the thickness of the Polymer-assisted TiO_2 thin film to study the effect of thickness on DSSCs.

In the context of polymer and composite studies, semiconducting materials are not limited to bulk inorganic solids. They are often processed as hybrid or composite thin-film systems in which inorganic nanoparticles are combined with organic binders, polymeric additives or flexible substrates to improve film formation, mechanical integrity, interfacial contact and device processability. TiO_2-based photoanodes used in dye-sensitised solar cells can therefore be viewed as functional semiconducting composite layers because the final device performance depends not only on the TiO_2 material itself but also on particle dispersion, porous nanoparticle-based composite architecture, binder-assisted coating and the interaction between the semiconductor layer, dye and electrolyte.

The TiO_2 nanoparticles were synthesised using a hydrothermal method from titanium isopropoxide precursor in ethanol and deionized water solutions and converted to a paste to be coated onto a fluorine-doped tin oxide glass substrate by a doctor blading process. The dye-sensitized solar cells were fabricated with different thicknesses of the Polymer-assisted TiO_2 thin films to study the effect of the thickness of the nanocrystalline Polymer-assisted TiO_2 thin film on the photovoltaic characteristics. We have studied the relationship between the structure of the semiconductor films and device properties by X-ray diffraction, scanning electron microscopy and transmission electron microscopy of the TiO_2 nanoparticles. The photovoltaic properties of the devices were studied by measuring the current voltage characteristics and the performance of the solar cells was evaluated in terms of open circuit voltage, short circuit current density, fill factor and photoelectric conversion efficiency. The study investigates

the influence of Polymer-assisted TiO_2 thin film thickness on DSSC performance, identifying the optimal thickness for improved DSSC performance. The research relates to the larger topic of semiconducting materials for next generation electronics and devices, since it shows that nanocrystalline semiconducting materials can be processed into a functional photovoltaic device and that the performance of electronic and energy conversion devices is not solely determined by the choice of semiconductor material but rather by processing and structural factors such as film thickness, morphology and surface area.

These structure-property relationships are also important in a wide variety of emerging electronics applications, including ZnO based devices [7, 8], two dimensional semiconductors [9–12], perovskite photovoltaics [13–16], organic semiconductors [17–22] and flexible polymer based electronics [23–32]. Hence, high quality, optimised TiO_2 thin films are likely to be useful in the production of lower cost, higher efficiency and more scalable photovoltaic devices and are likely to inform the design of other emerging electronic materials.

The present work is therefore positioned within the broader field of polymer-assisted and composite semiconductor films. Although TiO_2 is an inorganic semiconductor, its use in DSSCs involves paste formation, organic additives, coating control and multilayer device assembly. These features are closely related to composite material design, where the functional performance of a material depends on the interaction between the dispersed phase, processing medium, interface and final nanoparticle-based composite architecture. Studying Polymer-assisted TiO_2 thin film thickness in this context provides useful information for designing hybrid semiconductor composites for photovoltaic and emerging electronic applications.

MATERIALS AND METHODS

Materials

The titanium dioxide nanoparticles were created from titanium isopropoxide. Ethanol and deionized water were used as solvents, while adjusting the pH using nitric acid. The reaction was carried out using a hydrothermal synthesis method. The electrolyte consisted of iodine, lithium iodide, tetrabutylammonium iodide in acetonitrile and 4-tert-butylpyridine. The dye adsorbed on the TiO_2-based composite photoanode was N-719. Fluorine-doped tin oxide (FTO) glass was used as the transparent conducting substrate. The counter electrode was platinum-coated conducting glass. All chemicals were used in their analytical grade.

Ethyl cellulose and α-terpineol were used during paste preparation to improve the processability and coating behaviour of the TiO_2 nanoparticles. The presence of these organic processing components is important from a polymer and composite perspective because they assist in converting loose inorganic nanoparticles into a continuous thin-film structure suitable for device fabrication.

TiO_2 nanoparticles preparation

Nanocrystalline TiO_2 nanoparticles were prepared hydrothermally by adding titanium isopropoxide dropwise to the mixture of ethanol and deionized water, following broad principles of nanomaterial synthesis outlined by Chen and Mao [1]. The resulting solution was stirred and the pH was then adjusted to pH 0.7 with dilute nitric acid solution. After stirring for 3 hours, a milky white colloidal solution was obtained. The prepared colloidal solution was transferred to the internal Teflon reactor that was placed in a stainless-steel autoclave, as shown in Figure 1.

Ethanol was poured into the reactor to 80% of the volume of the autoclave. The autoclave was sealed tightly and placed in a hot-air oven at 180 °C for 6 h. The white precipitate was then separated by centrifugation at 2000 rpm and washed thoroughly with distilled water and ethanol to remove any

unreacted starting materials and other impurities. Finally, the TiO_2 nanoparticles obtained were dried in a vacuum oven at 100 °C for 5 hours.

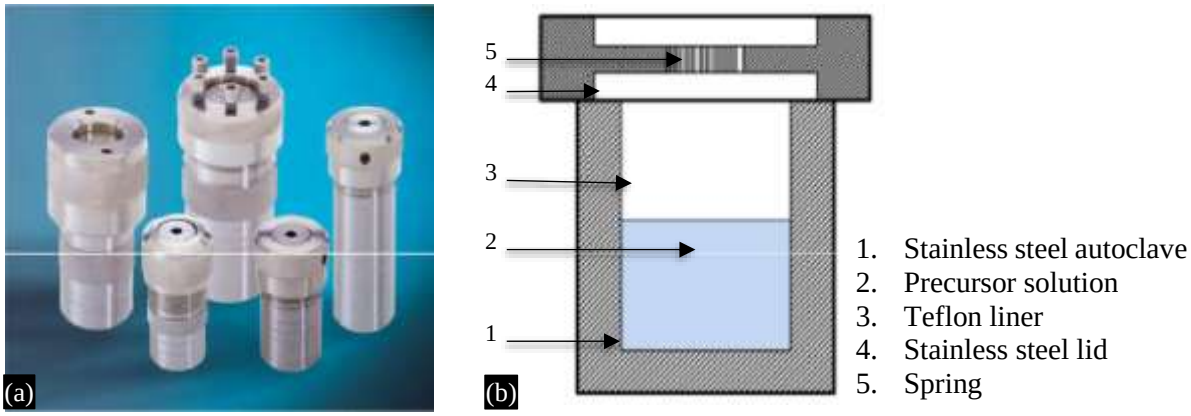


Figure 1. Autoclave and schematic diagram of hydrothermal synthesis setup: (a) Autoclave; (b) Schematic diagram of hydrothermal or solvothermal synthesis setup:

Preparation of TiO_2 Paste

The hydrothermally synthesised TiO_2 powder was converted into a paste for photoanode coating by mixing 0.5 g of TiO_2 powder, ethyl cellulose and α-terpineol in ethanol. The dispersion was mixed with the help of a mortar and pestle, then sonicated in an ultrasonic bath for 15 minutes to achieve a better dispersion and prevent agglomeration of particles.

The mixture was sonicated, then stirred at 50 °C in a thermostatic blender to remove the ethanol, and then the solvent and the water were finally removed with a rotary evaporator. The resultant TiO_2 paste was deposited by the doctor-blading method on a fluorine-doped tin oxide (FTO) glass substrate. The doctor-blading was repeated until the Polymer-assisted TiO_2 thin films reached the desired thickness, utilizing coating techniques similarly evaluated by Wang et al. [6] for forming composite gradient films.

This paste preparation step represents a polymer-assisted composite processing route. In this system, TiO_2 nanoparticles act as the functional inorganic semiconducting phase, while the organic medium supports dispersion, film continuity and coating uniformity. Therefore, the prepared Semiconducting thin-film composite layer can be interpreted as a semiconducting thin-film composite architecture rather than a simple powder coating.

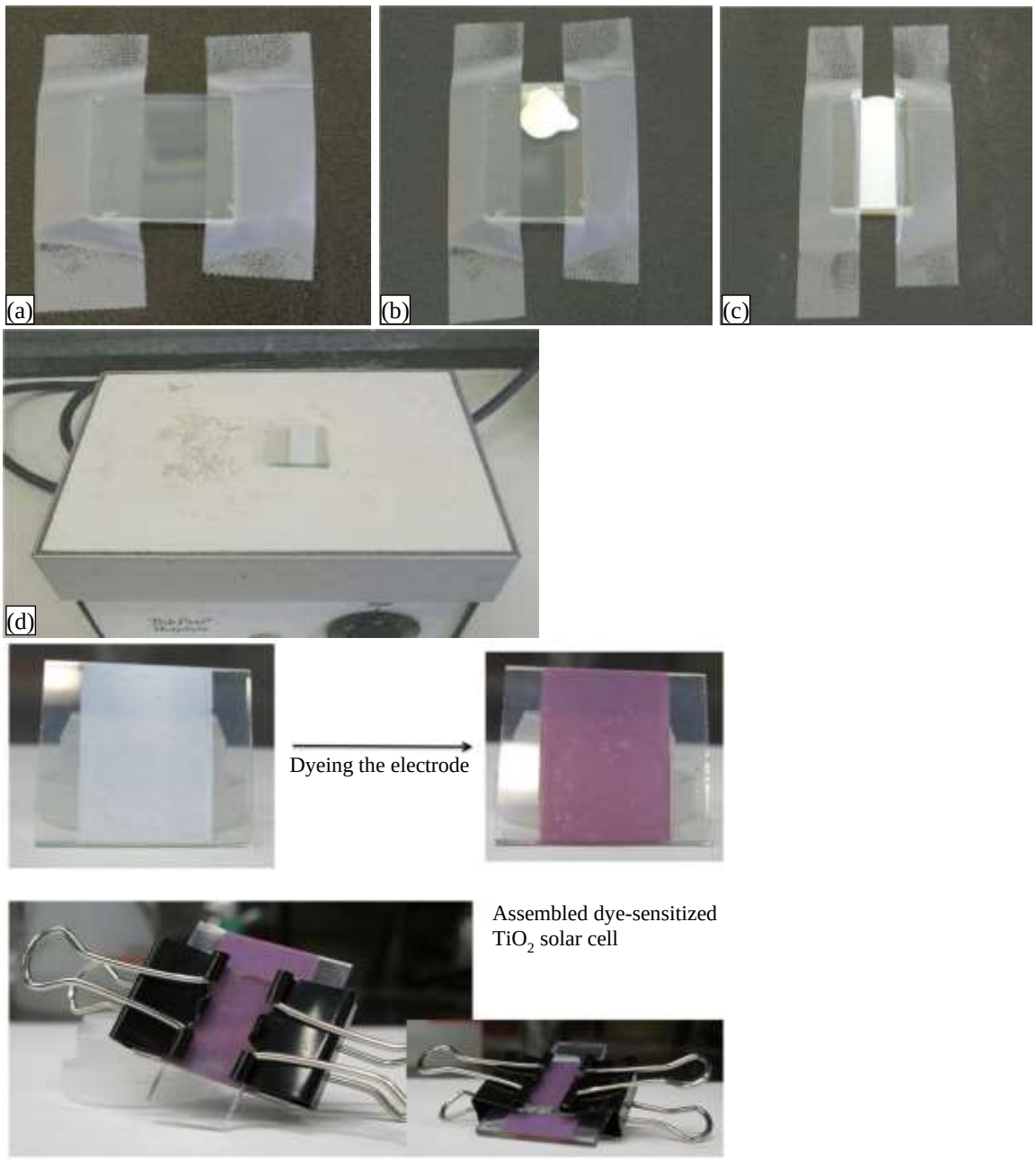
DSSC Fabrication

Fluorine doped tin oxide glass substrates are cleaned with tap water and detergent solution and ethanol, and stored in absolute ethanol after cleaning to avoid surface contamination. The TiO_2 paste is spread over the conducting side of the glass substrate by a doctor-blading technique, and heat-treated to improve the adhesion between the Semiconducting thin-film composite layer and the conducting substrate.

To accomplish the dye sensitization, the TiO_2 coated electrodes were immersed for 16 hours in a 0.3 mM ethanolic solution of the N-719 dye at room temperature. After the dye molecules were adsorbed onto the surface of the Polymer-assisted TiO_2 thin film, the electrodes were washed with ethanol to remove the loosely bound dye molecules and subsequently dried at 100 °C for 2 hours, consistent with typical sensitization methodologies discussed by Hagfeldt et al. [4]. Four TiO_2-based composite photoanodes with thicknesses of 10 µm, 12 µm, 14 µm, and 18 µm were synthesised, which all had an active area of 0.25 cm^2. The counter electrode was prepared by casting a platinum precursor solution on a conducting glass substrate and sintering it at 450 °C for 15 minutes. The DSSC was assembled by holding the dye-sensitised TiO_2 and platinum counter electrode together with binder clips. The complete fabrication steps of the dye-sensitized solar cell are outlined in Figure 2.

The electrolyte was composed of lithium iodide, iodine, tetrabutylammonium iodide and 4-tert-butylpyridine in acetonitrile. The electrolyte was injected between the dye-sensitised TiO_2 electrode and the platinum counter electrode. The completed dye-sensitised solar cell was illuminated at an irradiance of 100 mW cm^{-2} on an active area of 0.25 cm^2, mirroring the foundational evaluation structures established by O'Regan and Grätzel [2].

The fabricated DSSC represents a hybrid organic-inorganic device structure in which the TiO_2 semiconductor layer, dye molecules, electrolyte and conducting substrates operate together. From the viewpoint of composite materials, the performance of this device depends on interfacial compatibility, porous film structure and the controlled thickness of the semiconducting composite photoanode.



(a) Step 1: Put Scotch tape on the conducting side of ITO or FTO glass.
(b) Step 2: Put TiO_2 paste and flatten it with a razor blade on the conducting side of the FTO glass.

(c) Step 3: Place the coated electrode on a hot plate or in a hot-air oven at approximately 450°C for 10 minutes.
(d) Step 4: Dye the electrode and assemble the dye-sensitized solar cell.
Figure 2. Fabrication steps of dye-sensitized solar cell.

Photovoltaic Measurements

The photovoltaic responses of the fabricated dye-sensitised solar cells were measured by current-voltage characteristics using a xenon arc lamp as light source at a light intensity of 100 mW cm^{-2}. Current-voltage curves were measured by applying an external bias to the set-up with a digital source meter and by measuring the resulting photocurrent induced in the cell described by Yella et al. [5]. The main photovoltaic parameters calculated were open circuit voltage, short circuit current density, fill factor and photoelectric conversion efficiency, which is calculated using the following equation:

$$\eta = \frac{P_{max}}{P_{in}} \times 100$$

The fill factor was calculated as:

$$FF = \frac{P_{max}}{J_{SC} \times V_{OC}}$$

where maximum power is given by:

$$P_{max} = J_{max} \times V_{max}$$

Therefore, the fill factor can also be written as:

$$FF = \frac{J_{max} \times V_{max}}{J_{SC} \times V_{OC}}$$

The final equation for photoelectric conversion efficiency is:

$$\eta = \frac{J_{SC} \times V_{OC} \times FF}{P_{in}} \times 100$$

where J_{SC} is the short circuit current density, V_{OC} is the open circuit voltage, FF is the fill factor, P_{in} is the incident light power and P_{max} is the maximum power output of the cell. Table 1 presents a comparison of the I-V characteristics of DSSCs based on these formulas for Polymer-assisted TiO_2 thin films with different thicknesses.

The comparison of photovoltaic parameters seen in Table 1 shows that DSSC performance improved when the Polymer-assisted TiO_2 thin film thickness increased from 10 μm to 14 μm. The highest conversion efficiency of 2.55% was obtained for the 14 μm Polymer-assisted TiO_2 thin film. However, when the film thickness increased to 18 μm, the efficiency decreased to 0.85%, showing that excessive film thickness can reduce light transmission and increase recombination losses.

These results also show the importance of thickness control in semiconducting composite films. In a polymer-assisted TiO_2-based composite photoanode, film thickness affects not only light absorption but also dye penetration, electrolyte access, particle connectivity and electron transport. Therefore, the optimum 14 μm film can be understood as the most balanced composite architecture among the prepared samples.

RESULTS

In this section, structural, morphological, and photovoltaic characteristics of the prepared TiO_2 nanoparticles and fabricated DSSCs are presented. The crystallinity (phase) of TiO_2 was characterised using XRD. The surface morphology and particle size of the electrodes were investigated using scanning electron microscopy (SEM) and transmission electron microscopy (TEM). The structural

characterization of TiO_2 nanoparticles was performed using X-ray diffraction (XRD) analysis. The photovoltaic performance of DSSCs was studied by current-voltage measurements of Polymer-assisted TiO_2 thin films of different thicknesses.

XRD Analysis

The XRD pattern of the as-prepared TiO_2 nanoparticles was recorded in the $2.\theta$ range of 10° to 80° with CuKα radiation, and the average crystallite size of the anatase phase was calculated using the Debye-Scherrer equation:

$$D = \frac{K\lambda}{\beta \cos \theta}$$

where $D$is the crystallite size, $K$is the shape factor, $\lambda$is the wavelength of CuKα radiation, $\beta$is the full width at half maximum and $\theta$is the Bragg angle.

The generation of TiO_2 nanoparticles was confirmed by the XRD result. The results also showed that TiO_2 nanoparticles obtained were mainly in anatase phase, a crystalline phase that Chen and Mao [1] note as highly preferable for enhanced electron mobility. The major peaks observed in Figure 3 were 25.25°, 37.9°, 47.9°, 54.7° and 63.5° which correspond to the anatase form of TiO_2, and are associated with the tetragonal phase of titanium dioxide.

The broadening of these peaks suggests that nanosized crystallites can potentially be formed. Nevertheless, for dye-sensitized solar cell application (DSSC), the formation of anatase phase of TiO_2 is preferred because of its better electronic, surface and dye adsorption characteristics. The ethanol-rich condition and monitoring of acidic media during synthesis of TiO_2 nanoparticles assist in the formation of the anatase phase. Small crystallite sizes are considered helpful for DSSCs, since these nanoparticles offer larger surface area which can help improve dye loading and in turn charge production, as reinforced by Chen and Mao [1].

Table 1. Comparison of the I-V characteristics of DSSCs based on Polymer-assisted TiO_2 thin films with different thicknesses.

Sample	Polymer-assisted TiO_2 thin film Thickness (μm)	VOC (V)	JSC (mA cm^{-2})	FF	η (%)
1	10	0.60	3.48	0.593	1.23
2	12	0.79	4.05	0.6012	1.92
3	14	0.78	5.48	0.595	2.55
4	18	0.53	3.04	0.519	0.85

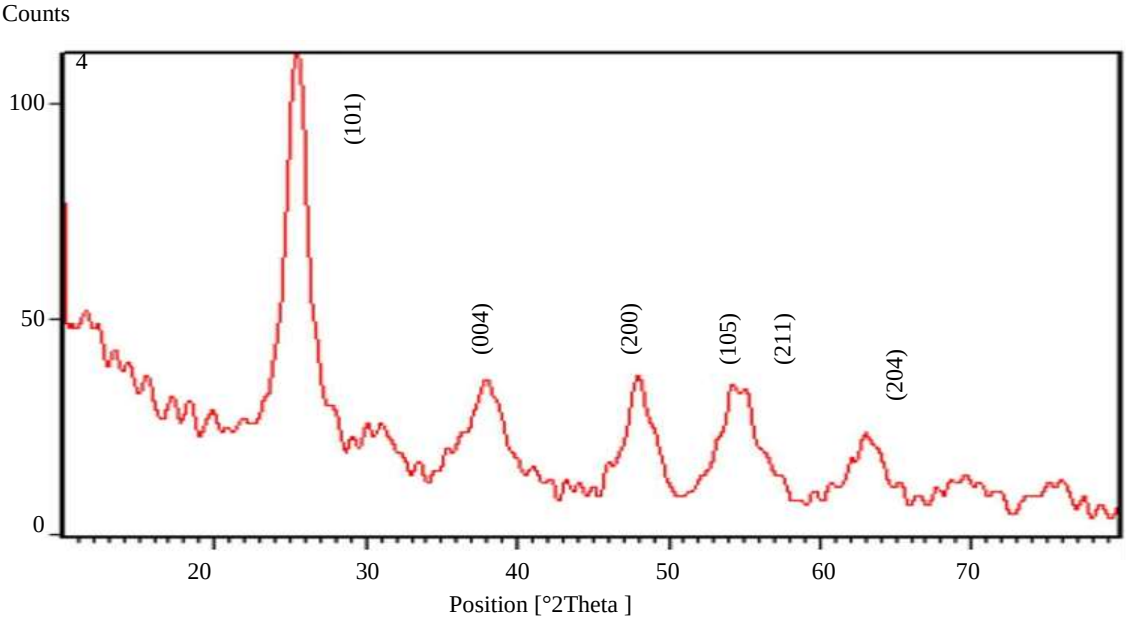


Figure 3. X-ray diffraction peaks of hydrothermally prepared TiO_2 nanoparticles.

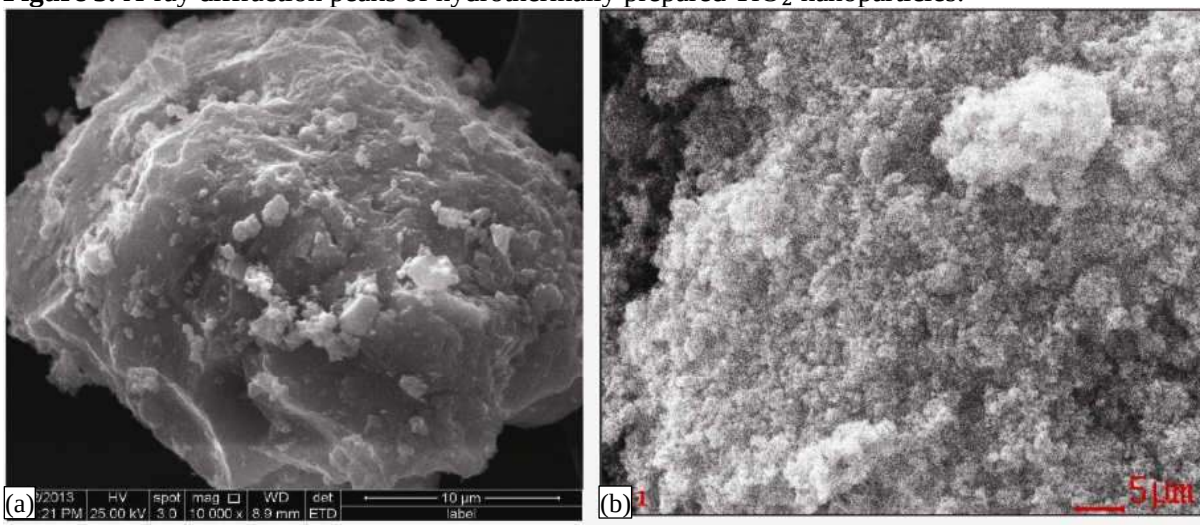


Figure 4."SEM micrograph of as-prepared TiO_2 nanoparticles.(a) SEM micrograph of as-prepared TiO_2 nanoparticles; (b) Higher magnification image.

SEM and TEM Analysis

The morphology of the prepared TiO_2 nanoparticles was further examined by means of scanning electron microscopy (SEM) and transmission electron microscopy (TEM). The SEM micrograph shown in Figure 4 revealed a clumpy surface morphology. This morphology can be due to the agglomeration of TiO_2 nanoparticles during the preparation and drying steps, although the agglomerated nanoparticles were not clearly appreciated in the SEM image. However, a connected particle network eases electron transfer through the Polymer-assisted TiO_2 thin film lattice structure.

The SEM images showed that the TiO_2 particles formed a porous rough structure; this is helpful for DSSCs because a rough structure increases the surface area for dye adsorption. Increasing the amount of dye that is adsorbed can improve light absorption and in turn increase the number of photoexcited electrons, making the surface morphology of the TiO_2 important to photovoltaic performance, mirroring findings established by Hagfeldt et al. [4].

TEM images, as depicted in Figure 5, further confirmed the nanoscale characterization of the prepared TiO_2 particles. The estimated size of the particles was around 20 to 25 nm, and their aggregates were also observed.

Nanoparticles with a small size have a large surface area, which helps in the efficient adsorption of dye molecules on the surface. However, particle agglomeration does cause degradation to the quality of the resultant film and impedes electron transport, meaning methods to improve particle dispersion and film preparation are needed. TEM analysis confirmed results obtained from the XRD analysis as nanocrystalline TiO_2 was observed. The nanometer-scale dimensions of TiO_2 help to improve surface contact, dye adsorption, and light harvesting of the dye-sensitized solar cells.

The observed porous and interconnected particle morphology is significant for polymer and composite studies because it reflects the role of particle packing, dispersion and interfacial contact in a functional thin-film composite system. Such morphology helps improve dye adsorption and electron movement, but excessive agglomeration may reduce film uniformity and limit charge transport.

Evaluation of DSSC Performance

The prepared TiO_2 electrodes sensitised with the N-719 dye were then used to fabricate the DSSCs, characterised by measuring their current-voltage (I-V) characteristics under simulated sun illumination. Figure 6 displays the photocurrent-voltage characteristics of DSSCs for different thicknesses of the Polymer-assisted TiO_2 thin film.

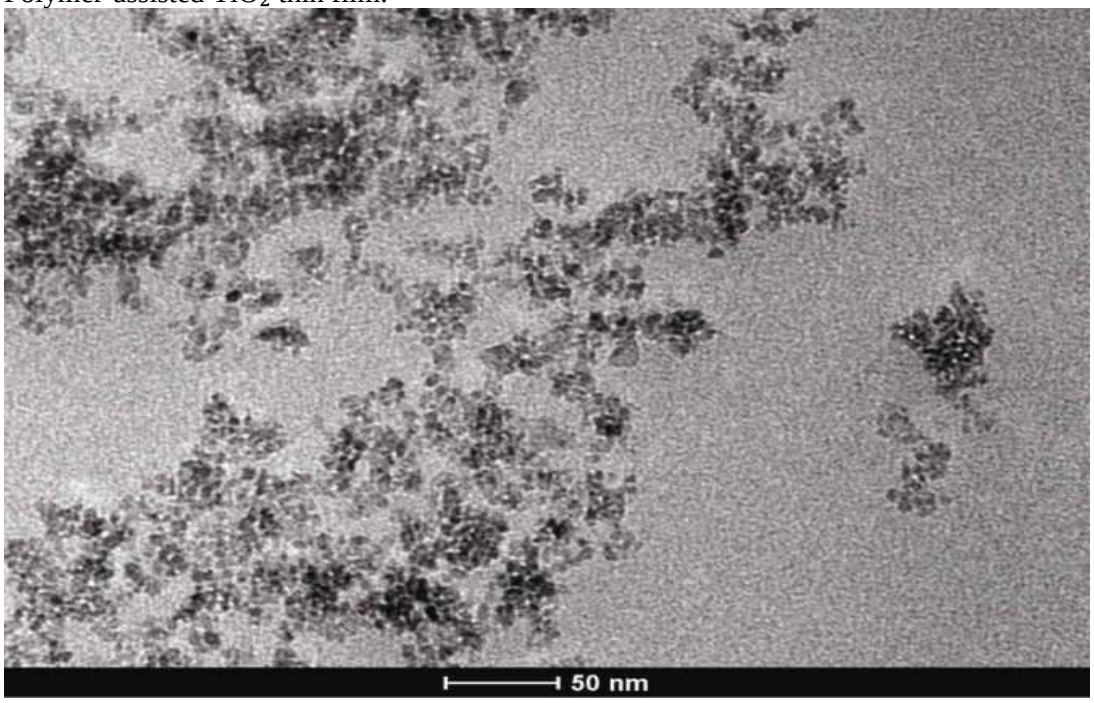


Figure 5. TEM micrograph of TiO_2 nanoparticles.

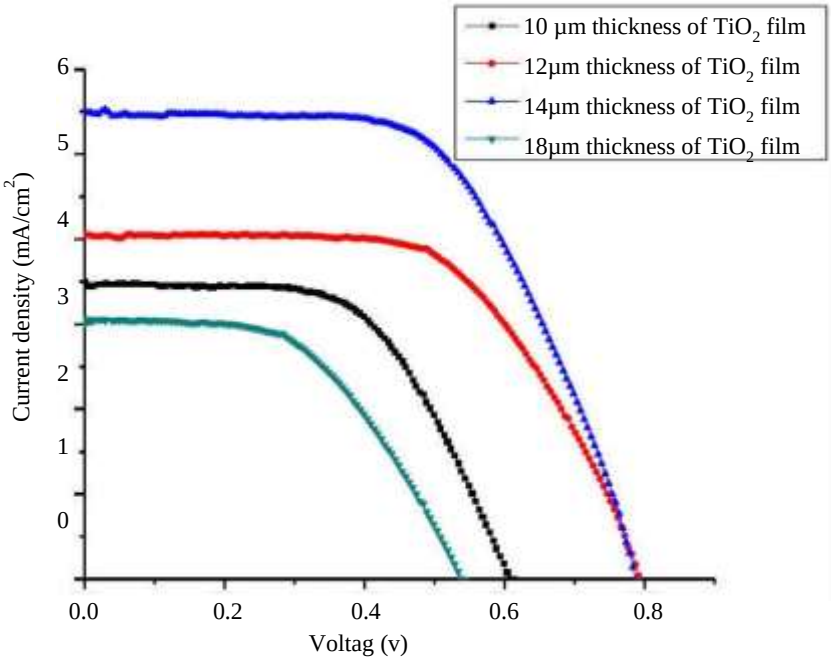


Figure 6. Photocurrent-voltage characteristics of DSSCs for different thicknesses of the Polymer-assisted TiO_2 thin film.

The DSSCs were prepared with differing Polymer-assisted TiO_2 thin film thicknesses of 10, 12, 14, 16, and 18 µm as shown in Table 1. Polymer-assisted TiO_2 thin film thickness could be used to modulate the performance of the DSSC. As the thickness was increased from 10 µm to 14 µm, the Jsc and efficiency improved. An increase in the short circuit current density from 3.48 mA cm^{-2} to 5.48 mA cm^{-2} was observed with an increase in the thickness of the film from 10 µm to 14 µm due to an increase in the surface area available for the adsorption of dye. Increasing the thickness of Polymer-assisted TiO_2 thin films increased the absorption of dye which in turn increased the number of photo-excited electrons, resulting in a higher open circuit voltage. An increase in thickness from 10 to 12 µm was observed to have this effect.

The 12 µm film had the highest open circuit voltage, whereas the 14 µm film had a similar open circuit voltage with a higher current density. This means that the increase in conversion efficiency was due to an increase in photocurrent rather than an increase in voltage.

The best performing DSSC, which had the highest energy conversion efficiency of 2.55%, was based on a 14 µm thick Polymer-assisted TiO_2 thin film, showing that this thickness was optimal amongst all the samples. This thickness allowed sufficient adsorption of dye, proper light absorption, as well as efficient electron transport in the Polymer-assisted TiO_2 thin film. The film was subsequently thick enough to adsorb additional dye molecules, but not so thick that electron-hole recombination losses were greater than the additional absorption.

Further, for an 18 µm thick Polymer-assisted TiO_2 thin film, an energy conversion efficiency of 0.85% was obtained, suggesting that there may be an optimum film thickness. Additionally, a very thick Semiconducting thin-film composite layer can also reduce transparency, increase the length of electron transport and provide recombination sites, all of which would reduce the amount of electrons available to the collecting electrode, thus decreasing photocurrent and efficiency. Figure 7 displays the variation of conversion efficiency, confirming that conversion efficiency increases with increasing TiO_2

thickness up to an optimum point.

As shown in Figure 7, the conversion efficiency was maximum with a thickness of about 14 μm. At 18 μm, it decreased. The Polymer-assisted TiO_2 thin film thickness must therefore be carefully optimised: a thin Polymer-assisted TiO_2 thin film has low dye adsorption and poor light absorption, while thick films limit charge percolation and increase recombination. Hence optimum film thickness for photovoltaic performance is an intermediate thickness. In summary, experimental results show that nanocrystalline TiO_2 is indeed a good semiconducting material for DSSC photoanodes and that the parameters of TiO_2 structure and morphology have a strong influence on dye adsorption, electron generation and electron transfer. The study illustrates the importance of Polymer-assisted TiO_2 thin film thickness optimization for achieving high-efficiency dye-sensitized solar cells, and is relevant to the use of nanostructured semiconducting materials in other emerging photovoltaic and electronic devices.

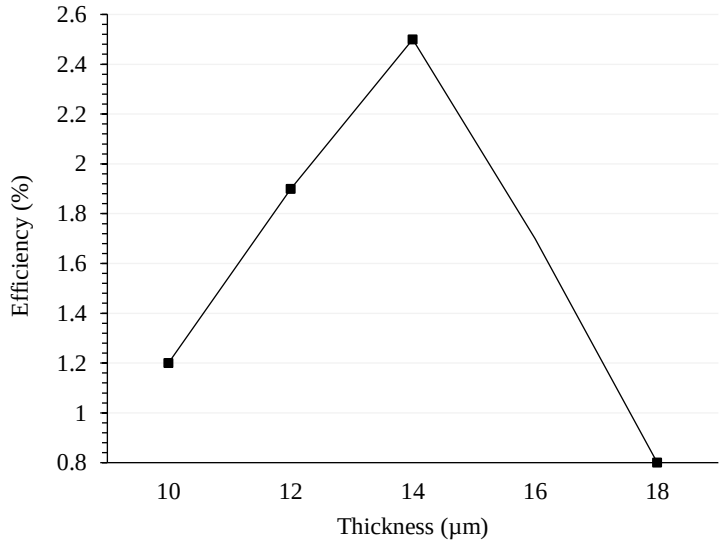


Figure 7. Variation of conversion efficiency of DSSCs with different thickness of the Polymer-assisted TiO_2 thin film.

DISCUSSION

The present study shows that nanocrystalline TiO_2 is a good choice as a semiconducting material in the photoanodes of DSSCs, owing to its structural integrity, nanoscale structure and good charge transport properties. TiO_2 has been considered as a prime semiconducting material for photocatalytic and photovoltaic applications, on account of its properties such as wide band gap, chemical stability and tunable nanostructure, as outlined by Chen and Mao [1]. Most of the resulting TiO_2 was identified by XRD as anatase TiO_2, which is a suitable material for the surface activity and electron transport generally associated with dye-sensitized solar cells. O'Regan and Grätzel [2] first demonstrated the role of dye-sensitized colloidal Polymer-assisted TiO_2 thin films for low cost photovoltaic applications. The dependence of structure of TiO_2-based composite photoanode on loading with dyes, electron injection and recombination was discussed by Grätzel [3] and Hagfeldt et al. [4]. The broad XRD peaks in this study indicated that the crystallites were nano-sized. Additional evidence of the aggregated presence of TiO_2 nanoparticles was obtained from the SEM and TEM images, where the particle size was approximately 20 to 25 nm. Nanosized TiO_2 is helpful: the surface area is maximised and multiple dye molecules can be adsorbed to generate more photoexcited electrons.

The photovoltaic measurements show that the thickness of the Polymer-assisted TiO_2 thin film had a strong influence on the performance of the dye-sensitized solar cells. When the thickness of the Polymer-assisted TiO_2 thin film was increased from 10 μm to 14 μm, the short circuit current density increased from 3.48 mA cm^{-2} to 5.48 mA cm^{-2}, and the conversion efficiency increased from 1.23% to

2.55% (Table 1). This performance improvement can be attributed to the better dye adsorption, light harvesting and electron generation characteristics of the thicker Polymer-assisted TiO_2 thin films. Wang et al. [6] also demonstrated that the architecture of the TiO_2-based composite photoanode can improve DSSC performance via control of the structure of the semiconductor film.

From a polymer and composite materials perspective, the improvement in DSSC performance with controlled TiO_2 thickness can be linked to the formation of an effective semiconducting composite photoanode. The TiO_2 nanoparticles provide the inorganic charge transport network, while the organic processing medium helps in film formation and coating stability. The photovoltaic performance is therefore governed by a structure-property relationship similar to that found in polymer composites, where filler distribution, interfacial contact, porosity and film continuity strongly influence functional behaviour. As reported by Yella et al. [5], the performance of the DSSC strongly depends on the efficiency of the sensitizer with respect to the semiconducting composite photoanode and the electrolyte. In this paper, a thickness of 14 μm was selected as a balance between surface area and the charge transport. This increase in efficiency arises because, although the open circuit voltage did not vary much between the 12 μm and 14 μm films, the short circuit current density increased considerably; therefore, increasing the film thickness mainly increases the photocurrent rather than the voltage.

The decrease in efficiency at 18 μm further confirms that composite film design requires optimization rather than simple material addition. A thicker Semiconducting thin-film composite layer may provide more surface area for dye adsorption, but it can also increase electron transport distance, reduce optical transparency and create more recombination sites. This behaviour is highly relevant to composite and polymer-based electronic materials, where excessive filler loading or uncontrolled film thickness can reduce rather than improve functional performance. The fact that the efficiency is reduced at 18 μm shows that a thicker Polymer-assisted TiO_2 thin film is not necessarily helpful. There are obvious advantages of increased dye-adsorption, but the distance that electrons must travel from the photoexcited dye increases as well. Increased distance may increase charge recombination losses, which results in lower charge collection. This explains the drop in efficiency from 2.55% at 14 μm to 0.85% at 18 μm. These results suggest that the nanoparticle-based composite architecture, morphology and processing conditions are as critical to device performance as the choice of material itself and, indeed, this is true for other emerging electronics. Morphological and process control is also important in organic semiconductors, electronic polymers and flexible photovoltaics, which has been discussed by Facchetti [19], Sirringhaus [22], Someya et al. [24], Krebs [27] and Søndergaard et al. [28]. In summary, our results suggest that optimised nanocrystalline TiO_2 thin films may be used for low cost photovoltaic applications and may act as a useful model for semiconducting material development for emerging electronic and technological applications.

CONCLUSION

Ultimately, the present study shows that nanocrystalline TiO_2 is a good semiconducting material for dye-sensitized solar cells and has a great potential in other electronic and photovoltaic devices. Hydrothermally synthesised TiO_2 nanoparticles are predominantly in the anatase phase and have nanosized dimensions and aggregated morphologies which are helpful for increasing the surface area for dye adsorption. Photovoltaic data shows that DSSC efficiency is related to the Polymer-assisted TiO_2 thin film thickness: increasing from 10 to 14 μm the efficiency goes from 1.23 to 2.55% due to the increase in dye loading, light harvesting and charge collection.The efficiency decreases to 0.85% for an 18 μm film due to transparency loss, enlarged electron diffusion distance and increased recombination losses. The optimum surface area, light absorption and charge transport combination was achieved using the 14 μm Polymer-assisted TiO_2 thin film. Thus it becomes clear that the resulting performance of semiconducting materials does not only depend on composition, but also on morphology, crystallinity, nanoparticle-based composite architecture and processing.

These findings show that the performance of semiconducting Polymer-assisted TiO_2 thin films depends not only on chemical composition but also on morphology, crystallinity, particle connectivity, film thickness and processing conditions. From the perspective of polymer and composite studies, the TiO_2-based composite photoanode can be interpreted as a polymer-assisted semiconducting thin-film composite in which inorganic nanoparticles are processed into a functional porous layer for photovoltaic use. The study therefore supports the design of hybrid, polymer-assisted and composite semiconductor films for low-cost photovoltaic devices and emerging electronic applications.

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Conflict of Interest Statement

The authors declare that there is no conflict of interest regarding the publication of this manuscript.

Data Availability Statement

The data supporting the findings of this study are available from the corresponding author upon reasonable request.

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