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Optimization of Tannic Acid Polymerization to Improve the Efficiency of Dye-Sensitized Solar Cells: Effect of Temperature and Monomer Concentration

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Abstract

This study investigates the effects of temperature and monomer concentration on tannic acid polymerization to enhance dye-sensitized solar cell (DSSC) efficiency. Tannic acid was polymerized using Trimethylolpropane Triglycidyl Ether (TMPGDE) as a crosslinker, with monomer concentrations of 2, 2.5, and 3 grams and temperatures ranging from 30°C to 110°C. UV-Vis analysis revealed a bathochromic shift in poly-tannic acid, indicating increased light absorption in the 200-800 nm range. FTIR confirmed the formation of new ether groups, signifying successful polymerization. The highest DSSC efficiency (8.984%) was achieved with 2.5 grams of monomer at 50°C, significantly outperforming unpolymerized tannic acid (1.35%). The optimal poly-tannic acid has an average molecular weight of 32,610.1568 and a polymerization degree of 19.182. This research demonstrates the potential of tannic acid polymerization for improving DSSC performance, paving the way for more efficient and cost-effective organic solar cells.

Keywords: *DSSC, Polymerization, Tanic Acid, Titanium Dioxide, Crosslinking.*

Introduction

Indonesia is a country with high energy consumption, growing at 7% per year, with 95% of it coming from fossil fuels [1–3]. The availability of fossil fuels on Earth is dwindling as demand increases every year, making the use of renewable energy sources a solution [4,5]. Solar energy is one type of alternative energy that can be used [6–9]. The amount of solar radiation energy received by the earth ranges from 1017 Watts per day, which is equivalent to burning 1 billion tons of coal per hour and solar cells can convert this sunlight into electrical energy [10–12]. One of the third-generation solar cell is dye-sensitized solar cell (DSSC) [8,9,13–17]. In DSSC, the photovoltaic effect converts photon energy into electrical energy [18–20]. DSSC does not require high purity materials, with low production costs and energy consumption [21–23].

Organic and synthetic dyes can be used in DSSC. Synthetic dyes have an average efficiency of 10%, but they are relatively expensive and have a toxic effect for the environment [24–28]. Organic dyes have low production costs and energy requirements, but the efficiency is still low [29]. The low efficiency produced is due to unstable dyes, low compound content in plant extracts, etc [17,30–35]. As a result, it is necessary to modify the dye to improve the efficiency of organic dye DSSC [17,26,36]. Organic dyes that have been used as DSSC applications include co-pigmented anthocyanins with 4.14 percent efficiency, beta-carotene with a 0.003 percent efficiency, chlorophyll with a 0.1249 percent efficiency, and curcumin with a 0.07 percent efficiency [30–32].

Tannic acid is a hydrolysable tannin exhibiting colorless to pale yellow solutions and have significant antioxidant properties in various applications [37–39]. The conjugated aromatic system of this compound can excite electrons or

photons from sunlight [40–43]. The amount of sunlight that can be converted into electrical energy is affected by the number of conjugated double bonds and autochrome substitution in the chromophores [44–47].

Polymerization is a method for synthesizing conjugated polymers with extended π-conjugation and chromophore incorporation [48–50]. Polymerization is a polymer formation reaction that involves a specific rearrangement [51]. Tannic acid can be polymerized through cross-linking with Trimethylolpropane Triglycidyl Ether (TMPGDE). Tannic acid will bind to TMPGDE to form ether and more linear OH groups [52,53]. In DSSC, tannic acid polymerization has been carried out, including the formation of covalent polymers (poly-tannin resins) through formaldehyde condensation in acidic conditions, which are then cross-linked with glutaraldehyde in alkaline conditions; this pathway extends the conjugated system, causing a shift in absorption (bathochromic) and a significant increase in cell efficiency [54,55]. In addition, metal coordination "polymerization" forms a metal–phenolic network that acts as a co-sensitizer; its performance is pH-sensitive and has been reported to outperform free TA on TiO_2 photoanodes [56–58].

This research investigates the effects of temperature and monomer concentration on tannic acid polymerization with Trimethylolpropane Triglycidyl Ether (TMPGDE) as a crosslinker to enhance DSSC efficiency. Tannic acid was chosen for its potential as an organic dye in DSSCs. Tannic acid serves as an effective dye and co-sensitizer in DSSCs due to its polyphenol groups, which exhibit strong binding to TiO_2 which facilitates efficient electron injection and regeneration processes, reducing electron recombination and allowing for better light absorption [59–61]. The resulting poly-tannic acid was characterized using UV-Vis spectrophotometry and FTIR analysis to confirm successful polymerization and improved light absorption properties. DSSCs electrical properties were measured to determine the optimal polymerization conditions. By examining these aspects, this research sought to enhance DSSC performance through the optimization of tannic acid polymerization, potentially paving the way for more efficient and cost-effective organic solar cells.

Experimental

Equipment

The equipment employed includes a digital balance, spatula, stirring rod, dropper, beaker, measuring cup, dark bottle, magnetic stirrer, oven, iron tongs, and Ostwald viscometer. Additionally, analytical instruments such as a UV-Vis Spectrophotometer, FTIR, and Multimeter are utilized.

Material

For materials, the list consists of sodium hydroxide (NaOH), potassium iodide (KI), acetonitrile, iodine (I_2), copper (II) chloride ($CuCl_2$), polyethylene glycol 400 (PEG 400), Degussa P-25 titanium dioxide, soy lecithin, trimethylolpropane glycerol diethoxylate (TMPGDE), ethanol, hexane, polyvinyl alcohol (PVA), candles, matches, and distilled water (aquadest). This comprehensive set of tools and materials suggests a complex experimental setup, likely involving chemical synthesis, solution preparation, and various analytical measurements.

Methods

Dye Preparation

A measured amount of tannic acid was dissolved in 5 mL of 0.2 M sodium hydroxide solution, with the quantity varying as per experimental requirements. A 0.5 mL aliquot of this solution was then dispersed in 15 mL of 0.1 M lecithin. This mixture underwent vigorous stirring with cyclohexane for 1 hour. Subsequently, 115 µL of trimethylolpropane glycerol diethoxylate (TMPGDE) was introduced to the mixture, followed by intense stirring for 12 hours at various temperatures, as specified by the experimental design. The resulting solution was then centrifuged at 12,000 rpm for 2 minutes, maintaining a temperature of 20°C. The precipitate obtained from centrifugation was subjected to a washing

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process using cyclohexane and an ethanol-water mixture. Following this, the washed samples were again centrifuged at 12,000 rpm for 2 minutes to separate the precipitate from the solution. Finally, the isolated precipitate underwent oven drying for one hour at 60°C. This procedure describes a complex process involving dissolution, dispersion, stirring, centrifugation, washing, and drying steps, likely for the preparation of a specific compound or nanoparticle formulation.

Electrolyte Preparation

In two separate containers, potassium iodide (KI) and iodine (I_2) were dissolved in acetonitrile. The solutions were combined and stirred until a homogeneous mixture was achieved. To create a gel-like consistency, polyethylene glycol (PEG) was introduced to the mixture and thoroughly blended [62,63]. This procedure describes the preparation of a PEG-based gel electrolyte containing KI and I_2. The use of acetonitrile as a solvent facilitates the dissolution of the ionic components, while the addition of PEG transforms the liquid solution into a gel-like material. Such PEG-based gel electrolytes are often employed in various applications, including energy storage devices and electrochemical systems, due to their favorable properties such as ionic conductivity and mechanical stability [62,64].

TiO_2 Paste Preparation

PVA (1 g) was dissolved in 10 ml distilled water, stirred at 80°C until suspended. The suspension was mixed with TiO_2 (Degussa P-25, 1 g) in a mortar. $CuCl_2$ was added, and the mixture was ground to a homogeneous paste.

Counter Electrode Preparation

The conductive surface of the ITO glass is exposed to a candle flame to deposit a carbon layer. The glass is held over the flame until a substantial carbon coating accumulates on the conductive side. Once coated, the carbon layer is shaped or trimmed to form a 1 cm x 1 cm square area on the ITO glass surface [65]. This method, known as the flame-soot technique, creates a carbon-based counter electrode for dye-sensitized solar cells. The candle soot provides a cost-effective and simple way to produce a catalytic carbon layer, which can enhance the performance of the solar cell by facilitating the reduction of tri-iodide ions [8,66]. The thickness and uniformity of the carbon layer can be controlled by adjusting the exposure time to the candle flame. This process results in a functional carbon electrode that can be used in conjunction with the dye sensitized TiO_2 electrode to complete the solar cell assembly.

Dye Sensitized Solar Cell (DSSC) Assembly

ITO glass was cut and prepared for TiO_2 deposition. The glass was dried at 60°C on a hot plate. The TiO_2 layer was soaked in dye solution for 30 minutes. The counter electrode was placed on top of the TiO_2 layer and clamped with clips. Liquid electrolyte was then added dropwise between the electrodes. This process outlines the assembly of a dye-sensitized solar cell, including photoanode preparation, dye sensitization, cell assembly, and electrolyte addition.

Electric current test

A 24-watt UV lamp is utilized as the light source. To measure current and voltage, the multimeter's positive lead is connected to the anode, while the negative lead is attached to the cathode.

Results and Disscusion

Synthesis of Poly-Tannic Acid Dyes

The dye was synthesized through the polymerization of tannic acid. This process involved mixing dissolved tannic acid (monomer) with 0.2 M NaOH and allowing the reaction to proceed for 15 hours. The mass of tannic acid used was varied, with monomer concentrations of 2 grams, 2.5 grams, and 3 grams. These variations were selected because the amount of tannic acid affects the degree of monomer binding to the crosslinking agent. In this reaction, NaOH served as an initiator, facilitating the cleavage of the epoxy group on the crosslinking agent, thereby enabling tannic acid to bond with it.

The crosslinking agent used was Trimethylolpropane Triglycidyl Ether (TMPGDE), a non-polar compound containing three reactive epoxy groups. When these epoxy groups react with NaOH, a radical carbon atom is formed, which binds to tannic acid, resulting in the formation of an ether group that links tannic acid to TMPGDE. Due to the polarity difference between tannic acid and TMPGDE, a surfactant—lecithin dissolved in hexane—was employed to ensure proper mixing. The tannic acid solution was stirred with the surfactant in hexane for one hour to ensure uniform dispersion before adding TMPGDE and continuing stirring for another 15 hours.

The polymerization reaction was conducted at various temperatures. Afterwards, the mixture was centrifuged at 20°C for 10 minutes to separate unreacted components from poly-tannic acid. The polymer, having a higher molecular weight, settled at the bottom as a brown precipitate. The mass of poly-tannic acid obtained varies depending on the initial tannic acid concentration and polymerization temperature.

The results showed that at a monomer concentration of 2 grams and a temperature of 50°C, the highest mass of poly-tannic acid (0.322 grams) was produced. This aligns with previous research indicating that increasing temperature increases polymer mass production due to faster reaction rates caused by increased particle movement and collision frequency. However, beyond 50°C, excessive heating led to a steady decline in polymer yield because tannic acid decomposes at high temperatures (98.89°C–101.67°C). At temperatures of 70°C, 100°C, and 110°C, decomposition reduced the amount of poly-tannic acid formed.

For monomer concentrations of 2.5 and 3 grams, the highest polymer masses were observed at 70°C—0.312 grams and 0.209 grams, respectively—while the lowest masses were recorded at 110°C (0.106 grams and 0.047 grams). At higher monomer concentrations, reactions occur more rapidly due to increased molarity, which accelerates particle collisions and reaction rates. However, excessive monomer concentrations can lead to unstable homopolymer formation and hinder diffusion within the crosslinking agent.

At 70°C, higher monomer concentrations resulted in faster polymerization compared to reactions at lower temperatures like 50°C. For concentrations of 2.5 and 3 grams, polymerization occurs more efficiently than at 2 grams due to faster reaction rates; however, excessive monomers also impeded diffusion in some cases. Notably, the polymer mass obtained at a concentration of 2.5 grams exceeds that at 3 grams because excessively high monomer levels can disrupt crosslinking efficiency by promoting homopolymer formation instead of stable polymers.

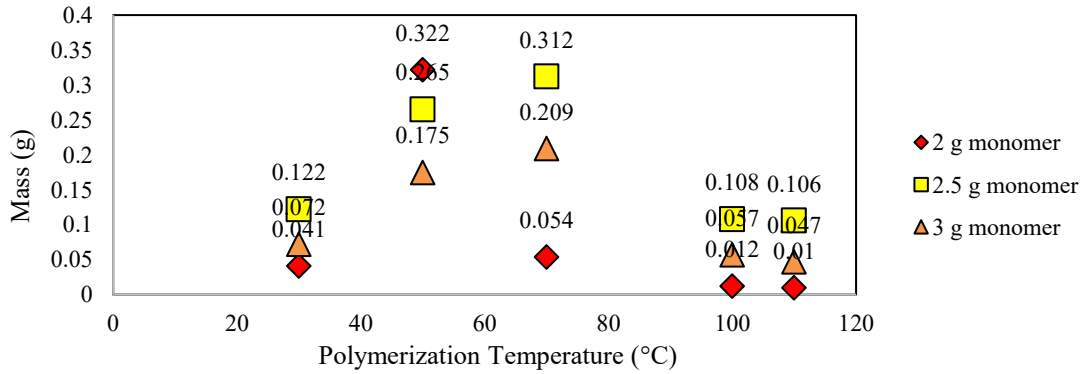


Figure 1. Poly-tannic acid mass produced at various polymerization reaction temperatures with initial monomer concentrations of 2.0, 2.5, and 3.0 grams.

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Characterization of Poly-Tanic Acid by UV-Vis and FTIR

The UV-Vis spectrophotometer test aims to analyze the impact of polymerization on the dye's optical properties. Based on the UV-Vis spectrophotometer analysis, the polymerized dye exhibits a bathochromic effect, also known as a red shift, where the maximum absorption wavelength shifts to a longer wavelength. In this case, the absorption peak shifted from 220 nm in tannic acid to 240–244 nm in poly-tannic acid, depending on temperature and monomer concentration variations (as shown in Table 2). This shift is attributed to changes in the electronic structure caused by polymerization, which extends conjugation and lowers the energy gap between molecular orbitals, facilitating light absorption at longer wavelengths.

Poly-tannic acid also demonstrates a broader absorption band compared to tannic acid. While tannic acid shows an absorption band within 200–500 nm, poly-tannic acid polymerized at 30°C extends its absorption band to 200–700 nm. At higher polymerization temperatures (50°C and 70°C), the absorption band further expands to 200–800 nm. This broader absorption range enhances light-harvesting capabilities, making the dye more effective for applications such as dye-sensitized solar cells (DSSCs).

The improved absorption properties are linked to the addition of electrons during polymerization, which increases light absorption efficiency. A stronger absorbance intensity within the UV-Vis spectrum correlates with higher energy capture and conversion potential. As poly-tannic acid exhibits higher absorbance intensity than tannic acid within the 200–800 nm region, it is expected to improve DSSC efficiency by enhancing light harvesting and electron injection into the semiconductor.

The relationship between temperature and monomer concentration also plays a critical role in determining dye performance. Higher temperatures generally increase polymerization rates due to faster molecular motion and collision frequency, resulting in greater conjugation and broader absorption bands. However, excessive heating beyond 98.89°C–101.67°C—the decomposition points of tannic acid—reduces polymer yield and absorption efficiency due to degradation of organic compounds.

Fourier-transform infrared (FTIR) spectroscopy was conducted to identify the functional groups present in both tannic acid and the poly-tannic acid compound produced through polymerization. This analysis provides insight into the structural changes that occur during the polymerization process. Table 3 summarizes the FTIR characterization of tannic acid, which reveals key functional groups.

Table 1. Table of variations of the variables used.

	30 ° C (X_1)	**50 ° C(X_2)**	**70 ° C(X_3)**	**100 ° C(X_4)**	**110 ° C(X_5)**
2.0 g (Y_1)	X_1Y_1	X_2Y_1	X_3Y_1	X_4Y_1	X_5Y_1
2.5 g (Y_2)	X_1Y_2	X_2Y_2	X_3Y_2	X_4Y_2	X_5Y_2
3.0 g (Y_3)	X_1Y_3	X_2Y_3	X_3Y_3	X_4Y_3	X_5Y_3

Table 2. UV-Vis results of tannic acid and poly-tannic acid.

Cell Sequence	Max Wavelength (nm)	Abs	Abs Area (nm)
X_1Y_1	241	2.599	200-650
X_1Y_2	241	2.812	200-700
X_1Y_3	241	2.787	200-650
X_2Y_1	244	2.730	200-800
X_2Y_2	244	2.839	200-800
X_2Y_3	244	2.778	200-800
X_3Y_1	243	2.699	200-800
X_3Y_2	243	2.773	200-800
X_3Y_3	243	2.975	200-800
X_4Y_1	240	2.703	200-500
X_4Y_2	240	2.810	200-500
X_4Y_3	240	2.713	200-500
X_5Y_1	240	2.141	200-500
X_5Y_2	240	2.321	200-500
X_5Y_3	240	2.342	200-500
Tannic acid	220	1.892	200-500

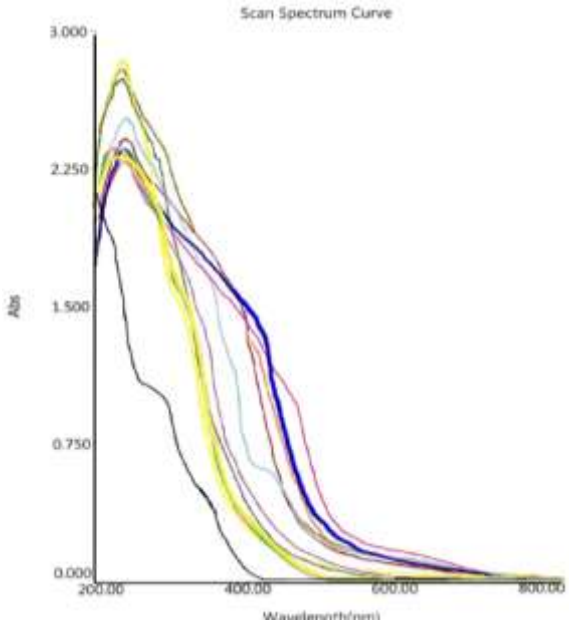


Figure 2. Tannic acid (black) and poly-tannic acid UV-Vis results (colored).

The FTIR spectrum of tannic acid shows a broad peak at 3300–3600 cm^{-1}, corresponding to the O-H stretching vibrations of phenolic groups. This broad band indicates strong hydrogen bonding, a characteristic feature of polyphenols. Additionally, a prominent peak at 1700 cm^{-1} represents the C=O stretching vibrations of carbonyl groups. More specifically, the tannic acid spectrum includes a broad band at 3323 cm^{-1} (O-H group of polyphenols) and a peak at 1763

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cm^{-1} (C=O group), confirming its aromatic nature. Other notable peaks are observed at 1636, 1609, and 1449 cm^{-1}, which are associated with aromatic ring vibrations and C=C stretching, further corroborating the presence of aromatic compounds.

Poly-tannic acid, formed through polymerization, exhibits significant changes in its FTIR spectrum compared to tannic acid. These changes reflect alterations in functional groups and bonding patterns due to crosslinking and polymerization. For example, shifts in the O-H and C=O peaks indicate modifications in hydrogen bonding and carbonyl interactions resulting from the formation of ether linkages between tannic acid molecules and crosslinking agents like TMPGDE. The broader absorption bands in poly-tannic acid suggest an increase in molecular complexity and conjugation.

FTIR analysis is crucial for understanding how polymerization affects the chemical structure of tannic acid. The observed changes in functional groups confirm successful polymerization, which enhances the dye's properties for applications such as dye-sensitized solar cells (DSSCs). The increased molecular complexity of poly-tannic acid contributes to improved light absorption and electron transfer capabilities, making it more effective as a dye material.

FTIR spectroscopy not only identifies functional groups but also highlights structural transformations during polymerization. These findings demonstrate how polymerization enhances the chemical and physical properties of tannic acid, enabling its use in advanced applications such as DSSCs. The detailed spectral data provides a foundation for optimizing polymerization conditions to achieve desired material properties.

The FTIR analysis of poly-tannic acid revealed the presence of key functional groups that confirmed the successful formation of the polymer. A broad peak at 3300–3600 cm^{-1} corresponds to the O-H stretching vibrations of phenolic groups, indicating strong hydrogen bonding. A distinct peak at 1700 cm^{-1} represents the C=O stretching vibrations of carbonyl groups, while a peak at 1050 cm^{-1} corresponds to the CH_2-O ether group. The presence of this ether group is significant as it confirms the formation of a polymer, resulting from the reaction between tannic acid and the crosslinking agent.

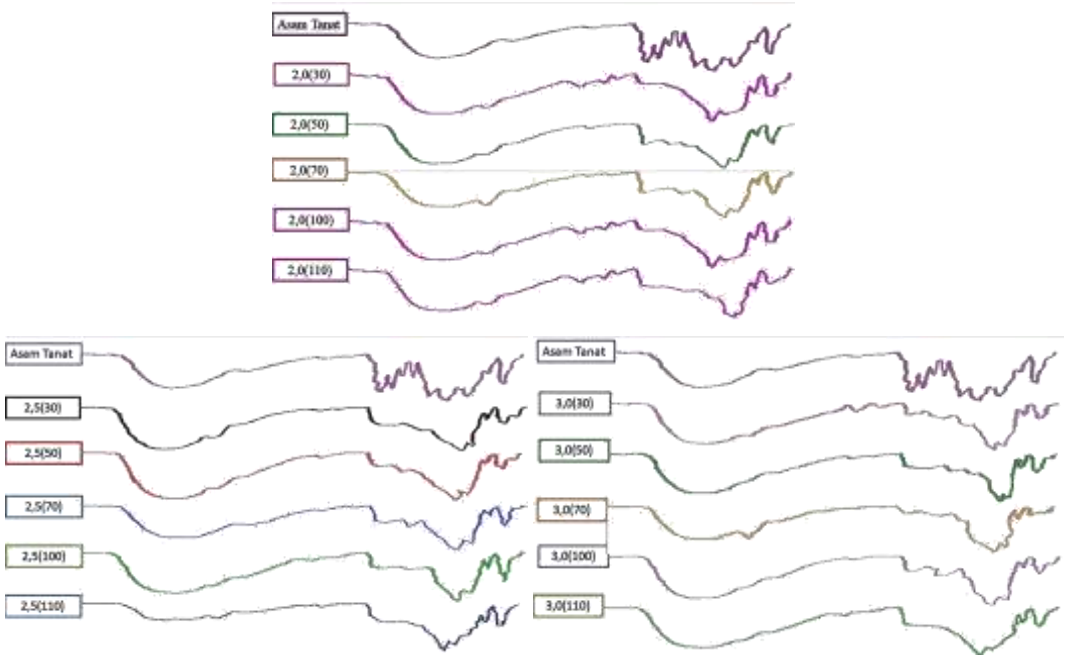


Figure 3. Tannic acid (tanic acid) and poly-tannic acid FTIR spectra at various temperatures with monomer concentrations of 2.0 g (upper), 2.5 g (below left), and 3.0 g (below right).

Table 3. Tannic acid FTIR data interpretation.

Wave number (cm^{-1})	Theoretical wave number (cm^{-1})	Functional groups
3332,67; 2721,60	3300-3600	-OH (phenol)
1703,38	1690-1760	C=O (aldehydes, ketones, carboxylic acid esters)
1636; 1609;15	1610-1680	C=C (alkenes, aromatics)
746,64	675-995	C-H (Alkenes, aromatic rings)
1449	1340-1470	C-H (alkane)

The appearance of an ether band at ~1050 cm^{-1} not only confirms the successful cross-linking between tannic acid and TMPGDE, but also implies an increase in dye stability. This bond reduces the solubility of poly-tannic acid in the electrolyte and protects the aromatic framework from degradation, allowing more dye to remain attached to the TiO_2 surface during operation. The π conjugation formed through cross-linking reduces the energy gap and broadens light absorption, facilitating more efficient photoexcitation and electron injection into the TiO_2 conduction band. This synergistic effect explains why poly-tannic acid exhibits a much higher DSSC efficiency (8.984%) compared to unpolymerized tannic acid (1.35%).

Additionally, because poly-tannic acid is derived from tannic acid, its FTIR spectrum retains characteristic features of tannic acid, such as phenolic O-H and carbonyl C=O peaks. These overlapping spectral features further confirm that poly-tannic acid is composed of tannic acid units linked by crosslinking agents. The retention of these functional groups also suggests that poly-tannic acid preserves some of the intrinsic chemical properties of tannic acid while gaining new properties from polymerization. In addition to spectroscopic and efficiency analyses, the polymerization results were evaluated in terms of average molecular weight and degree of polymerization, as described in the Molecular Weight section.

Cell Efficiency

The efficiency of dye-sensitized solar cells (DSSCs) was evaluated using a 24-watt UV lamp as the light source and a multimeter to measure voltage and resistance. The current generated by the DSSC was calculated using Ohm's Law, $V=I.R$, where V is the voltage (in volts), R is the resistance (in ohms), and I is the current (in amperes). This approach allows for a quantitative assessment of DSSC performance.

According to the data, the efficiency of DSSCs varied significantly with changes in polymerization temperature and monomer concentration. For a monomer concentration of 2 grams, the highest efficiency (3.386%) was achieved at 50°C, while the lowest efficiency (0.18%) occurred at 110°C. The trend indicates that cell efficiency initially increases with temperature due to improved polymerization but decreases sharply at higher temperatures as tannic acid decomposes. Similarly, for a 2-gram monomer concentration, efficiency rose from 7.54% at 30°C to 8.984% at 50°C before declining to 4.928% at 70°C and further dropping to 0.217% and 0.126% at 100°C and 110°C, respectively.

For a higher monomer concentration of 3 grams, efficiency also peaked at lower temperatures. At 30°C, the efficiency was 6.9%, increasing to 7.77% at 50°C but declining to 7.07% at 70°C and plummeting to 0.2301% and 0.032% at 100°C and 110°C, respectively. These results suggest that while polymerization reactions proceed more effectively at moderate temperatures, excessive

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heat leads to tannic acid decomposition, reducing both polymer mass and cell efficiency.

The performance of dye-sensitized solar cells (DSSCs) is closely influenced by the polymerization conditions, with temperature, monomer concentration, and dye-semiconductor interactions playing critical roles. Moderate temperatures, such as 50°C, are ideal for polymerization as they enhance molecular interactions without causing the decomposition of tannic acid, an organic compound that begins to degrade beyond its thermal stability range of approximately 98.89–101.67°C. In contrast, higher temperatures (100–110°C) lead to tannic acid decomposition, reducing the production of poly-tannic acid and ultimately lowering the quality of the dye and the efficiency of the cell.

Monomer concentration also significantly impacts DSSC performance. While increasing monomer concentrations can accelerate reaction rates and improve polymer yield, excessively high concentrations may hinder diffusion within the polymer matrix or result in the formation of unstable homopolymers, which negatively affect overall efficiency. Furthermore, the interaction between the dye and the semiconductor surface is crucial for efficient electron flow in DSSCs. The hydroxyl groups in poly-tannic acid facilitate strong bonding with the TiO$$_2$$ semiconductor surface, enhancing electron injection and improving overall cell performance. These factors collectively determine the effectiveness of DSSCs, highlighting the importance of optimizing polymerization conditions for achieving high energy conversion efficiency.

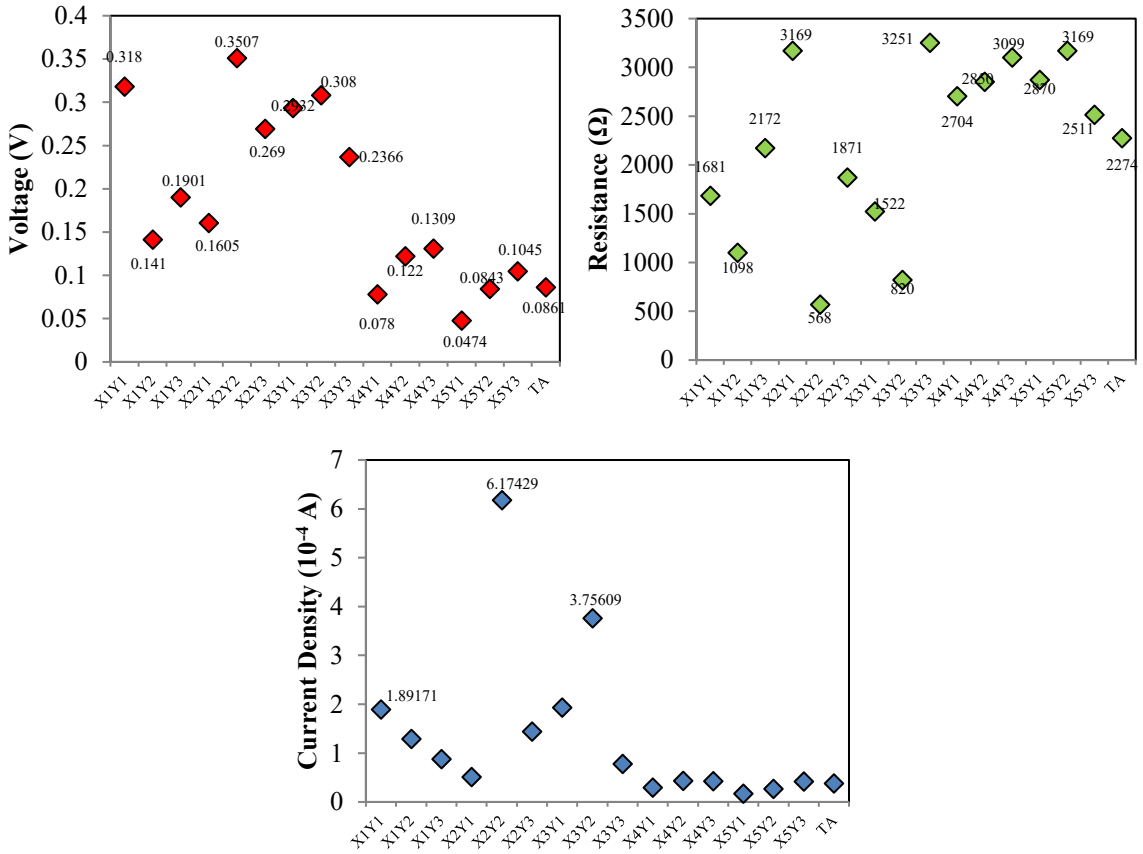


Figure 4. Electrical characteristics of DSSC devices with poly-tannic acid dye: (a) Voltage, (b) Resistance, and (c) Current density for different polymerization conditions.

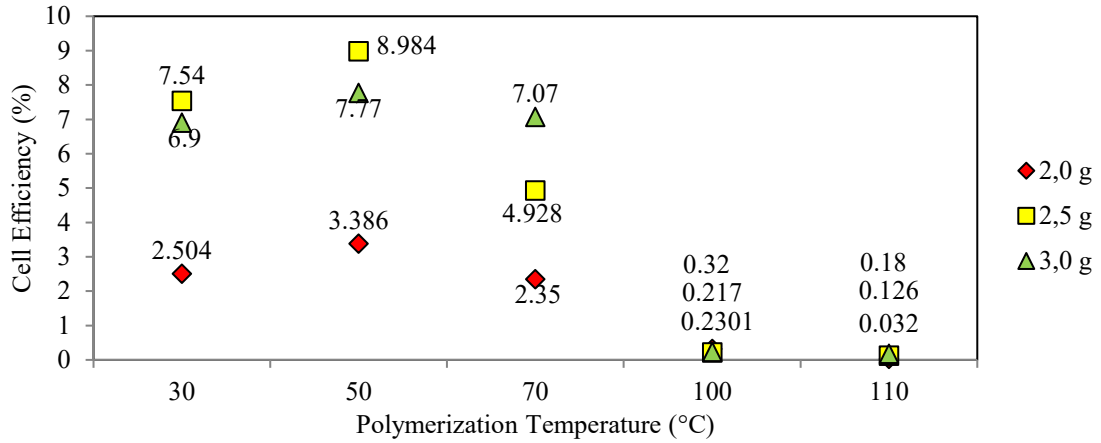


Figure 5. Cell efficiency at monomer concentrations of 2.0 grams, 2.5 grams and 3.0 grams in variations in polymerization temperature.

The highest efficiency observed (8.986%) occurred with a monomer concentration of 2.5 grams polymerized at 50°C (variation X2Y2). This combination likely optimized both dye quality and its interaction with the semiconductor, resulting in efficient electron transfer pathways within the DSSC circuit.

Polymerized tannic acid sensitizers produce a power conversion efficiency of 8.984%, far above most natural dyes, which generally fall below 3–5% [67–70]. Although slightly lower than the standard ruthenium N719 (9–11%), these results show that polymerized bio dyes are capable of approaching the performance of cutting-edge synthetic metal-organic dyes and offer a more environmentally friendly alternative [71].

In contrast, the lowest efficiency (0.012%) was recorded for a monomer concentration of 3 grams polymerized at 110°C (variation X5Y1). This poor performance can be attributed to tannic acid decomposition under excessive heat, which reduced both polymer mass and dye quality.

In conclusion, DSSC efficiency is influenced by multiple factors including temperature, monomer concentration, polymerization conditions, and dye-semiconductor interactions. Optimal conditions balance these factors to maximize light absorption, electron transfer, and energy conversion efficiency while minimizing losses due to decomposition or inefficient dye binding. These findings emphasize the importance of precise control over polymerization parameters in designing high-performance DSSCs for practical applications.

Molecular weight of Poly-tannic acid

In the proposed mechanism, the phenolic –OH group of tannic acid attacks the epoxy ring of TMPGDE, forming an ether bond (figure 6). As a result, tannic acid molecules cross-link with TMPGDE, producing poly tannic acid with longer conjugation and better stability. Polymerization of tannic acid with TMPGDE produces a three-dimensional network polymer because TMPGDE, as a crosslinker, opens the epoxide ring and binds to the hydroxyl groups of tannic acid. The result is a hydrophilic polymer with many free hydroxyl groups that form ether bridges between tannic acid units. This structure creates a gel or amorphous solid that has the ability to absorb metal ions or water, depending on the polymerization conditions and reactant ratio.

The average molecular weight of the polymer in the poly-tannic acid solution with the highest efficiency was determined using its intrinsic viscosity, which was measured with an Oswald viscometer.

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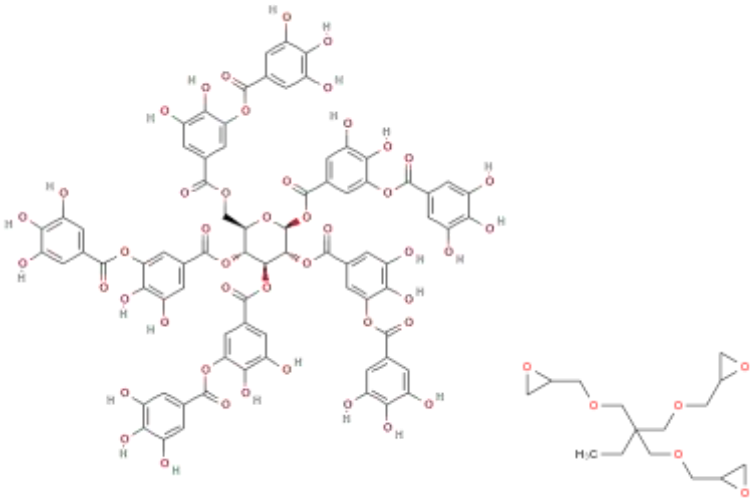


Figure 6. Structure of tannic acid (left) and TMPGDE (right).

In this process, the solid poly-tannic acid was dissolved in a polar solvent (water), and the viscosity of the resulting solution was measured. The Oswald viscometer operates on the principle of timing how long it takes for a liquid to flow through a specific distance under gravity. By comparing the flow time of pure water (solvent) with that of the poly-tannic acid solution, the specific viscosity of the polymer solution can be calculated.

The intrinsic viscosity, which represents the contribution of the polymer to the overall viscosity of the solution, was then determined using the Huggins equation. This equation relates specific viscosity to polymer concentration and provides a measure of how effectively polymer chains interact with the solvent. Once intrinsic viscosity is known, it can be used to calculate the average molecular weight of the polymer using the Mark-Houwink equation.

The degree of polymerization (DP), which indicates how many monomer units are linked in a polymer chain, was also calculated based on the molecular weight of poly-tannic acid. This provides insight into how effectively tannic acid molecules bind during polymerization. According to experimental results, when optimizing for cell efficiency, an average molecular weight of 32,610,156.8 g/mol and a degree of polymerization of 19,182 were achieved.

The relationship between polymer chain length and DSSC performance can be explained by considering the two main roles of polymerized tannic acid on the TiO_2 surface. The higher the molecular weight and degree of polymerization, the more phenolic groups can attach, resulting in stronger interactions between the dye and semiconductor and more optimal light absorption due to conjugation elongation [72]. This is consistent with the increase in photocurrent density at moderate polymer chain lengths. However, excessively long polymer chains can cause steric hindrance and interfere with charge transport, thereby increasing recombination losses [73,74]. Thus, there is an optimal degree of polymerization that balances light absorption, the amount of dye on the surface, and electron transport, thereby maximizing device efficiency.

These values confirm that the optimized polymer possesses sufficient chain length to enhance conjugation and binding to TiO_2, thereby contributing to the observed increase in DSSC efficiency. These findings are significant because they reveal how molecular weight and degree of polymerization influence DSSC performance. Higher molecular weights typically indicate longer polymer chains with

better light absorption properties and stronger binding to TiO_2, leading to improved electron transfer efficiency in DSSCs. The analysis underscores the importance of controlling polymerization conditions to optimize both molecular weight and cell efficiency for practical applications in solar energy conversion.

Conclusions

This study demonstrated that tannic acid polymerization significantly improved the efficiency of dye-sensitized solar cells (DSSCs). The highest efficiency achieved was 8.984% for poly-tannic acid, far exceeding the 1.35% achieved by unpolymerized tannic acid. The optimal polymerization conditions were obtained at a monomer concentration of 2.5 grams and a temperature of 50°C. UV-Vis characterization showed a bathochromic shift and a widening of the light absorption range of poly-tannic acid, indicating an increase in the light absorption ability in the range of 200-800 nm. FTIR analysis confirmed the success of the polymerization with the formation of new ether groups. The average molecular weight of the optimal poly-tannic acid was 32,610.1568 g/mol with a degree of polymerization of 19.182. The polymerization temperature and monomer concentration had significant effects on the polymer yield and cell efficiency, with the optimal temperature being around 50-70°C depending on the monomer concentration. Temperatures above 100°C cause decomposition of tannic acid and decrease in cell efficiency. This study demonstrates the potential of tannic acid polymerization as an effective strategy to improve the performance of organic DSSCs, paving the way for the development of more efficient and cost-effective solar cells.

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Declaration of Generative AI and AI-Assisted Technologies

The authors declare that no generative AI or AI-assisted technologies were used in the preparation of this manuscript.

Author Contributions

D.A.: Methodology, investigation, data collection, formal analysis, data curation, visualization, writing —original draft preparation. H.: Methodology, investigation, data collection. B.O.: Methodology, validation, supervision, and writing —review and editing. P.P.: Conceptualization, supervision, corresponding author responsibilities.

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Declaration of Conflicting Interests

The authors declare no conflict of interest.

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