



## Research Article

# Synthesis of compounds containing Cyano Anchor Groups, production of dye-sensitive solar cells and investigation of the antibacterial properties of these compounds

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## ABSTRACT

Global warming has become one of the most important factors threatening the world. The first place among the causes of global warming is taken by fossil fuels, which are mainly used for energy needs. The intensive consumption of fossil fuels causes the decrease of the world's fossil fuel reserves, environmental pollution and the greenhouse effect. Various renewable energy sources are shown as alternatives as a solution to these problems. Among all renewable energy forms, solar energy is the most popular. In this article, a dye-sensitive solar cell device of two compounds having a cyano anchor group containing fluorene and triphenyl amine as donor groups was prepared, and their antibacterial properties were investigated. The photovoltaic and electrochemical properties of these materials were evaluated, and it was investigated whether they could be used as photosensitizers in dye-sensitive solar cells. Photovoltaic cell efficiencies of the devices were in the range of 0.13-0.04% under 100 mW/cm<sup>2</sup> simulated AM 1.5 solar irradiance, and the highest open circuit voltage reached 0.45 V. When the photovoltaic cell efficiency values of the produced dye-sensitive solar cell devices were examined, it was seen that the L1 compound was higher than L2 due to the effect of the donor group. The antimicrobial properties of L1 and L2 compounds were examined, and the effect of donor groups was investigated. The conventional disk diffusion method was used to evaluate the antibacterial activity of the synthesized compounds against *Escherichia coli* and *Staphylococcus aureus*. As a result, it was seen that the produced compounds showed antibacterial activity, and the compound with high photovoltaic cell efficiency value had a high antibacterial properties.

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## INTRODUCTION

Photovoltaic technology is known to be one of the most important techniques aiming to produce clean energy from

sunlight [1]. Due to their low cost and simple fabrication methods, dye-sensitized solar cells (DSSCs), which convert sunlight into electricity, have been the subject of intense

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research over the past several decades as a novel system in the field of photovoltaics. Other known advantages of DSSCs are flexibility, chemical versatility, colorful appearances, and the possibility of synthesizing a wide range of different organic and organometallic molecular structures [2-4].

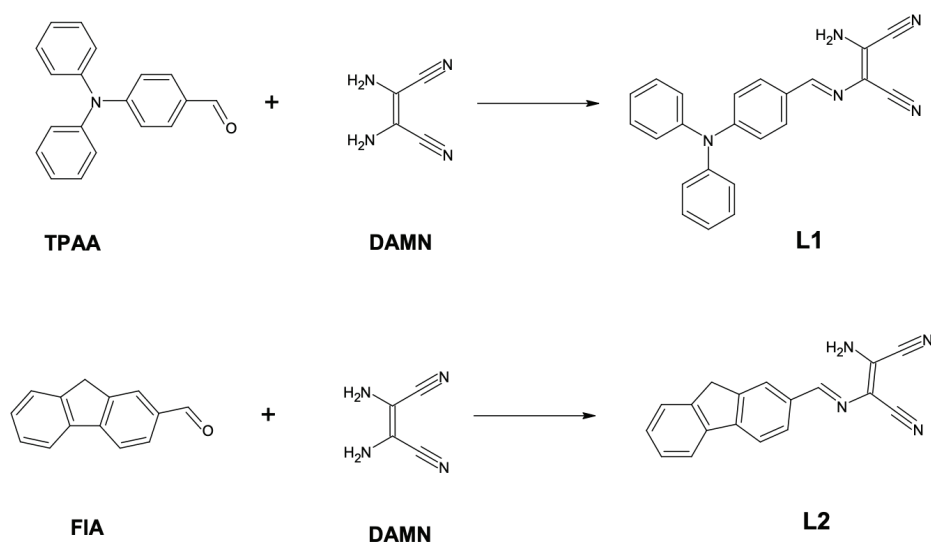
The device's photovoltaic and photocurrent levels affect how well solar energy is converted to electricity. Therefore, to enhance the photocurrent, the sensitizer's solar absorption spectra must be expanded from the visible to the near-infrared range, and its molar extinction coefficient must be raised. [5, 6].

In this case, novel chemical dyes with a red-shifted absorption band and increased molar absorptivity were created by attaching very prolonged conjugation units, such as triphenylamine and benzene, to the diaminomaleonitrile ligands. Within the organic photoelectric area, such as in organic light-emitting diodes (OLEDs) [7], organic field effect transistors (OFETs) [8], and perovskite solar cells (PSCs) [9], triphenylamine (TPA) and its derivatives, hole-transporting and emitting materials, are commonly utilized. Recent research revealed that because of its benefits, such as low excitation energy, less light damage, strong penetrability, etc., it has a promising future in the domains of biological probes and drug targeting [10]. Fluorene and its derivatives have been widely and successfully used in a variety of synthetic applications in recent years. For example, they are used in medical and optical applications. Additionally, fluorene derivatives show impressive spectroscopic and photophysical properties. Fluorine-based fluorophores are used significantly to improve dye absorption properties and fluorophore sensors [11, 12]. It is extensively employed in the production of several heterocyclic compounds.

2,3-Diaminomaleonitrile (DAMN), which is extensively employed in the production of several heterocyclic compounds, is a tetramer of hydrogen cyanide. DAMN derivatives have potential applications for the synthesis of heterocyclic compounds as well as the electron donor and/or due to the effects of the impact on the spectroscopic electron acceptor substituents, a wide variety of organic  $\pi$ -conjugated DAMN-based compounds and these compounds attention on the physicochemical properties, attractive and intense studies are in progress [13]. Because of their versatility in synthesis, mono-imitated DAMN-based compounds are also employed as intermediates in the synthesis of non-symmetrical ligands, pigments, and materials containing high nitrogen content [13,14]. It is commonly known that DAMN-based Schiff base is produced when DAMN reacts with aromatic aldehydes [15].

Azomethine groups ( $-RC=N-$ ) are present in Schiff bases structurally, and they are created when amines and active carbonyl compounds condense. Schiff bases play a significant role as synthetic intermediates and are utilized in the thermostable optical material business, chemical sensors, aggregation-enhanced emission luminogen, jet-printing inks, and optically active materials [16]. Numerous scientists worldwide are studying the substantial anti-tumor, antibacterial, and antifungal action that various Schiff bases exhibit. According to reports, the nitrogen atom in the azomethine group may contribute to the hydrogen bond formation with the active centers of cell components, thereby interfering with regular cell functions [17].

In our study, we describe the synthesis and dye-sensitized solar cell performance and antimicrobial activity of organic electronic materials with D- $\pi$ -A properties. We synthesized two dyes containing donor fluorene and triphenyl amine and containing a CN anchor group. It was determined whether these chemicals may be used as



**Scheme 1.** Synthesis scheme of L1 and L2 compounds.

photosensitizers in DSSCs by looking at their photovoltaic characteristics. The analysis results show that the L1 sensitizer used in the device has a strong donor group effect and increases the power conversion efficiency. When the antibacterial activities of these substances were investigated, it was determined that the synthesized substances showed the presence of antibacterial activity. Since it is a donor substance and has a high electron density, the highest antibacterial activity was observed in L1 substance.

## MATERIALS AND METHODS

### Materials

All of the chemicals were Fluka and Merck reagent grade. Unless specified otherwise, commercially available reagents were used without further purification, and solvents in processes were distilled from suitable drying agents before use. For column chromatography, Merck's silica gel 60 (0.04–0.063 mm) was used. The most important aspects of *S. aureus* and *E. coli* bacteria in the intestinal flora, which are quite common in nature and are found in dust, soil, on goods, human and animal skin, nasal mucosa, mouth, and nasopharynx flora, are chemotherapeutic. The rapid resistance too many of the substances, and therefore infections, are more common. *E. coli* is Gram negative; *S. aureus* is from Gram-positive bacteria group. The use of these two bacteria was deemed appropriate in our experimental study, since they are examples of both species and because they are widely used in antimicrobial studies in the literature. *E. coli* (ATCC25922) and *S. aureus* (ATCC 29213) strains were obtained from Yıldız Technical University, Faculty of Arts and Sciences, Chemistry Department, Biochemistry Laboratory Culture Collection.

## SYNTHESIS

### Synthesis of 2-amino-3-[(E)-{4-(diphenylamino)phenyl}methylidene]amino]-3-aminomaleonitrile (L1) [18]

To the solution of 2,3-diaminomaleonitrile (0.1 g, 0.462 mmol) dissolved in 10 mL methanol, slowly add triphenylamine aldehyde (0.250 g, 0.462 mmol) dissolved in 10 mL methanol. The mixture is refluxed for 36 hours and then cooled at room temperature until a light yellow precipitate forms. The product is filtered, washed several times with cold methanol and acetonitrile and dried at ambient temperature.  $C_{23}H_{17}N_5$  (L1), Yield: 0.24 g (72%). m.p. = 198 °C

### Synthesis of 2-(((E)-(9H-fluoren-2-yl)methylene)amino)-3-aminomaleonitrile (L2) [19]

To the solution of 2,3-diaminomaleonitrile (0.139 g., 1.28 mmol) dissolved in 10 mL methanol, slowly add fluorene 2-aldehyde (0.250 g, 1.28 mmol) dissolved in 10 mL methanol. The mixture is refluxed for 24 hours and then cooled at room temperature until a light yellow precipitate forms. The product is filtered, washed several times with cold methanol and acetonitrile and dried at ambient temperature.  $C_{18}H_{12}N_4$  (L2), Yield: 0.19 g (53%). m.p. = 195 °C

## RESULTS AND DISCUSSION

### Synthesis and Characterization

As shown in Scheme 1, L1 and L2 dyes were synthesized in good yields by the formation of Schiff base reactions. In the L1 and L2 FTIR spectra, the characteristic stretching bands of the amine ( $NH_2$ ) group are seen at 3480–3350  $cm^{-1}$ , and the stretching bands of the imine ( $C=N$ ) group are seen at 1619 and 1590  $cm^{-1}$ , respectively. These strong bands, which are indicative of the azomethine moiety of the majority of Schiff base compounds, are attributed to the

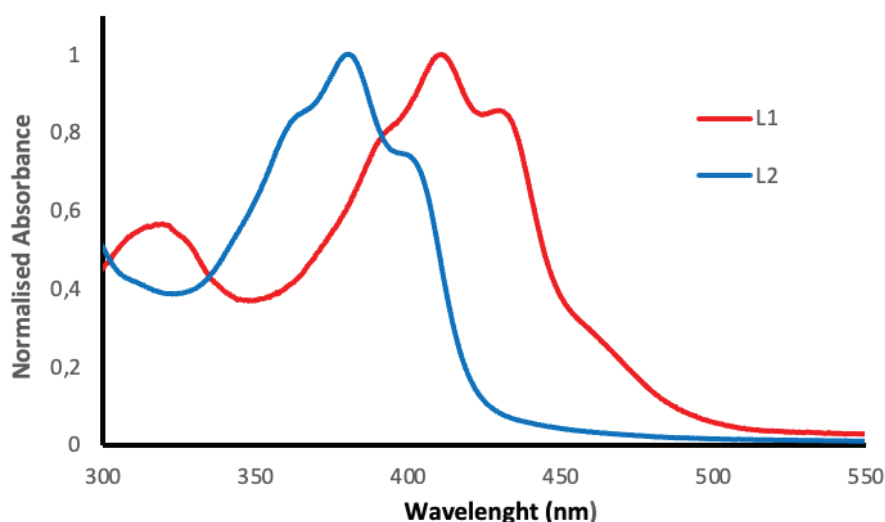


Figure 1. UV-vis spectra of L1 and L2 compounds in methanol.

ligands' C=N stretching frequencies. In the infrared spectra of L1 and L2, the absence of stretching vibrations at  $1686\text{ cm}^{-1}$  belonging to the carbonyl group (C=O) in the starting compound TPA and FIA structure indicates the formation of the Schiff base.

UV-vis absorption spectra of L1 and L2 compounds were taken by preparing  $1 \times 10^{-4}\text{ M}$  in methanol. L1 and L2 showed two distinct bands that were different from the starting materials but similar to each other. In the absorption spectra of L1 and L2, the  $\pi\text{-}\pi^*$  transitions with the highest energy levels were observed at peaks at 411 and 381 nm, respectively, as seen in Figure 1. Absorption peaks of strong charge transfer (CT) were not observed in the UV-vis spectrum of both substances.

### Production and Characterization of DSSCs

In order to fabricate DSSCs, two transparent  $\text{TiO}_2$  pastes were applied, as previously reported [20, 21], on the conductive side of the fluorine-doped tin oxide (FTO) substrate using Doctor Blade's method. The coated FTO substrates were immersed in an EtOH solution including 0.3 mM dye solution to create the dye-sensitized (L1 and L2)  $\text{TiO}_2$  films (photoanodes). Using a platinum paste solution, the Pt counter electrodes were created on the FTO substrates. Surlyn film was used to seal the counter electrode and photoanode. Through a bored hole at the counter electrode, the redox electrolyte was injected between the electrodes. The current density-voltage (J-V) characteristics were investigated by using the AM 1.5 global one sun illumination ( $100\text{ mW cm}^{-2}$ ) from a solar simulator.

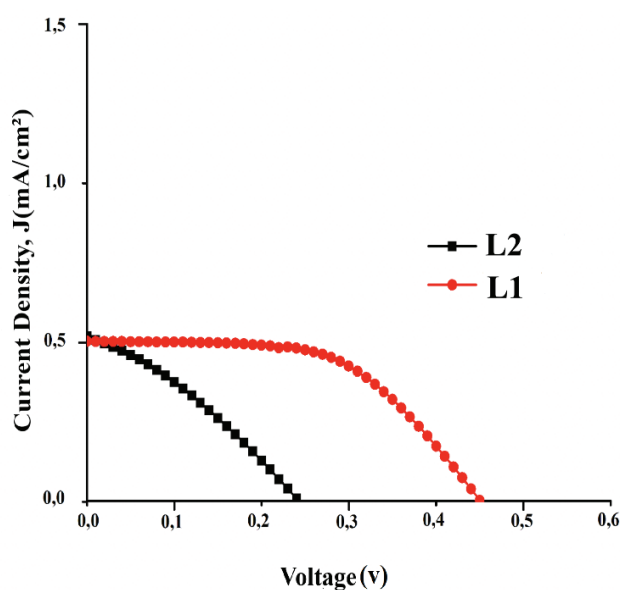
The electrical performance of a solar cell is determined by the short-circuit current ( $I_{sc}$ ), open-circuit voltage ( $V_{oc}$ ), current at the maximum power point ( $I_{mp}$ ), voltage at the maximum power point ( $V_{mp}$ ), maximum power ( $P_{max}$ ), fill factor (FF), and photovoltaic cell efficiency ( $\eta$ ). The solar cell efficiency and fill factor are calculated using the Eq. 1 and Eq. 2 equations, respectively [21].

$$\eta = \frac{P_{max}}{P_{in}} = \frac{I_{sc} \cdot V_{oc} \cdot FF}{P_{in}} \quad (1)$$

where the FF is most commonly determined from measurement of the I-V curve, is given by

$$FF = \frac{I_{mp} \cdot V_{mp}}{I_{mp} \cdot V_{oc}} \quad (2)$$

The DSSC parameters:  $J_{sc}$ ,  $V_{oc}$ , and FF were extracted from the J-V curve to find the cell efficiency as illustrated



**Figure 2.** Current density-voltage curves with sensitizers L1 and L2 of illuminated DSSCs.

in the Fig. 2. Furthermore, the effective area of the devices in our DSSC device was  $0.52\text{ cm}^2$ , which is greater than that of the reported.

A clear association between the sensitizer's structure and the device's power conversion efficiency (PCE) can be seen in the current-voltage curves that were obtained in both light and dark conditions. The high values of the L1-based DSSC devices  $J_{sc}$ ,  $V_{oc}$ , FF, and PCE from the generated devices contributed to a higher PCE value.

The higher PCE value obtained from the device fabricated with L1 sensitizer is a result of the TFA donor group having higher electron density compared to other L2 sensitizer derivatives and more electrons are presented to the  $\text{TiO}_2$  surface. The values related to the results of the optimized DSSC device are presented in Table 1. The larger molar extinction coefficient and wider absorption in the visible region of L1 compound resulted in higher  $J_{sc}$  values compared to L2 compound. Weak bonding and electron transfer on semiconductor  $\text{TiO}_2$  as well as the presence of anchor CN groups in the compound structures, are the causes of the low PCE value that was achieved. Moreover, the poor performance of L1-based DSSC might have been caused by electron recombination due to the aggregation of its molecules on the  $\text{TiO}_2$  surface.

**Table 1.** Photovoltaic parameters for DSSCs sensitized with L1 and L2

Sample	$J_{sc}$ (mA/cm <sup>2</sup> )	$V_{oc}$ (V)	FF	$\eta$ (%)
L1	0,5	0,45	0,59	0,13
L2	0,5	0,25	0,32	0,04

There are few articles in the literature where the cyano group is used alone as an anchor/acceptor. In the sensitizer structure, the cyano group is generally located together with the carboxylic acid anchor group, and in this case, the life of the prepared devices is short due to the hydrolysis of the carboxylic acid. The L1 and L2 sensitizers used in the DSSC device were evaluated with the CN anchor/acceptor groups found in the literature. As a result, it was observed that higher Voc values were obtained in the produced devices, but lower Jsc and FF values were achieved. Low Jsc and FF values cause the power conversion efficiency of our device to decrease. Direct electron transfer from the donor group to the anchor group in the sensitizing structure leads to an increase in the performance of sensitized solar cells [21]. The existence of the  $\pi$ -conjugation system that provides the transfer of these electrons, as well as the electron density that will form from the donor group to the anchor group, is an important issue in terms of the performance of solar cells [22].

The functionalized dye (JY1) was synthesized by J. Cong et al. and included an electron-withdrawing nitro group along with CN and COOH anchor groups. The dye's power conversion efficiency was found to be  $\eta = 0.81$  [23]. In a separate investigation, Badawy et al. synthesized two cyanoacetamide and four novel phenylacetone nitrile dye derivatives and examined the effects of a thiophene spacer and several supplementary acceptors on the photophysical and electrochemical properties of the DSSC devices. Additionally, they assessed the DSSCs' total photo conversion efficiencies and photovoltaic characteristics for the sensitizers SFA5 and SFA6, obtaining the highest result reported in the literature [24]. In our next studies, we will synthesize new sensitizers by changing the acceptor and anchor groups in the structure to increase the efficiency of our dyes.

In the study conducted by S. M. Abdalradi et al., three new 2,3-diaminomaleonitrile (DAMN) derivative dyes were prepared by simple Schiff base reaction in a one-pot reaction. The compounds were designed as a sensitizer in dye-sensitized solar cells (DSSCs) and were used in DSSCs. The data was found to show the best performance for SA3 dye with 0.38% efficiency at AM 1.5, followed by SA2 with 0.22% and the last dye was SA1 with 0.09%, 5.4% compared to the control cell (N719) [25].

#### Antimicrobial tests

*E. coli* (ATCC25922) and *S. aureus* (ATCC25923) were used as reference strains for Gram-negative and Gram-positive bacteria, respectively. Strains were grown on nutrient agar. The synthesized samples (L1 and L2) were tested for antibacterial activity by the standard disc diffusion method. Each bacterium was added to 100  $\mu$ L of inoculum agar medium made by culturing for 18 to 24 hours and adjusting the turbidity to meet the 0.5-McFarland criterion. The *E. coli* broth culture was then inoculated onto nutrient agar plates for 24 hours. Sterile cotton swabs were used to swab each strain uniformly onto the separate plates.

**Table 2.** Antibacterial activity parameters of L1 and L2 samples

Bacteria	Inhibition zone (mm)	
	L1	L2
<i>Echerichia coli</i>	15	13
<i>Staphylococcus aureus</i>	19	17

Gel puncture was used to create 10 mm-diameter wells on nutrient agar plates. Ethanol was used as the solvent and 50  $\mu$ L of sample solutions (1 mg/mL) were applied to each well in all plates with pipettes. Subsequently, the plates were incubated for 24 hours at 37 °C. The zone incubation of each well was measured in millimeters (mm), and the results were recorded.

When looking at the inhibition zone formed by sample L1, one of the Schiff bases used in the study, it was seen that the antibacterial effect was the highest. Alsantali et al. A new series of (4-(2,7-dichloro-9H-fluoren-4-yl)thiazol-yl) acetamide derivatives were synthesized, characterized, and evaluated for their antimicrobial activity. Additionally, the synthesized fluorene derivatives were found to show remarkable activity as antibacterial and antifungal agents when evaluated as antimicrobial agents. Although there is limited literature, these studies can be considered to open a new alternative field in the field of medicinal chemistry of fluorene derivatives [26].

By synthesizing new mono-, di- and tri-phosphonium ionic liquids and evaluating their antibacterial activity on Gram-positive and Gram-negative bacteria, Brunel et al. found that triphenylamine-phosphonium ionic liquid derivatives have a unique mechanism of action. They observed that although TPA-P compounds with structures with different chains, spacer lengths and charges were active on Gram-positive bacteria, this distinction was in favor of Gram-negative bacteria as the spacer length, and charge density increased and proved that TPA-P derivatives could constitute a useful biological tool [26].

#### CONCLUSION

In this study, compounds containing fluorene and triphenyl amine as donor groups and cyano as anchor groups were synthesized and used in the production of dye-sensitive solar cell devices. Photovoltaic cell efficiencies of the devices were in the range of 0.13-0.04% under 100 mW/cm<sup>2</sup> simulated AM 1.5 solar irradiance, and the highest open circuit voltage reached 0.45 V. It was observed that the L2<L1 efficiency of the produced dye-sensitive solar cell devices increased, respectively. The effective photovoltaic performance of sensitizer L1 in dye-sensitive solar cell devices appears to have a donor triphenylamine group in their structure and strongly present electrons to the TiO<sub>2</sub>

surface. Our best result in this study was achieved with an L1-based dye-sensitive solar cell that exhibited a power conversion efficiency of up to 0.13%. It was observed that the antibacterial effect in the inhibition zone formed by the compounds L1 and L2 was the highest in the L1 compound and can be used in applications. Antibacterial activity and power return efficiency values of compounds L1 and L2 were measured and it was seen that compound L1 had higher values in both studies. We hope that in future studies this relationship will be used in some medical and biological applications.

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## AUTHORSHIP CONTRIBUTIONS

Authors equally contributed to this work.

## DATA AVAILABILITY STATEMENT

The authors confirm that the data that supports the findings of this study are available within the article. Raw data that support the finding of this study are available from the corresponding author, upon reasonable request.

## CONFLICT OF INTEREST

The author declared no potential conflicts of interest with respect to the research, authorship, and/or publication of this article.

## ETHICS

There are no ethical issues with the publication of this manuscript.

## STATEMENT ON THE USE OF ARTIFICIAL INTELLIGENCE

Artificial intelligence was not used in the preparation of the article.

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