SPEKTRA: Jurnal Fisika dan Aplikasinya
Volume 10, Issue 3, December 2025
Homepage: doi.org/10.21009/SPEKTRA

DOI: doi.org/10.21009/SPEKTRA.103.04

Comparative Study of the Incorporation of Gold and Silver Nanoparticles into N-719 Dye on the Performance of Dye-Sensitized Solar Cell (DSSC)

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Received: 18 August 2025
Revised: 17 December 2025
Accepted: 19 December 2025
Online: 27 December 2025
Published: 30 December 2025

SPEKTRA: Jurnal Fisika dan Aplikasinya
p-ISSN: 2541-3384
e-ISSN: 2541-3392

ABSTRACT

The Dye-Sensitized Solar Cell (DSSC) is a promising third-generation solar cell technology with very significant enhancement in device efficiency. The modification in the fabrication of DSSC is still being studied today in order to increase the stability and lifetime of the devices. The aims of our research is to compare the incorporation of gold and silver nanoparticles capped with 3-Mercaptopropionic Acid (AuMPA or AgMPA) into N-719 dye to enhance DSSC performance. Metal nanoparticles exhibit a unique optical property called Localized Surface Plasmon Resonance (LSPR), which maximizes light absorption. In our experiment, AuMPA and AgMPA were prepared using the chemical reduction method and then incorporated into the dye solution and then in the DSSC assemble, the electrode-TiO_2 layer were immersed into dye contained NPs solution. Optical characterization, including absorbance and photoluminescence spectra, shows a blue shift in the spectral position and an overlap between the intrinsic peak of N-719 and the plasmonic peak of AuMPA or AgMPA, indicating a stable nanoparticle dispersion within the dye matrix. Fourier Transform Infrared (FTIR) spectra confirm the chemical interaction through the changes in the S-H and -COOH vibrations. Scanning Electron Microscopy (SEM) observation reveals the presence and growth of AuMPA and AgMPA nanoparticles within the mesoporous TiO_2 layer. The addition of AuMPA into the dye solution increases the PCE of the device from 3.32% to 4.19%, while AgMPA yielded a PCE of 3.76%. Enhancement of our DSSC performance is attributed to the LSPR effect, which enhances light absorption and charge transport. The better performance by the addition of AuMPA into the dye is attributed to a highly substantial plasmonic effect and stability, while AgMPA tends to aggregate, limiting its effectiveness.

Keywords: dye-sensitized solar cell, gold nanoparticles, silver nanoparticles, dye N-719, plasmonic effect

INTRODUCTION

The majority of the current energy supply remains heavily dependent on non-renewable fossil fuels such as petroleum and coal. As a sustainable alternative, solar energy presents significant potential due to its abundant availability and environmentally benign characteristics [1-2]. Solar cells utilize the photovoltaic effect to convert solar energy into electrical energy [3]. Various types of solar cells continue to be developed, ranging from silicon-based solar cells to Dye-Sensitized Solar Cells (DSSC), which are third-generation solar cells [4]. DSSC is a type of solar cell that has attracted significant attention because it is cheap, ease of manufacture, low toxicity profile, environmentally friendly, effectiveness, and ability to operate in low-light conditions [3, 5-6]. Nonetheless, theoretically, the DSSC can achieve an efficiency of 32%, but the highest efficiency reported today is only about 13%. Therefore, the research and development process regarding DSSC continues to be carried out [7-8].

In other research, it has been established that Gold (Au) and silver (Ag) are two types of metal nanoparticles commonly used because they exhibit unique optical properties known as Localized Surface Plasmon Resonance (LSPR) when incoming light interacts with surface electrons in the conduction band. LSPR arises when light waves are trapped inside conductive nanoparticles that are smaller in size than their wavelength, creating resonance conditions. This resonance occurs when the frequency of incoming light is proportional to the frequency of the collective oscillation of electrons in nanometer-scale structures [9].

Several approaches can be taken to improve the work efficiency of DSSC, one of which is the addition of nanoparticles to the active layer components of DSSC. The addition of metal nanoparticles to the DSSC structure plays a crucial role in maximizing light absorption by harnessing the plasmonic effects of nanoparticles. This effect can be activated through various applications on the active layer of DSSC, one of which involves the photosensitizer component [10]. Photosensitizers are one of the important components in DSSC because they play a role in absorbing sunlight [11]. Dye N-719 is a photosensitizer that is popularly used in DSSC because of its ability to absorb light in the region of visible light, and has unique characteristics such as ultra-fast electron injection and its ability to bind to the surface of TiO_2 [10].

Several studies have been conducted to investigate the effect of adding metal nanoparticles to DSSC devices, including research by Pujiarti et al., which demonstrated an increase in power conversion efficiency (PCE) from 4.18% to 5.14% in DSSC devices after the addition of AuNP capped with oleylamine to the dye N-719. In contrast, Rahayu et. al. found that the addition of 5.66 wt.% AuNP by capping dodecanethiol increased PCE from 1.58% to 2.77%, and a similar trend also occurred in the addition of silver nanoparticles [9, 11]. The capping agents play an important role in preventing the agglomeration of colloidal particles by surrounding the particles. The use of different capping agents cannot only control the material's particle size, agglomeration, and morphology but also stability over time of nanostructures [12].

Based on previous research, this research aims to incorporate metal nanoparticles, specifically gold nanoparticles and silver nanoparticles capped with Mercaptopropionic Acid (3-MPA) (AuMPA and AgMPA), into the DSSC photosensitizer component, namely dye N-719. 3-MPA is one of the capping molecules commonly used for surface functionalization of gold or silver cores due to the presence of both thiol and carboxyl functional groups [13-14]. We compare the types of AuMPA and AgMPA regarding their interaction with that of the dye to determine which type yields better performance in the fabricated devices.

METHODS

1. Synthesis of AuMPA and AgMPA

AuMPA and AgMPA were synthesized by used of modified chemical reduction method. For the synthesis of AuMPA, it was carried out by dissolving $HAuCl_4$ at a concentration of 0.0417 M in water in a three-neck flask. The solution was heated to boiling temperature under reflux conditions and stirring constantly. Separately, the encapsulation solution was made by mixing 1.5 wt% of 3-MPA and trisodium citrate dihydrate and then injected to $HAuCl_4$ solution. After mixing, the colour of the solution changes from yellow to red wine, indicating the formation of gold nanoparticles. The heating and stirring process was continued for another hour, and the solution was then allowed to cool to room temperature.

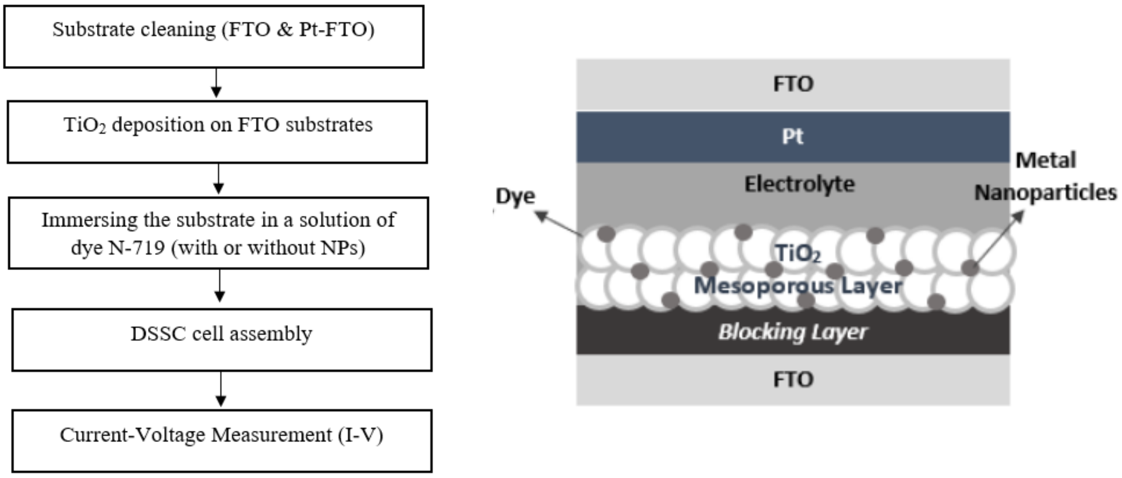
The synthesis of AgMPA was carried out by dissolving $AgNO_3$ (1.5 mM) in water and heating in a three-neck flask. After the solution reached the boiling temperature, a mixture of 1.5 wt% 3-MPA and trisodium citrate dihydrate was added. The heating and stirring process was continued until the color solution changes from clear to a brownish-yellow, indicating the formation of silver nanoparticles. The solution was then cooled using an ice bath.

2. Preparation of N-719 dye solution incorporated with AuMPA or AgMPA

In our experiment, we mixed the active material of dye N-719 with that of AuMPA or AgMPA in order to increase the absorbance of photon and to enhance the number of charges reveal in the device. The preparation of dye N-719 was carried out by mixing 0.825 mg/mL of N-719 *(Ruthenizer 533-bis TBA*) with CDCA (*Chenodeoxycholic Acid*) in ethanol by following the method and concentration of our previous study [9, 11]. The mixture was then stirred slowly with magnetic stirrer in room temperature until the solution is dissolved and homogeneous. The addition of AuMPA (0.046 mg/mL) and AgMPA (0.016 mg/mL) into the dye solution based on optimum result in our previous experiment. The mixture of dye solution and metal NPs form in homogenous solution without any aggregation exist in bottom solution [9, 11, 14]

3. Fabrication of DSSC with and without incorporation of AuMPA or AgMPA

Fabrication begins by cleaning FTO and Pt-FTO substrates with the size of (1.25×2.50) cm. Next, the FTO substrate was coated with TiO_2 mesophorous by used of screen-printing technique in the area of 0.5×0.5 cm. In our experiment, the thickness of this TiO2 mesophorous layer is around 200 nm. After the sintering process, the substrate was immersed in an N-719 dye solution for 12 hours, either with or without the addition of nanoparticles. The fabrication process is systematically explained in FIGURE 1.



(a) (b)

FIGURE 1. (a) Flowchart of DSSC device fabrication, and (b) DSSC configuration without and with the addition of metal nanoparticles [9].

The performance of the DSSC was evaluated through current-voltage (I-V) measurements using a solar simulator with an intensity of 100 mW/cm², under both dark and standard illumination conditions.

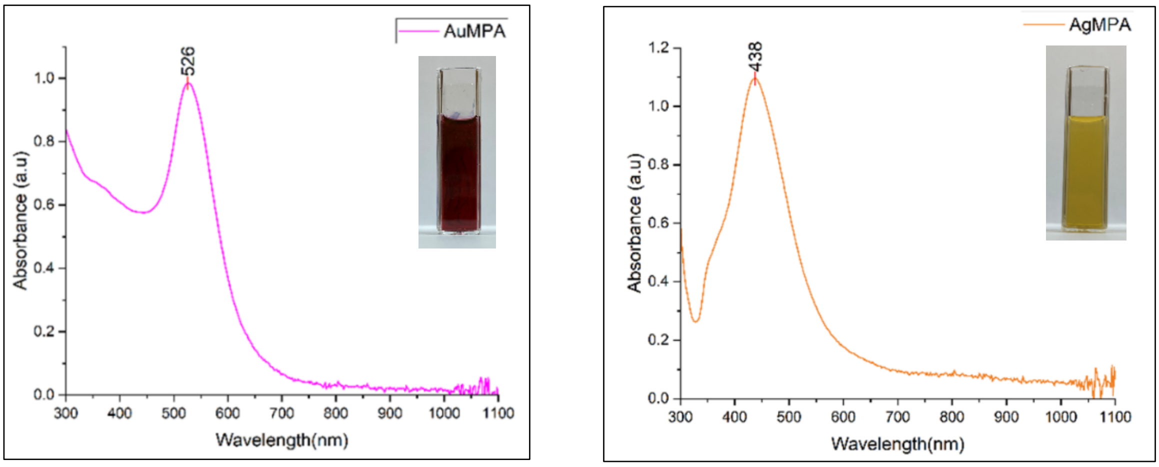
4. Characterizations

The characterization in this study comprises UV-Vis and Photoluminescence spectroscopy for optical analysis, Fourier Transform Infrared Spectroscopy (FTIR) spectroscopy for the identification of functional groups, and Scanning Electron Microscopy (SEM) to observe surface morphology.

RESULTS AND DISCUSSIONS

1. Optical properties of coloidal AuMPA and AgMPA

The formation of AuMPA and AgMPA can be identified by the appearance of specific absorbance peaks called plasmonic peaks as shown in FIGURE 2. The plasmonic peak of AuMPA is at a wavelength of 526 nm with a red-wine solution colour that is characteristic of colloidal AuNPs, while for AgMPA, the plasmonic peak is at a wavelength of 438 nm with a brownish-yellow solution color [15-16].

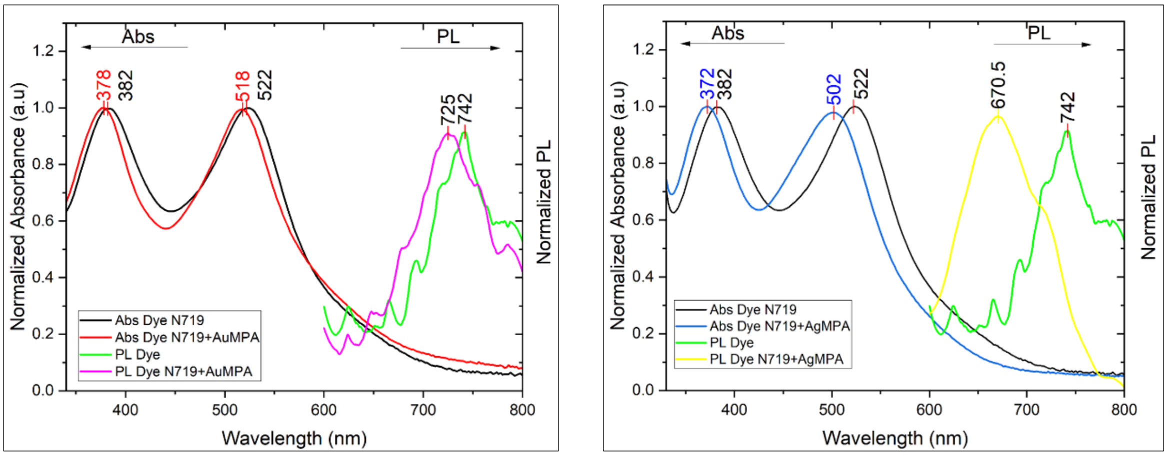


(a) (b)

FIGURE 2. Absorbance Spectrum of (a) AuMPA and (b) AgMPA solution

2. Optical and chemical properties of N-719 dye with incorporation of AuMPA or AgMPA

FIGURE 3 present a comparison of the optical characteristics between the reference dye and the dye to which AuMPA or AgMPA has been added based on the absorbance and photoluminescence spectra. The absorbance spectrum of N-719 dye consists of two intense (broad) bands with peaks at 382 nm (violet-blue regions) and 522 nm (green region) [17]. These peaks attribute to the Metal-to Ligand Charge Transfer (MLCT) transition [14]. The dominant absorbance area of N-719 dye is found to overlap with that of the plasmonic peak of AuMPA or AgMPA, which reveals a blue shift of about ~4 nm for dye with the addition of AuMPA and ~20 nm for dye with the addition of AgMPA. This overlapping phenomenon indicates that the metal nanoparticles are successfully dispersed and stable within the dye matrix, without undergoing significant aggregation that can interfere with the dye absorption profile.



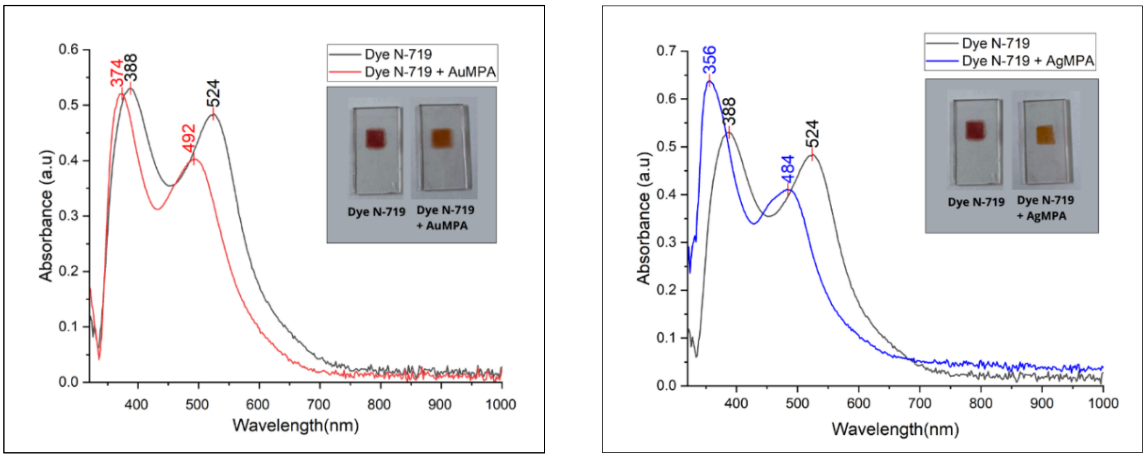
(a) (b)

FIGURE 3. Dye absorbance and photoluminescence spectra with the addition of (a) AuMPA and (b) AgMP.

These results are also supported by the findings of photoluminescence (PL) spectroscopy, which reveals a shift in the luminescence peak towards shorter wavelengths (blue shift) of the dye when AuMPA or AgMPA is added. This shift indicates a chemical interaction between the dye and capping material of metal nanoparticles, which can modify the electronic and optical properties of the dye and can be attributed to the recombination rate of photo-induced charge carriers [17]. This modification can be applied to optoelectronic devices based on the N-719 dye as the active layer of DSSC.

The performance efficiency of DSSC is determined by the dye's capacity to absorb light and by the diffusion of electron injection across the TiO2 mesoporous layer [18]. UV-Vis spectroscopy measurement on mesoporous TiO2 are performed to illustrate the actual conditions when the dye adheres to the surface of the semiconductor within the DSSC, as the efficiency of electron injection into the TiO2 conduction band is highly dependent on the absorbed dye bond as well as the electrostatic and chemical interactions between the surface of the TiO2 and the dye molecules [19].

From the UV-Vis spectra of the thin film specimens in FIGURE 4, it is evident that the metal nanoparticles (AuMPA and AgMPA) do not interfere with the main spectral characteristics of the N-719 dye; however, they have a significant effect on the absorption spectra [20]. The addition of AuMPA or AgMPA nanoparticles causes a shift in the absorption peak position of the immersed mesoporous TiO2 layer spectrum on the N-719 dye without the addition of nanoparticles. This shift occurs towards a short wavelength (blue shift) due to the overlap between the spectrum of N-719 dye and the plasmonic spectrum of AuMPA or AgMPA metal nanoparticles. The observed enhancement in the optical properties of dye molecules indicates that the dipole moments of the dye and the LSPR are within a minimal spatial range [21].



(a) (b)

FIGURE 4. Absorbance spectra of dye N-719 and dye N-719 with: (a) AuMPA and (b) AgMPA, on thin film TiO_2 mesoporous.

The chemical characterization as shown in FIGURE 5 show transmittance spectra for the samples before and after the interaction dye N-719 and AuMPA. The main vibrational peaks of the N-719 dye consist of N=C=S bonding, COOH, and C=C bending [23]. The addition of AuMPA to the N-719 dye result in the disappearance of the vibration band corresponding to the N=C=S bond and causes both a shift and broaden band of COO- bond from 1622 cm^{-1} to 1638 cm^{-1} and from 1381 cm^{-1} to 1384 cm^{-1}, indicating a possible disruption of the dye's COO- bond due to the presence of AuMPA or the formation of chemical bond with the 3-MPA capping agent. Similarly, the spectral bands corresponding to the C=C and C-O bonds in the N-719 dye also exhibit shifts upon the addition of AuMPA, which may be attributed to the overlap between the dye's chemical bonds and the nanoparticles.

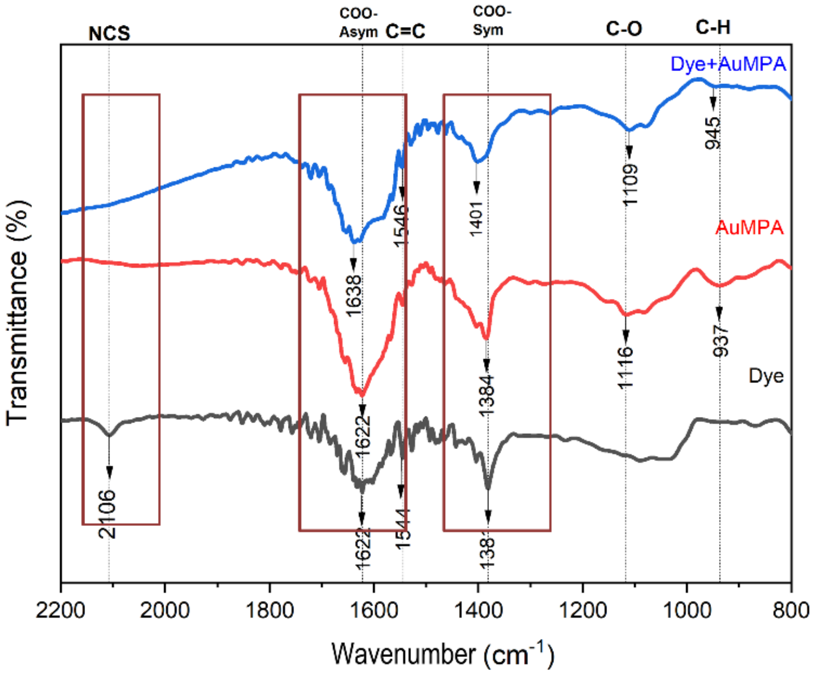


FIGURE 5. FTIR spectra of Dye N-719, AuMPA, and Dye N-719 with AuMPA.

TABLE 1. Band assignment of N-719, AuMPA, and N-719 dyes with the addition of AuMPA [22].

Number of waves (cm^{-1})			**Band Assignment**
Dye	**AuMPA**	**Dye with AuMPA**	
2106	-	-	N=C=S stretching
1622	1622	1638	COO- Asym
1544	-	1546	C=C bending
1381	1384	1401	COO- Sym
-	1116	1109	C-O stretching
-	937	945	C-H bending

The change in vibrational characteristics due to the addition of AgMPA to the dye, as shown in FIGURE 6, is indicated by a shift and broadening of the COO vibration band from 1622 cm^{-1} to 1538 cm^{-1} and from 1381 cm^{-1} to 1385 cm^{-1}, as well as a change in the C=C and C-O bands. The disappearance of the N=C=S band at 2106 cm^{-1} in the spectrum of the dye with AgMPA addition also indicates a chemical interaction between the functional groups of the dye and the surface of the silver nanoparticles.

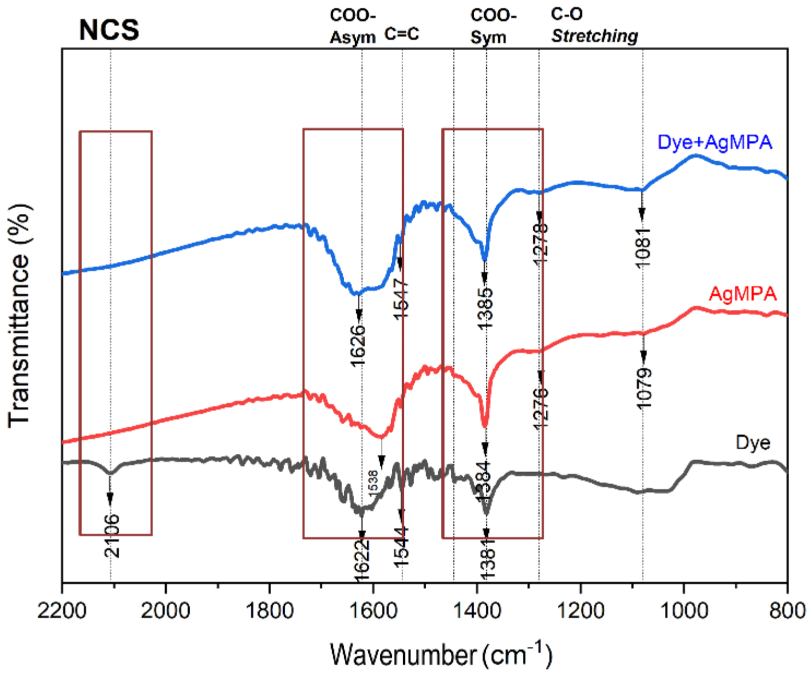
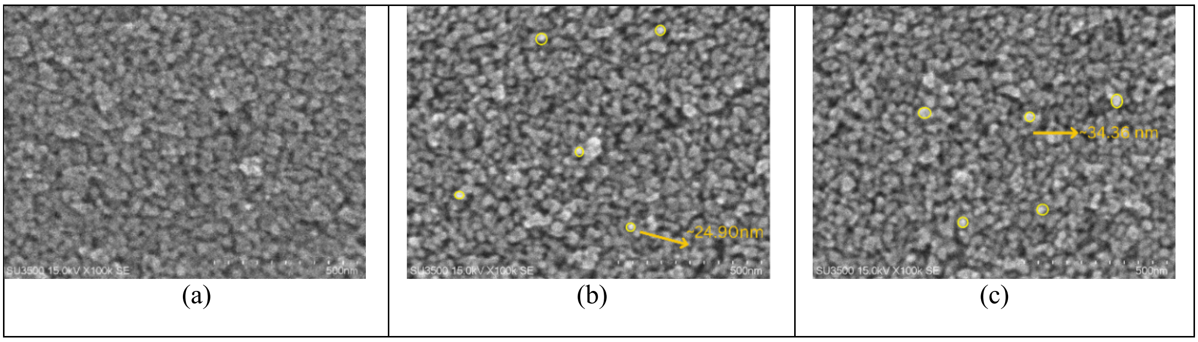


FIGURE 6. FTIR specta of Dye N-719, AgMPA, and Dye N-719 with AgMPA.

TABLE 2. Band assignment of N-719, AuMPA, and N-719 dyes with the addition of AuMPA [22].

Number of waves (cm^{-1})			**Band Assignment**
Dye	**AuMPA**	**Dye with AgMPA**	
2106	-	-	N=C=S stretching
1622	1538	1626	COO- Asym
1544	-	1547	C=C bending
1381	1384	1385	COO- Sym
-	1276-1079	1278-1081	C-O stretching



(a) (b) (c)

FIGURE 7. (a) SEM image of TiO_2 surface immersed in dye N-719, (b) SEM image of TiO_2 surface immersed in dye N-719 with AuMPA, and (c) SEM image of TiO_2 surface immersed in dye N-719 with AgMPA.

FIGURE 7(a) shows the results of the surface morphological characterization of the TiO_2 layer immersed in dye N-719 solution using Scanning Electron Microscopy (SEM). The imaging results reveal the surface morphology of the N-719 dye, indicating that the surface appears to be composed of relatively uniform fine particles with a fairly even distribution. This structure suggests that the N-719 dye effectively covers the surface, with minimal large agglomerations present. After the addition of AuMPA in FIGURE 7(b), there is an increase in density and a slight increase in roughness compared to the reference dye. It can be seen that the size of the grains becomes more varied, and bright dots indicate the presence of metal nanoparticles. This suggests that AuNPs are successfully bound to the surface. Meanwhile, the addition of AgMPA in FIGURE 7(c) shows a morphology with a denser texture and a rougher surface than the previous two samples. The addition of AuMPA or AgMPA to the N-719 dye is observed in the mesoporous TiO_2 layer, with nanoparticles having an average size of approximately ~24.90 nm (AuMPA) and ~34.36 nm (AgMPA) due to the growth of nanoparticles within the mesoporous TiO_2 layer. The observed particle sizes are consistent with the sizes of the nanoparticles used in this study.

3. Performance of DSSC with incorporation of AuMPA or AgMPA

FIGURE 8 show the current density-to-voltage (J-V) curve of the three DSSC devices without and with the addition of AuMPA or AgMPA nanoparticles, as well as the performance parameter data of each device shown in TABLE 3.

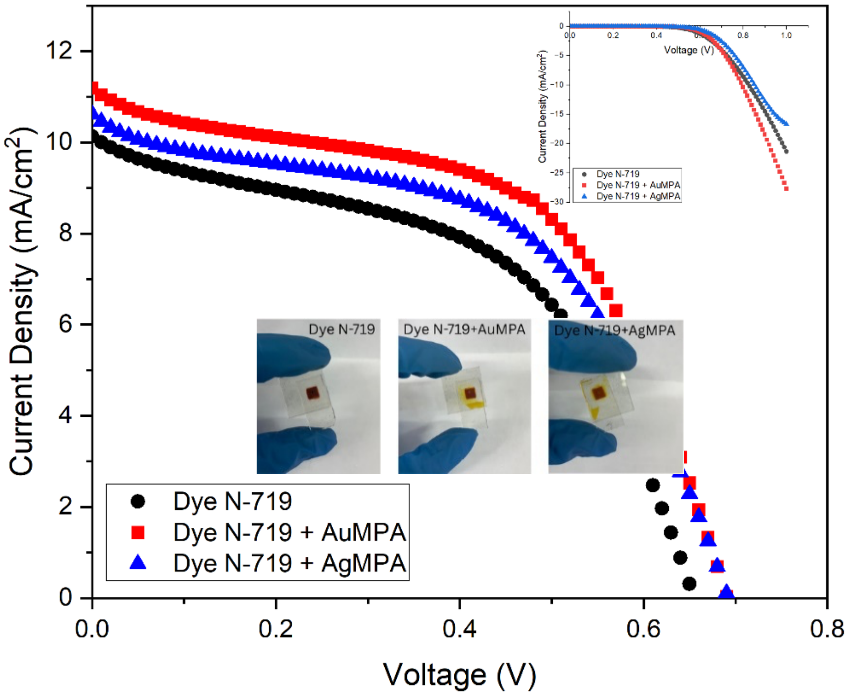


FIGURE 8. J-V characteristics of DSSC device incorporated AuMPA and AgMPA in dye N-719.

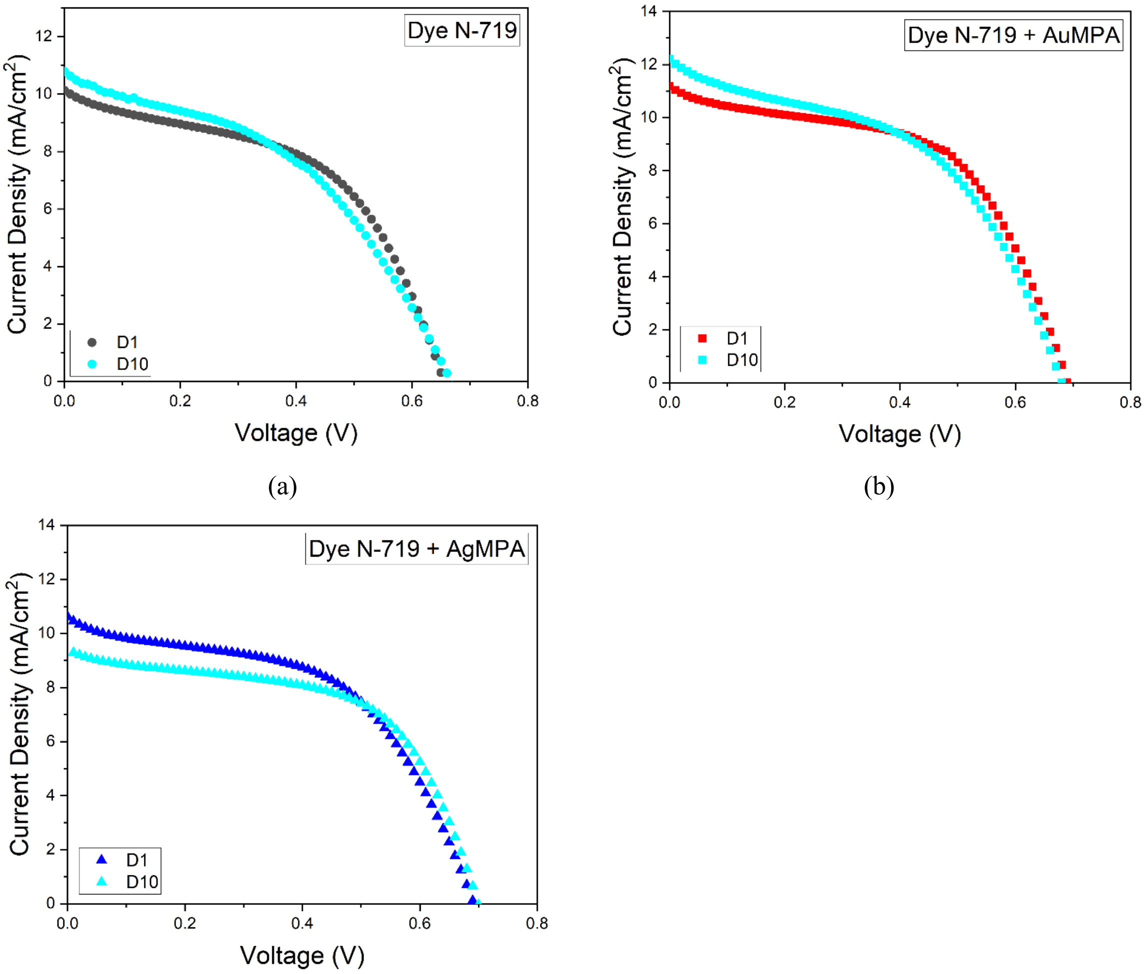
TABLE 3. DSSC performance parameters incorporated AuMPA and AgMPA in dye N-719.

Device name	V_{oc} **(V)**	J_{sc} **(mA/cm^2)**	***FF***	**η (%)**
Dye N-719	0.66	10,14	0,50	3,32
Dye N-719 with AuMPA	0.69	11,19	0,54	4,19
Dye N-719 with AgMPA	0.69	10,62	0,51	3,76

Based on these data, it can be seen that the addition of metal nanoparticles indicates an improvement in the performance of solar cells. DSSC device with the addition of AuMPA shows the highest power conversion efficiency (PCE) of 4.19%, with an increase in short-circuit current density (Jsc) to 11.19 mA/cm^2 and the open-circuit voltage (Voc) to 0.69 V, in comparison to the DSSC without nanoparticles, which showed a PCE of 3.32% with Jsc of 10.14 mA/cm^2 and a Voc of 0.66 V. The addition of AgMPA also shows a similar trend, with a PCE increase to 3.76%. The improved efficiency of AuMPA results from its more pronounced plasmonic effect and good stability, in contrast to AgMPA, which is more prone to aggregation, which reduces its effectiveness. The improvement of solar cell performance with the addition of AuMPA or AgMPA nanoparticles is attributed to their plasmonic effect, which enhances light absorption, improves charge transport through the light-harvesting mechanism in the visible spectrum region, and contributes to beneficial scattering properties [20-21, 24-25].

In this study, the performance measurement of the fabricated DSSC device was evaluated over a 10-day period to assess its lifetime. Measurements are made to monitor changes in efficiency over time, as well as to observe the effect of adding AuMPA or AgMPA metal nanoparticles on the device's resistance to degradation. The measurement results are shown in FIGURE 9 and TABLE 4 below.

The performance degradation of the reference dye device shows that its performance decreases after 10 days compared to the first day, as reflected by the reduction in FF values. This decreasing performance indicates the extent of dye degradation or an increase in the rate of recombination resulting from the storage process. The addition of AuMPA or AgMPA into the dye exhibits a similar trend to the reference device, with a slight decrease in efficiency of approximately 6.21% for AuMPA (from η = 4.19% to 3.93%) and 1.33% for AgMPA (from η = 3.76% to 3.71%). This decrease on the 10th day is due to a reduction in the Fill Factor (FF), which is also influenced by the value of *Jsc*, *Voc*, and the maximum power output of the device. Overall, the device with AgMPA nanoparticles shows a smaller decrease in efficiency compared to the device with AuMPA or the device without nanoparticles. Nevertheless, the efficiency decrease observed in these three devices is not significant, indicating that the devices are still capable of maintaining good performance over a 10-day period.



(a)

(b)

(c)

FIGURE 9. Lifetime performance of our fabricated DSSC (a) reference device, (b) device incorporated AuMPA, and (c) device incorporated AgMPA (for 10 days measurements).

TABLE 4. DSSC performance parameters of our fabricated DSSC (a) reference device, (b) device incorporated AuMPA and (c) device incorporated AgMPA (for 10 days measurements).

Device name	V_{oc} (V)	J_{sc} (mA/cm^2)	*FF*	η (%)
Dye N-719	0,67	10,79	0,43	3,11
Dye N-719 with AuMPA	0,68	12,22	0,47	3,93
Dye N-719 with AgMPA	0,70	9,40	0,56	3,71

This research offers a new perspective by integrating Au and Ag nanoparticles capped by 3-MPA into the dye matrix, demonstrating that AuMPA enhances initial efficiency through a more substantial plasmonic effect. At the same time, AgMPA provides relatively better short-term stability, which may guide practical material selection for DSSC optimization.

CONCLUSION

This study presents a comparison of the addition of gold and silver nanoparticles, capped with 3-mercaptopropionic acid (AuMPA and AgMPA), to the N-719 dye solution in terms of optical characteristics, chemical structure, surface morphology, and working performance of DSSC. The Optical characterization shows that the intrinsic absorbance peak of Dye N-719 overlapped with the plasmonic peak of the nanoparticles, indicating the stability of the nanoparticles in the dye matrix. The characterization of FTIR reveals the formation of chemical interactions between nanoparticles and functional groups in the dye, as evidenced by changes in the molecular bonds of Dye N-719. The results of SEM characterization, with the addition of AuMPA or AgMPA to the mesoporous TiO_2 layer, show the presence of nanoparticles with slightly enlarged sizes, which are approximately 24.90 nm (AuMPA) and 34.36 nm (AgMPA), due to the growth of nanoparticles within the TiO_2 mesoporous layer. The TiO_2 layer also appears more homogeneous after the addition of AuMPA or AgMPA. Meanwhile, the DSSC's power conversion efficiency (PCE) increases after the addition of AuMPA or AgMPA into dye. The DSSC incorporating AuMPA into the dye exhibits better performance compared to AgMPA due to a more substantial plasmon resonance effect and enhanced stability. The lifetime measurements of the devices show that all three fabricated devices maintain good performance over 10 days, with the device incorporating AgMPA exhibiting the smallest efficiency decline in comparison to the device with AuMPA or the one without nanoparticles. Longer-term stability evaluations are necessary to assess the durability of the devices comprehensively. This research suggests opportunities for exploring the incorporation of Au and Ag nanoparticles or further modification of capping agents to achieve an optimal balance between performance and long-term stability in DSSC.

ACKNOWLEDGMENTS

This research is supported by Penelitian Kemendikbudristek, Skema PPS-PTM, Contract No: 257/IT1.B07.1/SPP-LPPM/VI/2024.

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