

# Nickel Oxide/Graphene Oxide (NiO/GO) as Double Hole Transport Layer in Perovskite Solar Cell

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**Abstract:** The hole transport layer is one of the most crucial parts of the perovskite solar cell. Recently, some studies have improved perovskite solar cell performance by modifying the surface of the hole transport layer using double fabrication of inorganic-organic materials. Their studies generate higher power conversion efficiency than inorganic materials but have not generated good stability. Therefore, this study aims to develop a double-hole transport layer using nickel oxide (NiO) and graphene oxide (GO) to improve the perovskite solar cells performance. The experiment utilizes the spin-coating method to observe the effect of combining NiO and GO in perovskite solar cells. The experimental results showed that the double hole transport layers produce better power conversion efficiency of 15.3% compared to single-layer NiO. The double holes can smoothen the holes of the perovskite interface layer and cause molecular uniformity to reduce recombination. In addition, NiO/GO surfaces show better stability to environmental conditions.

**Keywords:** double hole transport layer; graphene oxide; nickel oxide; perovskite solar cell

## 1 INTRODUCTION

A solar cell (also called a photovoltaic cell) is a device that can convert light energy into electrical energy by utilizing photovoltaic technology. The photovoltaic technology uses photons contained in sunlight at specific wavelengths to excite electrons in the semiconductor material. The development of photovoltaic technology has created silicon-based solar cells, thin-film-based solar cells (such as a-Si:H, CdTe, CuInSe<sub>2</sub>, and CIGS), dye-sensitized solar cells (DSSCs), organic photovoltaic cells, and organic-inorganic hybrid cells (perovskite) known as perovskite solar cells [1].

Perovskite solar cell (PSC) is a type of solar cell that contains an organic-inorganic metal halide perovskite material as the light-harvesting active layer. The perovskite material is a compound with the chemical formula ABX<sub>3</sub> (A = CH<sub>3</sub>NH<sub>3</sub>, B = Pb or Sn, and X = Cl, Br, or I) [2-4]. The PSC configuration is evolved from dye-sensitized solar cells (DSSCs) by replacing the pigments in the DSSC with two inorganic dihybrid organic-based hybrid peroxides CH<sub>3</sub>NH<sub>3</sub>PbI<sub>3</sub> [5].

The PSC's structure consists of five components, i.e., Transparent Conductive Oxide (TCO) substrate, Hole Transport Layer (HTL), perovskite layer, Electron Transport Layer (ETL), and counter electrode [6-8]. The TCO substrate is a solar cell body with a conductive layer that functions to flow electric charges. The perovskite layer functions absorbing light to generate electron-hole pairs leading to the Electron Transport Layer (ETL) and the Hole Transport Layer (HTL), respectively, to create currents. The HTL reduces recombination and increases absorption in PSC. The ETL is a layer that transfers electrons resulting from the perovskite intrinsic layer. The counter electrode is a catalyst to accelerate the reaction kinetics of the triiodide reduction process in TCO.

One of the most important components in PSCs is HTL. It functions for optimizing the interface, adjusting the energy compatibility, helping to get higher *PCE*, and causing stability of the PSC device [1, 8, 9]. The most prevalent organic HTL material used in solar cells is poly (3, 4 - ethylene-dioxythiophene): poly (styrenesulfonate) (PEDOT:PSS) because of its simple processing, high

conductivity, and good film processing capability [10]. However, PEDOT:PSS yields low power conversion efficiency (i.e., 11.65%) and low stability due to its acidic, hygroscopic, and electrical properties [11]. Another organic HTL material is spirobifluorene (Spiro-OMeTAD) [12]. This material can produce higher *PCE* than PEDOT:PSS (i.e., 13.5%).

Furthermore, several researchers developed HTL using inorganic materials to increase stability. For example, CuI [13], CuSCN [14], NiO [15], CuO [15], Cu<sub>2</sub>O [16], and MoO<sub>3</sub> [17]. Compared to organic HTL materials, inorganic materials have the advantages of high bore mobility, wide bandgap, low cost, and material availability, which show promising prospects as selective hole contacts in PSCs. NiO is an interesting inorganic HTL material used in solar cells because of its large bandgap, high stability, and high open-circuit voltage value [18]. Nevertheless, NiO gives relatively low efficiency and lower fill factor values due to inadequate hole extraction capability and poor contact of NiO with perovskite [19].

Recently, some studies have modified the surface of HTL to increase the performance of PSCs. Their studies use double fabrication of inorganic-organic materials, such as PEDOT: PSS/NiO, PEDOT: PSS/SrGO [20], and Cu: NiO/PhNa [18]. These materials work by reducing the number of cavities in the perovskite layer to generate higher *PCE* than inorganic materials. However, the various efforts in previous studies have not generated high *PCE* and good stability at the same time. Thus, the lack of stability will become an obstacle to the PSC's commercialization.

Based on the problem above, this study aims to develop a double HTL using a thin-film NiO facilitated with an inorganic material (i.e., graphene oxide (GO)) to yield an efficient and stable PSC. NiO serves to maintain PSC stability. GO as an oxidized derivative of graphene is a material that has very attractive processing or even better optical properties [21] than PEDOT:PSS. The addition of GO as double HTL in PSC functions as a catalyst to accelerate electron flow and induce delocalized electrons with a positive charge on perovskite material. Thus, the stability and efficiency of PSC will increase.

## 2 MATERIAL AND METHODS

This study proposed the development of perovskite solar cells using a double hole transport layer, i.e., Nickel Oxide (NiO) and Graphene Oxide (GO). NiO and GO function as a hole transport layer, known as the double hole transport layer, because perovskite solar cells have two-hole transport layers. The first layer is NiO and the second layer is GO. NiO serves to increase PSC stability, and GO functions to increase the efficiency of PSC. The experimental method utilizes spin-coating to investigate the effect of combining NiO and GO to perovskite solar cells. The development of perovskite solar cells consists of three main stages, i.e., synthetic material preparation, solar cell fabrication, and evaluation. The following subsection explains the detail of these stages.

### 2.1 Synthetic Material Preparation

There are five processes to prepare the synthetic material. The first process is the NiO film material preparation. This process is carried out by mixing nickel acetate tetrahydrate and monoethanolamine in ethanol with a 0.1 M concentration and stirring this mixture for 4 hours using a magnetic stirrer at 70 °C temperature up to the solution is homogeneous and green [22].

The second process is perovskite solution preparation that includes  $\text{CH}_3\text{NH}_3$  solution and  $\text{PbI}_2$  solution. The  $\text{CH}_3\text{NH}_3$  solution is produced by dissolving 90 mg of methylammonium iodide powder in isopropanol 2 mL. The  $\text{PbI}_2$  solution is obtained by dissolving 900 mg of  $\text{PbI}_2$  powder in a dimethylformamide solvent 2 mL and stirring for 24 hours at 70 °C temperature. The centrifugation process is then carried out to get a clear  $\text{PbI}_2$  solution.

The third process is GO preparation using the modified Hummer method [23]. This method is carried out by stirring 2 g graphite powder and 2 g  $\text{NaNO}_3$  in 100 mL  $\text{H}_2\text{SO}_4$ , which yields a 100 mg/mL GO solution.

The fourth process is the ZnO solution preparation using zinc acetate dihydrate (2.95 g; 13.4 mmol) and KOH (1.48 g; 23 mmol). Firstly, the zinc acetate dihydrate is dissolved in 125 ml methanol and stirred using a magnetic stirrer at 65 °C temperature and KOH is dissolved in 64 mL methanol and heated at 60 - 65 °C temperature for 15 minutes. The two solutions are then mixed and stirred at 65 °C temperature for 2.5 hours. Finally, the solution is cooled at room temperature and filtered using a 0.45  $\mu\text{m}$  PVDF syringe filter to get ZnO nanoparticles [24].

The fifth process is silver electrode counter preparation. This process is performed by coating silver in Fluorine doped Tin Oxide (FTO) glass substrate and heating it with evaporation process at  $10^{-6}$  mbar pressure.

### 2.2 Solar Cell Fabrication

This stage aims to fabricate the solar cell on FTO glass substrates. Before fabricating the solar cells, the FTO pattern was cleaned ultrasonically using acetone, detergent, and UV-Ozone Cleaner for 25 minutes [5]. Our study has two solar cells fabrication, i.e., solar cell fabrication with coating NiO/GO and solar cell fabrication with coating NiO as a comparison. The difference between the two solar cells lies in the hole transport layer. Fig. 1 shows the steps in solar cell fabrication with coating NiO/GO as the double hole transport layer.

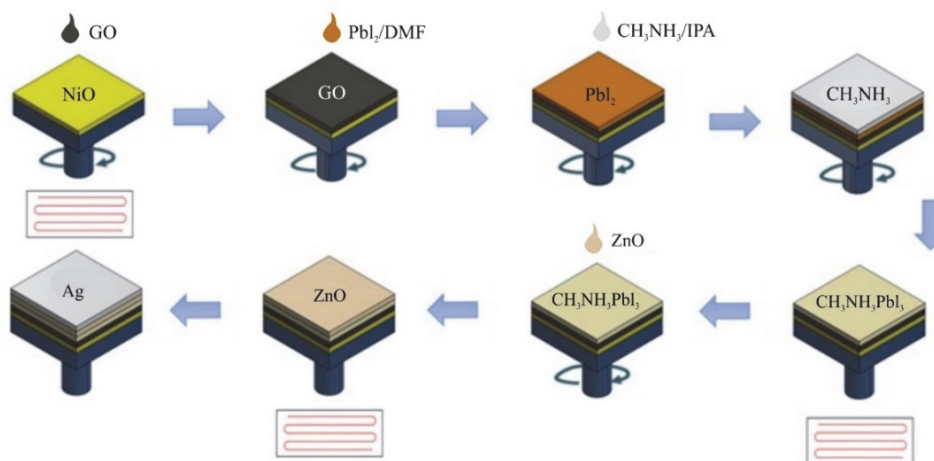


Figure 1 The steps in solar cell fabrication with coating NiO/GO

Based on Fig. 1, the first step is to deposit NiO in FTO glass substrates and perform spin-coating at 1000 rpm for 45 seconds. The GO solution is then dripped in NiO film deposition and processed with spin-coating at a speed of 1000 rpm for 30 seconds and annealing at 200 °C temperature for 1 hour to form double hole transport layers (NiO/GO). The next step is to drip  $\text{PbI}_2$  solution over the NiO/GO layer and carry out the spin-coating process at 1000 rpm speed for 30 seconds, then drip  $\text{CH}_3\text{NH}_3$  solution over the  $\text{PbI}_2$  layer and carry out the spin-coating process

at 1000 rpm speed for 30 seconds. This step results in the conversion from  $\text{PbI}_2$  to perovskite  $\text{CH}_3\text{NH}_3\text{PbI}_3$ . After that, the annealing process is performed at 100 °C temperature for 3 minutes, and the perovskite  $\text{CH}_3\text{NH}_3\text{PbI}_3$  is coated with ZnO solution using the spin-coating process at 1000 rpm speed for 30 seconds, and the annealing process is performed at 100 °C temperature for 3 minutes. Finally, the Ag electrode with a thickness of 100 nm is deposited using a thermal evaporator at a pressure of  $1 \times 10^{-6}$  mbar.

Fig. 2 illustrates the complete structure of perovskite with NiO/GO as the double hole transport layers. The graphene oxide works as a double hole transport layer with FTO/NiO/GO/Perovskite/ZnO/Ag structure. The GO layer functions as a bridge for accelerating ion  $h^+$  leads to NiO cathode and blocking layer to avoid electron recombination. Furthermore, solar cell fabrication with NiO coating is carried out in similar steps without adding a GO solution.

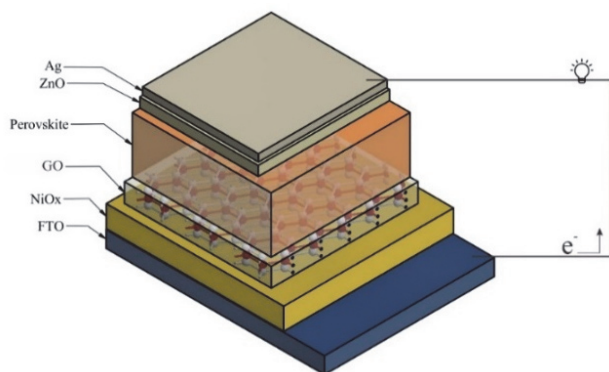


Figure 2 The complete structure of perovskite with NiO/GO

### 2.3 Evaluation

This stage aims to evaluate the proposed perovskite solar cell (PSC) performance. In this study, we evaluate the characteristic of PSC material and the electric current and voltage testing of the solar cells. The evaluation of PSC material uses Scanning Electron Microscope (SEM) testing and UV-Vis test. Meanwhile, PSC's electric current and voltage testing using measurement instruments, including data logger, digital multimeter (MS8250C), and solar simulator.

The SEM testing on PSC functions to observe the characteristics of the surface morphology, texture, shape, and particle size that make up the object, and get information on the arrangement of the grains in the item is observed. The UV-Vis test measures the relative light energy transmitted, reflected, or emitted to material as a wavelength function. Meanwhile, the electric current and voltage testing function to get the efficiency of PSC with the research installation scheme is shown in Fig. 3.

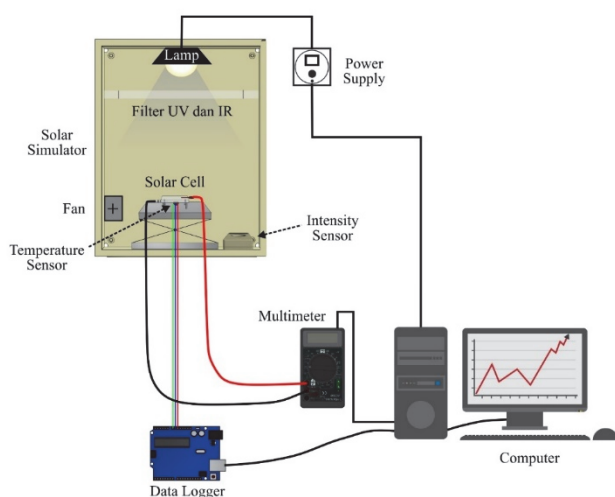


Figure 3 Research installation schema

Based on Fig. 3, the research installation scheme contains a solar simulator with lighting on the PSC to generate the current and voltage. The solar simulator measuring  $500 \times 600 \times 700$  mm includes a halogen lamp with an intensity of  $1000 \text{ W/m}^2$ , an intensity sensor, a temperature sensor, and a data logger MS8250C. This scheme works by generating the electric current and voltage on PSC using the halogen lamps illumination in the solar simulator. The data logger measures the generated current and voltage changes to determine the performance of PSC. This test was done at the Solar Energy Laboratory, Mechanical Engineering Department, Universitas Brawijaya.

## 3 RESULT AND DISCUSSION

### 3.1 SEM Testing

The SEM testing aims to analyze the surface morphology characteristics of the PSC layer. Fig. 4 shows the morphology of the PSCs surface with different hole transport layers (i.e., NiO and NiO/GO). The active area of the PSC is  $1 \text{ cm}^2$ . Therefore, this study uses a magnification of 30.000 times in SEM testing.

Fig. 4a illustrates that the perovskite layer surface with NiO as hole transport layer is hollow or porous. It shows that there are still many cavities between perovskite molecules, marked by dark color. The cavity with NiO as hole transport layer looks rough, marked by the presence of pores that appear cracked. The morphology of the hole transport layer strongly influences the crystal structure [25], as shown in Fig. 4. A rougher surface can produce a worse fill factor so that the power conversion efficiency (PCE) decreases [26]. Meanwhile, a smoother hole transport layer can improve the perovskite solar cell (PSC) performance. It is due to the rough surface having a potential for large recombination, thus lowering the fill factor. The cavity in the NiO as hole transport layer causes the electron transfer between perovskite molecules to be inhibited so that the PSC performance is low.

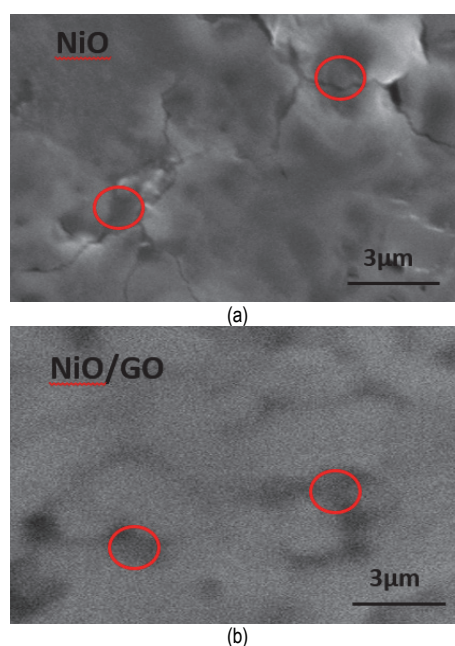


Figure 4 SEM images: (a) morphology of perovskite surface with NiO, (b) morphology of perovskite surface with NiO/GO

The use of a porous NiO layer causes the connectivity between NiO particles to be reduced significantly. The reduced connectivity can cause an increase in photoelectron transport resistance which facilitates electron recombination, thereby reducing the PSC efficiency. Photoelectron transport is the release of electrons from an object's surface when illuminated by light due to the absorption of electromagnetic radiation. Photoelectron transport across the porous NiO layer can only be through the pathway where the NiO particles are interconnected and connected to the perovskite.

Fig. 4b shows that the density of the voids or gaps decreases with the addition of graphene oxide (GO). The addition of GO as a double-hole transport layer can improve the connectivity between perovskite particles and smooth surfaces. The smooth surface will result in closer contact between the perovskite and GO as HTL, thereby accelerating charge transfer and reducing leakage current between the cathode and perovskite absorber. The improved connectivity between perovskite and GO causes a reduction in electron flow resistance and creates a short path, thereby reducing the possibility of recombination and increasing the PSC efficiency. In addition, the NiO/GO as double hole transport layer enhances the perovskite layer morphology and may originate from electrostatic interactions between functional groups on the side chain of the GO polymer and the perovskite precursors. The decrease in pore size in NiO/GO active perovskite films is beneficial for PSC devices' short-circuit currents ( $J_{sc}$ ) and fill factor.

Thus, the SEM test result shows that rougher surfaces will produce fill factor and worse *PCE* [26], and smoother HTL affects to improve the PSC performance. It happens because the rough surface has a large chance of recombination, thus lowering the fill factor. Our observations show that NiO/GO cells' increased efficiency can occur with a higher work function than NiO and a smoother surface morphology. Therefore, we have demonstrated that the double hole transport layer positively affects perovskite/HTL interfaces and bulk perovskite, resulting in a significant increase in the performance of PSC devices. High stability during air storage is an important benefit of the inorganic layer [27].

In addition to surface morphology analysis, the factor that needs to be considered in the selection of solar cell materials is the energy bandgap, i.e., the minimum energy between the ends of the valence band and the conduction band. This study also observed the absorption of work function (WF) on the performance of solar cells using an energy bandgap, as shown in Fig. 5.

Based on Fig. 5, there is a small significant difference in the observed absorption properties, but the WF value significantly affects the PSC efficiency. The WF value of NiO is  $-5.05$  eV, while the WF of NiO/GO is  $-5.00$  eV. It indicates that the larger WF in the double hole transport layer (NiO/GO) is due to GO's heterogeneous electron structure, a mixture of  $sp_2$  and  $sp_3$  hybridization, which can be explained by valence bonds and gradual conduction [28]. In particular, higher WF increases the internal field and minimizes the energy mismatch between the perovskite and the interlayer, which can result in better load transport and PSC performance [29]. In other words,

higher WF in NiO/GO can result in better cell efficiency [30].

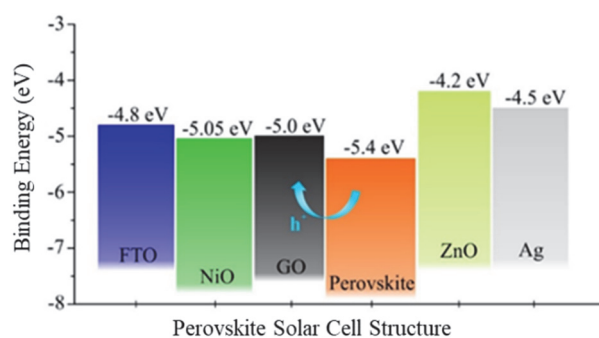


Figure 5 Schematic diagram of PSC binding energy with NiO/GO

Fig. 5 also shows that NiO still has many peaks, which causes the relationship between NiO and the perovskite layer to be low. It results in significant opportunities for recombination. In contrast to the NiO/GO layer, which has a small peak, the relationship with the perovskite layer is good, and the possibility of recombination is little. Thus, it can be concluded that the improved PSC performance obtained using NiO/GO could stem from better energy level matching, lower charge recombination, and better carrier transport than NiO with single HTL. Therefore, double HTL is a promising inter-layer candidate for creating high-performance PSCs with high device stability.

Finally, to further prove that NiO/GO can be a better alternative than NiO in terms of cell stability and cell efficiency, we evaluated the stability of the device as a function of the time duration of environmental conditions [31]. As shown in Fig. 5, the photovoltaic characteristics of NiO/GO have good stability without corrosion on the FTO substrate and a deep work function that will increase the high open-circuit voltage [32]. Meanwhile, a graphene sheet is a single carbon atom layer with a honeycomb lattice that will provide stability for chemicals and efficiently block the penetration of small molecules such as oxygen, hydrogen, and water [33]. Therefore, the increased stability of the device could be due to the hydrophobic nature and chemical inertness of the graphene sheet in GO [31]. It indicates that NiO/GO is a more effective way to maintain high efficiency in the long term.

### 3.2 UV-Vis Testing

The UV-Vis testing aims to study the transport properties of the HTL charge carrier spectrum. This testing uses a photoluminescence circuit to analyze the dependence of open-circuit voltage ( $V_{oc}$ ) on different light intensities. Fig. 6 shows the absorbance of each hole transport layer with a wavelength between 0 and 250 nm. The absorbance of Ni/GO offers a better absorption effect than another single layer (NiO) in all wavelengths. It shows that the ability to absorb photons is more remarkable for NiO/GO, and the resulting holes in the perovskite layer are more efficient. The transport efficiently using double hole transport layer (NiO/GO) results in the carrier recombination in bilayer-based PSCs was relatively decreased compared to single-layer based PSCs [26]. The double hole transport layer (NiO/GO) can induce faster and more efficient hole extraction from perovskites, increasing

solar cell properties, particularly the  $J_{sc}$  and fill factor values.

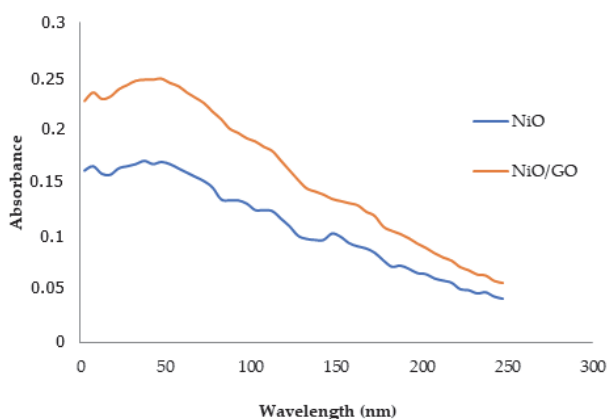


Figure 6 UV-vis spectra

### 3.3 The Current and Voltage Testing

Measurement of current and voltage aims to determine the performance of PSC in terms of power conversion efficiency ( $PCE$ ). The  $PCE$ 's measurements were carried out on PSC with a single hole transport layer (NiO) and PSC with a double hole transport layer (NiO/GO). Examination of electric current, voltage, and efficiency by adding GO as a bilayer aims to determine electric current and voltage changes in 1000 W/m<sup>2</sup> halogen lamps. The testing utilized the research installation scheme, as shown in Fig. 3.

Fig. 7 illustrates the result of the J-V curves of perovskite solar cells graphically with different HTL using the research installation schema that presents the electric current, voltage, maximum fill factor, and maximum efficiency of PSC.

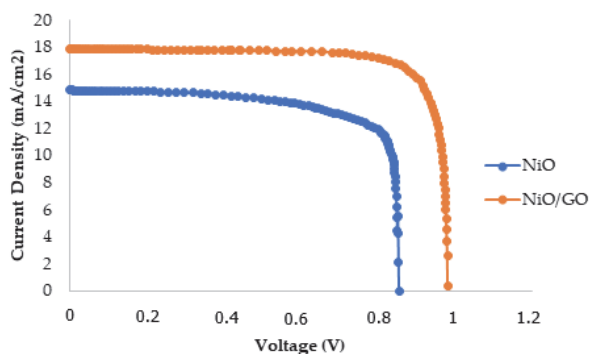


Figure 7 J-V curves of perovskite solar cells with different HTL

Based on Fig. 7, we can compare the photovoltaic properties of PSC with different HTLs, as shown in Tab. 1. The double hole transport layer (NiO/GO) provides higher performance values than the single hole transport layer (NiO). The NiO/GO as a double hole transport layer improves  $V_{oc}$  of 0.127 V,  $J_{sc}$  of 3.1 mAcm<sup>-2</sup>, fill factor of 25.4%, and  $PCE$  of 7.5%. Note those equivalent  $PCE$  parameters and data statistics obtained from 100 different cells.

Table 1 Photovoltaic properties of PSCs with different HTL

HTL	$V_{oc}$ / V	$J_{sc}$ / mA/cm <sup>2</sup>	FF / %	$PCE$ / %
NiO	0.855	14.8	61.64	7.8
NiO/GO	0.982	17.9	87.04	15.3

Finally, Fig. 8 shows the effect of GO as HTL on the perovskite layer, which can reduce the occurrence of recombination due to its function as a blocking layer. As a result of the blocking layer can reduce the recombination between positive and negative ions resulting from photon absorption by the perovskite layer. The decrease in recombination causes the current and fill factor to increase so that the efficiency is greater. Thus, it shows that the insertion of a NiO/GO bilayer can effectively improve the performance of p-i-n planar-structure PSCs.

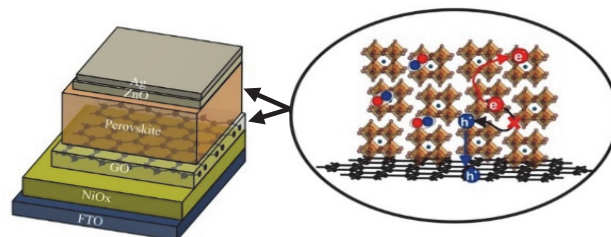


Figure 8 Schematic of a PSC with NiO/GO and the process of electron transfer from perovskite to graphene oxide

The PSC's efficiency value increasing using GO as double HTL occurs because the carbon bond performs electron delocalization. It causes h<sup>+</sup> ions from perovskite to move quickly to HTLNiO so that NiO (as a cathode) is more positive. Meanwhile, the e<sup>-</sup> ions move to the ETLZnO so that the potential difference is getting bigger. The greater the potential difference, the faster the electrons move, causing the current to increase. It causes the efficiency of NiO/GO to be large compared to NiO in PSC.

## 4 CONCLUSIONS

We infer that the combination of NiO and GO as a double hole transport layer can increase the efficiency and stability of the inverted PSCs. The experimental results showed the efficiency of PSCs using NiO/GO and NiO are 15.3% and 7.8%, respectively. The increase in  $PCE$  in NiO/GO is due to the compatibility between the perovskite layer and GO better so that the extraction of holes from perovskite to HTL is more efficient. The density of holes in HTL causes the opportunity for recombination to decrease, which can increase the  $J_{sc}$  and fill factor values. It is essential to the uniformity of the ion more evenly than the single NiO, which results in decreased recombination. Comprehensively, those hybrid interlayers can be a versatile approach for higher performance realization.

However, there is still room for research to improve the efficiency of this PSC. In future research, it is necessary to explore using different inorganic materials in HTL to improve the relationship between the surface of the HTL layer and perovskite in order to increase efficiency.

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