

RESEARCH ARTICLE

The Application of Dye-Sensitized Solar Cell Using rGO and MBs in Series-Parallel Under Low Illumination

JUNG-CHUAN CHOU¹, (Senior Member, IEEE),
JUN-XIANG CHANG¹, (Student Member, IEEE), PO-HUI YANG¹, (Member, IEEE),
CHIH-HSIEN LAI¹, (Member, IEEE), PO-YU KUO¹, (Member, IEEE),
YU-HSUN NIEN², (Member, IEEE), RUEI-HONG SYU¹, AND PO-FENG CHENG¹

¹Graduate School of Electronic Engineering, National Yunlin University of Science and Technology, Douliu 64002, Taiwan

²Graduate School of Chemical and Materials Engineering, National Yunlin University of Science and Technology, Douliu 64002, Taiwan

Corresponding author: Jung-Chuan Chou (choujc@yuntech.edu.tw)

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ABSTRACT The advantage of the dye-sensitized solar cell (DSSC) is utilized indoor fluorescent light illumination, but the output power of DSSC is not high enough. In this study, we provided a structure for DSSCs, and characterized the photovoltaic performances under air mass 1.5 global and indoor fluorescent light illumination. The photoanode of DSSC is based on titanium dioxide (TiO₂) – magnet beads (MBs) – reduced graphene oxide (rGO) composited photoanode (TMGP), which was fabricated by hydrothermal method, spin coating, and doctor blade. According to the experimental results, adding MBs and rGO to DSSCs can enhance the charge transfer ability, reducing the occurrence of charge recombination, thereby improving the photovoltaic performance. DSSCs with TMGP photoanode can maintain a photovoltaic conversion efficiency of around 14 % under indoor light.

INDEX TERMS Dye-sensitized solar cells (DSSCs), electrochemical impedance spectroscopy (EIS), graphene, titanium dioxide (TiO₂).

I. INTRODUCTION

The demand in energy supply has accelerated fossil fuel depletion. It is predicted that the reserves of fossil fuel can only last forty years, sixty years for natural gas, and two hundred for coal [1]. The renewable energy has been considered in recent years, such as photovoltaic, wind power, geothermal heat, hydropower, and biomass energy. Among all renewable energy technologies, the photovoltaic technology is the particularly promising technology for direct conversion sunlight into electricity energy [2], [3], [4], [5], [6]. The developments of the solar cells are able to be divided into four generations: (1) the silicon-based solar cells (single, polycrystalline, and amorphous silicon); (2) the thin-film cells (CdTe, CIGS, and CIS); (3) the organic matter and

nanotechnology cells. However, silicon-based solar cells are high production costs and confine to application in terrestrial photovoltaic. In comparison with silicon-based solar cells and thin-film cells, the dye-sensitized solar cell (DSSC) has low manufacturing costs, high-temperature environment, transparency, flexibility, and generate electricity with indoor light sources [7], [8], [9], [10], [11]. Due to the lower illumination application, the DSSC can work efficiently in dark conditions, for example, dawn or dusk. Hence, DSSC can be the most promising photovoltaic device. The DSSC consists of three parts with the sensitized working layer (photoanode), electrolyte, and the counter electrode. In general, the photoanode material is titanium dioxide (TiO₂), because the TiO₂ has good compatible with dye molecules. Moreover, the photoanode of DSSC is acted as a very important role, which takes responsibility for transforming sunlight into electric power [12]. Therefore, the photovoltaic conversion efficiency

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of DSSC is usually improved by modifying photoanode. Moreover, the topic of our research group is the enhancement of photovoltaic conversion which is ameliorated by modifying the nanostructure of active layer (semiconductor layer) or retardation of dark reaction at interface. For instance, Chou *et al.* in [13], the indium gallium zinc oxide (IGZO), the indium gallium zinc oxide (IGZO), IGZO barrier layer is based on the photoanode with DSSC via sputtering. It can retard dark reaction and back reaction to improve the photovoltaic conversion efficiency. Furthermore, Chou *et al.* in [14], graphene oxide (GO) and zinc oxide (ZnO) have been applied to photoanode to increase the amount of dye-loading and reduce electron transfer impedance, respectively. In addition, Chou *et al.* in [15], GO and magnetic beads (MBs) have also introduced into counter electrode for photocatalytic activity and fast electrocatalyst of triiodide reduction. Furthermore, Chou *et al.* in [16], the Fe_3O_4 has been applied to active layer for the decrease in electron recombination. The advantage of the dye-sensitized solar cell (DSSC) is utilized indoor fluorescent light illumination, but the output power of DSSC is not high enough. Therefore, we propose the titanium dioxide (TiO_2) – magnet beads (MBs) – reduced graphene oxide (rGO) composited photoanode (TMGP) photoanode to enhance the photovoltaic conversion efficiency under low illumination. It under the indoor light condition can maintain photovoltaic conversion efficiency (*PCE*) by about 14 %.

II. EXPERIMENTAL DETAILS

A. MATERIALS

The organic sensitized dye was N719, which purchased from Solaronix Aubonne (Switzerland). The platinum target (99.99% purity) for sputtering was purchased from Ultimate Materials Technology Co., Ltd. (Taiwan). The electrolyte contained with methoxy propionitrile (MPN), iodine (I_2), 4-Tert-Butylpyridine (TBP), lithium iodide (LiI), and 1-propyl-2,3-dimethylimidazolium iodide (DMPII). The titanium tetrachloride (TiCl_4) and acetylacetone (AcAc) were Sigma-Aldrich, (United States).

B. PREPARATION OF THE COLLOIDAL PASTE

The MBs- TiO_2 spin paste had consisted of 1.5 g P25 TiO_2 powder, 0.1 ml acetic acid, 3 ml deionized water (D.I. water) and 0.125 mM MBs. The rGO- TiO_2 doctor blade paste was prepared as follows: Firstly, the 5 mg rGO powder put in 4 ml D.I. water that the solution into an ultrasonic oscillator for one hour. The purpose was dispersed rGO powder evenly in the solution. After that, 2 g TiO_2 powder and 0.4mL absolute alcohol were added into the rGO solution. Finally, the rGO- TiO_2 solution mixture was stirred for one day to obtain rGO- TiO_2 colloid paste. Besides, we prepared pure TiO_2 colloid paste without MBs and rGO powder [17].

C. FABRICATION OF THE PHOTOANODE FOR DYE-SENSITIZED SOLAR CELL

The fluorine-doped tin oxide (FTO) glass ($7 \Omega / \text{cm}^2$) was cleaned by acetone, ethanol, and D.I. water, respectively. The TiCl_4 stock solution was diluted with D.I. water to the 40 mM concentration TiCl_4 treatment solution via the ice bath method. Then the cleaned FTO was immersed into 40 mM TiCl_4 solution at $\sim 70^\circ\text{C}$ for the half-hour to make the TiO_2 compact layer then annealing at 450°C for the half-hour at the furnace. The working layer was based on the TiO_2 compact layer by using the spin method and doctor blade method that the photoanode annealing at 450°C for the half-hour. Finally, the annealed photoanode used TiCl_4 treatment and annealing for a half-hour. Annealing the photoanode film at an ambient temperature of 450°C enables the removal of organic impurities in the photoanode film and strengthens the contact between nanoparticles within the film.

D. INSTRUMENTATION

The photocurrent-voltage curve measurements were conducted under Xe lamp solar simulator (MFS-PV-Basic-HMT, Taiwan) with the sunlight intensity of $100 \text{ mW}/\text{cm}^2$ and indoor fluorescent light source (T5) and light decay filter. Besides, we used light filter 80%, 50%, 30%, and 10% to investigate the DSSCs performance of different sunlight intensities. The Nyquist plot was used to measure the frequency range of 1 MHz to 50 Hz in the dark at a potential of 0.7 V.

III. RESULTS AND DISCUSSION

A. PHOTOVOLTAIC PERFORMANCES OF THE DSSC WITH TiO_2 AND TMGP

The current-voltage curves (J-V) of DSSC based on TiO_2 and TMGP, the photovoltaic conversion efficiency (*PCE*) is enhanced by 31.75 % from 5.20 % to 6.85 %, as shown in Fig. 1. This improvement in photovoltaic conversion efficiency is due to the enhancement of short-circuit current density (J_{SC}) by magnetic beads (MBs) and reduced graphene oxide (rGO). First, the MBs provides another charge transfer path, which can improve the charge transfer characteristics in the TiO_2 film and reduce the electron recombination opportunity. Besides, the rGO is acted as a bridge to accelerate the excited-electron from the conduction band (CB) of titanium dioxide (TiO_2) to the CB of FTO. This role of rGO can reduce the dark reaction between the excited-electron and oxidized-dye molecule [18], [19], [20], [21], [22], [23].

Moreover, the equivalent circuit for DSSC in this study is exhibited in Fig. 2, and it would not exhibit in electrochemical Impedance Spectroscopy (EIS) measurement as below. It composes of R_s , R_1 , R_2 , C_1 , and C_2 . First, the R_s indicates the resistance between FTO substrate and wire. Besides, Z_1 indicates high-frequency impedance of interface between electrolyte and counter electrode, which is the

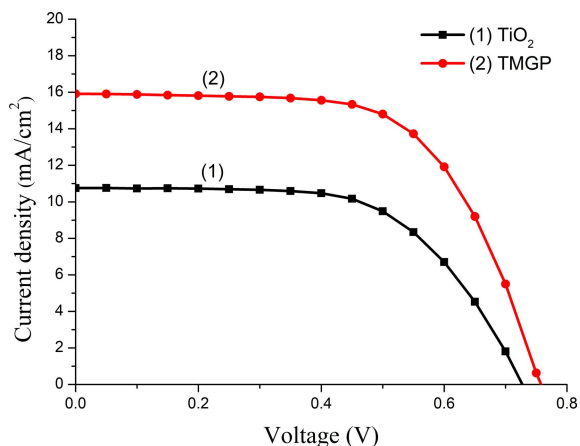


FIGURE 1. Schematic photo of the flexible arrayed NiO biosensor.

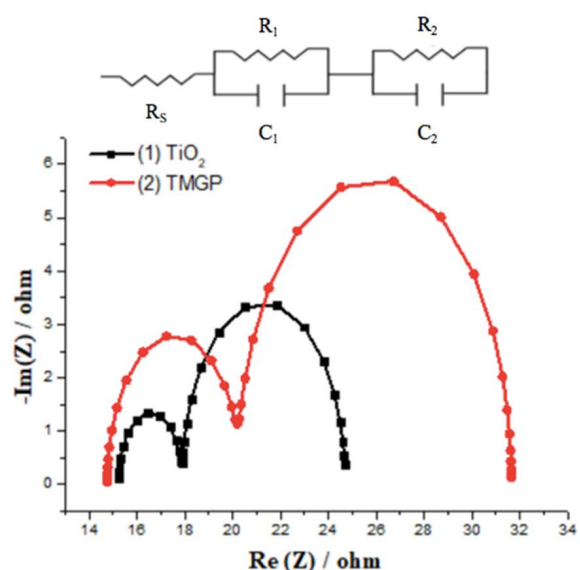


FIGURE 2. Nyquist plots of the DSSCs with different photoanodes and equivalent circuit for DSSC.

first semicircle at the left-hand side. An equivalent circuit, Z_1 indicates the parallel connection between R_1 and C_1 , and R_1 is the interface resistance between the counter electrode and electrolyte. Additionally, Z_2 is the frequency impedance of interface between the electrolyte and the active layer (photoanode), which is the second semicircle at the right-hand side. An equivalent circuit, the Z_2 is the parallel connection between R_2 and C_2 , and R_2 is the interface resistance between the active layer (photoanode) and electrolyte. Additionally, the DSSC is operated in direct current, that the capacitance can be ignored.

It can be seen from Fig. 2 that R_2 is reduced from 11.4Ω to 6.8Ω because MBs provide another charge transfer path for the photoanode for better electron transfer, and the rGO can increase the amount of dye in the photoanode and reduce charge recombination. Because of the above properties, MBs

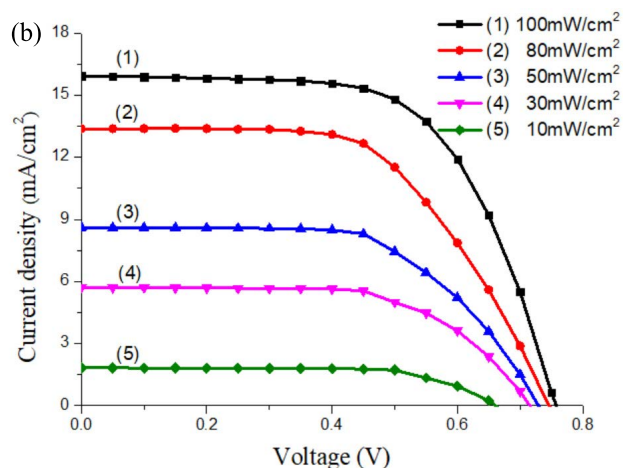
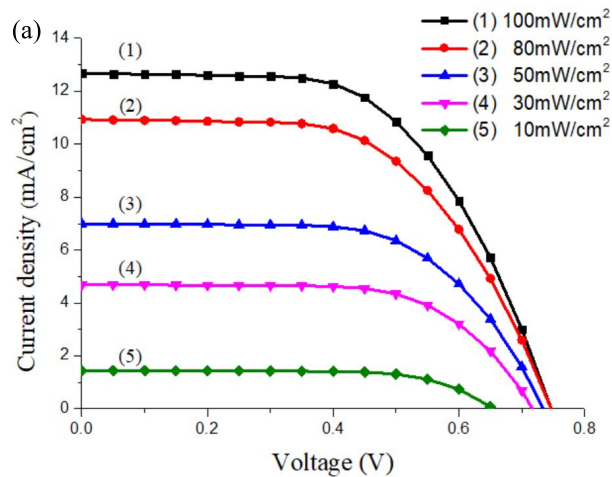


FIGURE 3. J-V curves of DSSCs based on (a) TiO_2 photoanode and (b) TMGP photoanode under low illumination.

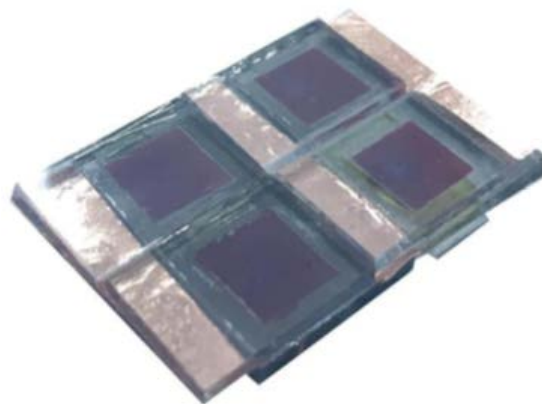


FIGURE 4. Photograph of DSSCs in two series and two parallel.

and rGO enable more excited electrons to be transported to external circuits. Finally, the J_{SC} current can be enhanced [24], [25]. Furthermore, the details of MBs and rGO were shown in our previous research [17].

TABLE 1. Photovoltaic parameters of DSSC based on TMGP under different light intensities.

Xe Lamp Solar Simulator				
Intensity(mW/cm ²)/ Lux (lx)	V_{OC} (V)	J_{SC} (mA/cm ²)	$F.F.$ (%)	PCE (%)
100/ 683 K	0.75	15.73	59.33	6.85
80/ 546 K	0.74	13.45	62.37	7.76
50/ 341 K	0.72	8.65	65.63	8.17
30/ 204 K	0.71	5.75	68.10	9.27
10/ 68 K	0.69	1.98	72.29	9.88
Indoor Fluorescent Lamp				
Intensity(mW/cm ²)/ Lux (lx)	V_{OC} (V)	J_{SC} (mA/cm ²)	$F.F.$ (%)	PCE (%)
100/ 683 K	0.63	518.4	75.52	14.08
80/ 546 K	0.61	458.3	74.38	14.13
50/ 341 K	0.61	303.2	75.76	14.58
30/ 204 K	0.60	157.4	74.29	14.69
10/ 68 K	0.53	51.9	68.86	8.52

TABLE 2. Photovoltaic parameters of DSSC based on TiO₂ under different light intensities.

Xe Lamp Solar Simulator				
Intensity(mW/cm ²)/ Lux (lx)	V_{OC} (V)	J_{SC} (mA/cm ²)	$F.F.$ (%)	PCE (%)
100/ 683 K	0.74	12.61	55.70	5.23
80/ 546 K	0.73	10.80	58.24	5.45
50/ 341 K	0.71	7.01	62.73	5.84
30/ 204 K	0.71	4.67	65.10	6.17
10/ 68 K	0.65	1.42	70.29	6.49
Indoor Fluorescent Lamp				
Intensity(mW/cm ²)/ Lux (lx)	V_{OC} (V)	J_{SC} (mA/cm ²)	$F.F.$ (%)	PCE (%)
100/ 683 K	0.62	462.5	69.49	11.39
80/ 546 K	0.61	389.2	66.41	10.44
50/ 341 K	0.59	242.6	64.97	9.69
30/ 204 K	0.57	157.4	62.69	8.79
10/ 68 K	0.49	46.8	52.74	5.50

B. PHOTOVOLTAIC PERFORMANCE OF THE DSSCs IN SERIES-PARALLEL MODULES

Fig. 3 (a) and (b) show the photovoltaic conversion efficiency (PCE) of DSSC based on TiO₂ and TMGP under low illumination. The TMGP photoanode of DSSC is enhanced 44.23 %, from 6.85 % to 9.88 %, and the TiO₂ photoanode of DSSC is enhanced 24.29 %, from 5.23 % to 6.49 % In addition, the open-circuit voltage (V_{OC}) is decreased from 0.75 V to 0.69 V, which can be attributed to the decrease in the amount of excited-electron. Because the V_{OC} is the difference between Fermi-level of active layer and redox potential

TABLE 3. Photovoltaic parameters comparison of photovoltaic parameters of DSSC with other literature [26].

Xe Lamp Solar Simulator [26]				
Intensity(mW/cm ²)/ Lux (lx)	V_{OC} (V)	J_{SC} (mA/cm ²)	$F.F.$ (%)	PCE (%)
100/ 683 K	0.65	15.01	57.13	5.66
80/ 546 K	0.64	12.51	58.31	5.84
50/ 341 K	0.62	8.40	59.44	6.29
30/ 204 K	0.61	5.47	60.68	6.86
10/ 68 K	0.59	1.71	61.36	5.88
Indoor Fluorescent Lamp [26]				
Intensity(mW/cm ²)/ Lux (lx)	V_{OC} (V)	J_{SC} (mA/cm ²)	$F.F.$ (%)	PCE (%)
100/ 683 K	0.59	330.0	65.80	6.90
80/ 546 K	0.58	260.0	69.33	7.02
50/ 341 K	0.57	170.0	74.68	7.46
30/ 204 K	0.57	110.0	80.15	7.97
10/ 68 K	0.56	20.0	83.02	7.42

TABLE 4. Photovoltaic parameters of DSSC based on TMGP in series and parallel.

Connection	Active area (cm ²)	V_{OC} (V)	I_{SC} (mA/cm ²)	$F.F.$ (%)	PCE (%)	Ref.
Parallel	1.00	0.73	15.50	58.55	6.61	This study
Series	1.00	1.41	7.65	57.28	6.16	This study
Series & Parallel	2.00	1.42	15.23	55.04	5.93	This study
Series	2.56	1.46	13.08	59.13	4.48	[27]
Parallel	2.56	0.73	28.16	59.14	4.35	[27]

of electrolyte. Besides, the amount of excited-electrons are reduced with decline of light intensity. Similarly, because the amount of excited-electron is decreased, the J_{SC} current is also reduced. The enhancement in photovoltaic conversion efficiency is due to the increase in fill factor ($F.F.$). The fill factor is increased from 59.33 % to 72.29 %. However, the better PCE of DSSC can be obtained under lower illumination than indoor fluorescent, which are due to fewer amount of excited-electrons. Moreover, the higher improvement in photovoltaic conversion efficiency under low illumination can be obtained from DSSC based on TMGP. Because the amount of excited-electrons is little, the decrease of dark reaction is helpful for the improvement of photovoltaic conversion efficiency of DSSC under low illumination.

In the result with Table 1 and Table 2, the PCE of DSSC based on TiO₂ decreased from 11.39 % to 5.50 % as the light intensity was decreased from 1.75 mW/cm² to 0.22 mW/cm². In addition, the J_{SC} dropped from 462.5 μ A/cm² to 46.8 μ A/cm² as the light intensity was decreased from 1.75 mW/cm² to 0.22 mW/cm². Moreover, the V_{OC} of DSSC based on TiO₂ decreased from 0.62 V to 0.49 V as the light

intensity was decreased from 1.75 mW/cm² to 0.22 mW/cm². Because the dye molecule has a better response with indoor fluorescent lamp, the V_{OC} of 1.75 mW/cm² was higher than that of 10 mW/cm². In addition, the FF was decreased from 69.49% to 52.74%, while the light intensity was decreased from 1.75 mW/cm² to 0.22 mW/cm². Moreover, the result in the PCE decreased from 11.39 % to 5.50 %. Photovoltaic conversion efficiency decreased from 11.39 % to 5.50 % while light intensity was decreased from 1.75 mW/cm² to 0.22 mW/cm².

In Table 3, we compare DSSCs under different light intensities with other literature. Chou *et al.* in [26], the DSSC in the IGZO retardation structure. The PCE is enhanced by 21.20 %, from 5.66 % to 6.86 %. In Table 4, we compare the DSSC module with other literature. Chou *et al.* in [27], the DSSC in parallel connection and series connection are fabricated by silver paste connection. The I_{SC} , V_{OC} , PCE of series connection DSSC are 13.08 mA/cm², 1.46 V and 4.58 %. The I_{SC} , V_{OC} , PCE of parallel connection DSSC are 28.16 mA/cm², 0.73 V and 4.35 %.

According to previous literature [26], [27], the reduced dark reaction for photoanode of DSSC is a key point to enhance photovoltaic performances, but there are a few literature to apply the reduced dark reaction on DSSC. Moreover, DSSCs in series-parallel has not been investigated, and it has the potential to develop to drive the device.

IV. CONCLUSION

In summary, the properties of DSSCs with MBs and rGO in series-parallel have been investigated. The DSSCs with MBs and rGO can obtain higher photovoltaic conversion efficiency under low illumination, which is due to the retardation of dark reaction. Because the amount of excited-electrons is few under low illumination, a decrease in reverse recombination is a key point to increase the photovoltaic conversion. MBs provides another charge transfer path, which can improve the charge transfer. Besides, the rGO is acted as a bridge to accelerate the excited-electrons from the conduction band of FTO to the conduction band of titanium dioxide (TiO₂). In other words, rGO can retard the dark reaction between the excited-electrons and oxidized-dye molecules. The advantage of the dye-sensitized solar cell (DSSC) is utilized indoor fluorescent light illumination, but the output power of DSSC is not high enough under indoor fluorescent light illumination. Moreover, the output power can be enhanced by series and parallel.

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JUNG-CHUAN CHOU (Senior Member, IEEE) was born in Tainan, Taiwan, in July 1954. He received the B.S. degree in physics from the Kaohsiung Normal College, Kaohsiung, Taiwan, in 1976, the M.S. degree in applied physics from Chung Yuan Christian University, Chung-li, Taiwan, in 1979, and the Ph.D. degree in electronics from National Chiao Tung University, Hsinchu, Taiwan, in 1988. From 1979 to 1991, he worked as a Lecturer, an Associate Professor, and the Director of the Department of Electronic Engineering, Graduate School of Electronic Engineering, Chung Yuan Christian University. From 1997 to 2002, he was the Dean of the Office of Technology Cooperation, National Yunlin University of Science and Technology, Yunlin, Taiwan. From 2002 to 2009, he was the Chief Secretary with the National Yunlin University of Science and Technology. From 2009 to 2010, he was the Director of Library with the National Yunlin University of Science and Technology. From 2010 to 2011, he was the Director of the Office of Research and Development, National Yunlin University of Science and Technology. From 2011 to 2017, he worked as a Distinguished Professor with the Department of Electronic Engineering, National Yunlin University of Science and Technology. From 2013 to 2018, he was the Director of Administration, Testing Center for Technological and Vocational Education. Since 1991, he has been working as an Associate Professor with the Department of Electronic Engineering, National Yunlin University of Science and Technology, where he has been a Professor, since 2010. Since 2018, he has been working as the Lifetime Chair Professor with the Department of Electronic Engineering, National Yunlin University of Science and Technology. Since 2018, he has been the Deputy Director of Headquarters, Testing Center for Technological and Vocational Education. His research interests include sensor material and device, biosensor and systems, microelectronic engineering, optoelectronic engineering, solar cell, and solid state electronics.



JUN-XIANG CHANG (Student Member, IEEE) was born in Taichung, Taiwan, in October 1995. He received the bachelor's degree from the Department of Microelectronic Engineering, National Kaohsiung University of Science and Technology, Kaohsiung, Taiwan, in 2018, and the master's degree from the Graduate School of Electronic Engineering, National Yunlin University of Science and Technology, Yunlin, Taiwan, in 2021. His research interest includes dye-sensitized solar cell.



PO-HUI YANG (Member, IEEE) received the B.Eng. degree in marine electronics engineering from National Taiwan Ocean University, in 1993, the M.S. degree in industrial education from National Taiwan Normal University, in 1993, and the Ph.D. degree from the Institute of Electrical Engineering, National Chung Cheng University, in 2001. From 2001 to 2003, he was an Assistant Professor with the Department of Electronics, Southern Taiwan University of Science and Technology. From 2003 to 2004, he was a High-Performance Digital IC Design Engineer and a Circuit Design Section Manager with the SoC Technology Center (STC), Industrial Technology Research Institute (ITRI), Hsinchu, Taiwan. He joined the Department of Electronic Engineering, National Yunlin University of Science and Technology, as an Assistant and an Associate Professor. His research interests include high-speed and low-power CMOS IC design, advanced IC packaging, and low-power biosensor IC design.



CHIH-HSIEN LAI (Member, IEEE) was born in Taichung, Taiwan, in 1968. He received the B.S. and M.S. degrees in electrical engineering and the Ph.D. degree in photonics and optoelectronics from National Taiwan University, Taipei, Taiwan, in 1990, 1992, and 2010, respectively. He worked with Telecommunications Industry for a number of years, whilst also worked extensively as an Assistant Professor with the Department of Electronic Engineering, Hwa Hsia Institute of Technology, Taipei, from 2004 and 2012. In 2012, he joined the Department of Electronic Engineering, National Yunlin University of Science and Technology, Yunlin, Taiwan. He served as the Chairperson of the Department of Electronic Engineering, from 2017 to 2020. He is currently a Professor with the Department of Electronic Engineering, National Yunlin University of Science and Technology. His current research interests include optical and terahertz guided-wave structures, nanophotonic devices, and optoelectronic devices.



PO-YU KUO (Member, IEEE) was born in Taichung, Taiwan, in 1980. He received the M.S. and Ph.D. degrees in electrical engineering from The University of Texas at Dallas, in 2006 and 2011, respectively. In 2013, he joined the Department of Electronic Engineering, National Yunlin University of Science and Technology, Yunlin, Taiwan, where he is currently an Associate Professor. His research interests include analog circuits, power management circuits, and biosensors measurement.



YU-HSUN NIEN (Member, IEEE) received the Ph.D. degree from the Department of Material Sciences and Engineering, Drexel University, Philadelphia, PA, USA, in 2000. He is currently a Professor and the Director of the Center for Industrial Pollution Prevention Research, National Yunlin University of Science and Technology, Yunlin, Taiwan. His research interests include materials engineering and sustainable energy development.



PO-FENG CHENG was born in Nantou, Taiwan, on July 1999. He received the bachelor's degree from the Department of Electrical Engineering, National Kaohsiung University of Science and Technology. He is currently pursuing the master's degree with the Graduate School of Electronic Engineering, National Yunlin University of Science and Technology, Yunlin, Taiwan. His current research interest includes dye-sensitized solar cells.

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RUEI-HONG SYU was born in Changhua, Taiwan, in February 1998. He received the bachelor's degree from the Department of Electro-Optical Engineering, Southern Taiwan University of Science and Technology, Tainan, Taiwan, in 2020, and the master's degree from the Master's Program of Electronic Engineering, National Yunlin University of Science and Technology, Yunlin, Taiwan. His research interests include dye-sensitized solar cell and electrochemical impedance spectroscopy.