Research Article

Development of Paint-Type Dye-Sensitized Solar Cell Using Carbon Nanotube Paint

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This paper proposes paint-type dye-sensitized solar cells (DSCs). DSCs, one type of solar cell, generally consist of a dye-attached semiconducting electrode, a metallic electrode, and an electrolyte. The DSC generates power through the excitation of the electrons in the dyes and the oxidation-reduction reaction between the dyes and the electrolyte. For our paint-type DSC, we made two electrodes by painting two types of paint on substrates. We used carbon nanotubes (CNTs) as the paint material because they have both semiconducting and metallic properties. This enabled us to prepare semiconducting and metallic electrodes easily by simply painting with the CNT paint. As a result of testing, we determined that our DSCs were capable of power generation. Our paint-type DSCs have the potential to provide power as a unique and useful device for daily life in the near future.

1. Introduction

Recently, clean power generation, such as solar power, has attracted much attention because of energy consumption and environmental problems. Many kinds of solar cells have been developed. This paper focuses on one unique type of solar cell: the dye-sensitized solar cell (DSC). DSCs generally consist of a dye-attached semiconducting electrode and a metallic electrode. There is an electrolyte between electrodes, which face each other. The DSC generates power through the excitation of electrons in the dye and the oxidation-reduction reaction between the dyes and the electrolyte. DSCs are known for their customizability and are expected to be used where customizability is needed such as interior design.

In this study, we propose a unique DSC: a paint-type DSC. We made two electrodes for our DSC by painting two kinds of paint on the substrates. We used carbon nanotubes (CNTs) [1–5] as the paint material. CNTs are commonly known for having many useful properties, such as high electrical conductivity [6], thermal conductivity [7], mechanical strength [8, 9], and chemical stability [10]. In particular, we focused on one trait of CNTs, namely, they have both semiconducting and metallic properties [11–13]. There have been many studies using CNTs, particularly for

electrodes of solar cells [14–18]. We aimed to make both semiconducting paint and metallic paint by applying the CNTs. In doing so, we could make semiconducting and metallic electrodes easily by simply painting with the CNT paint. In addition, CNTs are known to have large surface areas, so we could attach enough dye to the surface of the CNTs. In this study, we constructed our paint-type DSC, examined its usability, and aimed to improve its performance. In the research area of solar cells, there are many reports applying CNTs or other nanocarbon materials, e.g., organic solar cells, to improve device performance [19–23]. For DSCs, CNTs have been applied as a counter electrode [24] or a transparent electrode [25]. In contrast, by advances in the research area of printed electronics, CNTs can be painted on various substrates including plastic films [26]. However, the full paint-type DSC as we propose has not been reported yet. We believe our approach is unique and will be useful for our life in near future.

2. Bases of Dye-Sensitized Solar Cell and Carbon Nanotube Paint

2.1. Dye-Sensitized Solar Cell. Modern DSCs were invented by Prof. Michael Grätzel of the École polytechnique fédérale

de Lausanne (EPFL) in 1991 [27]. After that, DSCs garnered much attention and were reported to show more than 10% of photoelectric conversion efficiency [28–32]. A DSC (Figure 1) generally has a semiconducting (negative) electrode (on a transparent electrode) and a metallic (positive) electrode, and the two electrodes keep an electrolyte between themselves. Typically, DSCs use titanium dioxide for the semiconducting electrode, platinum for the metallic electrode, and iodine-based material for the electrolyte. Its flow of operation is as follows. First, the dyes are excited by sunlight that irradiates the semiconducting electrode, and excited electrons transfer to the metallic electrode via the semiconducting electrode and an external circuit. Then, the electrons move from the metallic electrode to the electrolyte and change from iodine into iodide ion. The iodide ion reduces the oxidized dyes and is turned back to iodine. Through this series of operations, electric current flows in the DSC circuit.

2.2. Carbon Nanotube Paint.

A CNT is a nanoscale material, so it is hard to use or handle CNT as it is. For this reason, there are many studies on how to make them easy to use [33]. In this study, we use single-walled CNT [34] dispersion as paint. Substrates can be given semiconducting and metallic properties, which are characteristics of CNTs, by painting them with CNT paints. Substrates made in this way are used as the electrodes of our paint-type DSCs. In the future, our DSCs will be able to be directly installed anywhere paint can be used. Therefore, they will contribute to the continued expansion of solar cells and can be expected to be applied to various objects.

3. Experimental Methods

3.1. Concept of Our Paint-Type Dye-Sensitized Solar Cells.

Our DSC needed two electrodes: one semiconducting and one metallic. As the first step of this study, we used sketchbook papers as substrates of the electrodes. Each paper substrate had an area of 15 by 15 mm^2. We prepared the semiconducting and metallic CNT paints for the semiconducting and metallic electrodes. Each of the electrodes was made by painting the CNT paints to the paper substrate. Figure 2 shows the schematic of this way to make an electrode. The electrodes were wired, made to face each other, and electrolyte was put between them.

3.2. Making Metallic Electrodes.

We used single-walled carbon nanotubes, specifically, HiPco (NanoIntegris Inc., powder), for our metallic CNT paint because it is known that HiPco type CNTs are one of the typical CNTs which have high conductivity and contain both semiconducting and metallic CNTs when they are fabricated. Although they contain semiconducting CNTs, we can assume them to be metallic CNTs as a whole because of contained metallic ones. We also used sodium dodecyl sulfate (SDS) as a dispersant [35, 36]. First, we made CNT-mixed liquid by blending 6 mL of pure water, 14 mg of HiPco, and 14 mg of SDS. Then, we made CNT dispersion
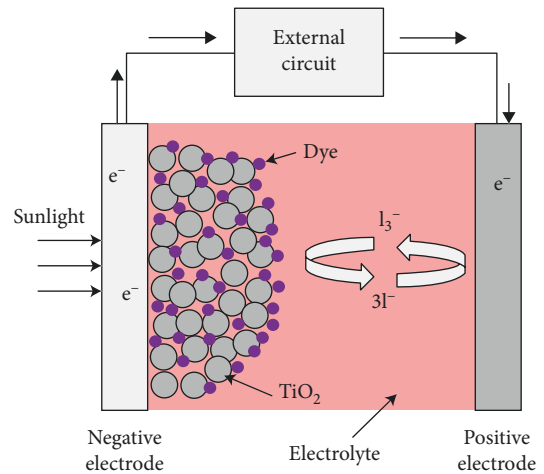

FIGURE 1: General operation principle of DSC.

by irradiating the CNT-mixed liquid with ultrasonic waves [37] for 1 hour. We then painted about 0.25 mL and 1 mL of CNT dispersion to the paper substrate and dried it at approximately 100°C. Figure 3(a) shows the obtained sample by this way. Finally, we wired the paper substrate using conductive silver paste.

3.3. Making Semiconducting Electrodes.

We used 99% pure semiconducting CNTs (NanoIntegris Inc., dispersion). This dispersion contains 0.01 mg of the CNTs per 1 mL. The reason why we used these CNTs is that the semiconducting property must be required to prepare the aiming semiconducting electrodes. As described above, the typical CNTs contain both semiconducting and metallic CNTs. However, they show metallic property. Therefore, we must prepare refined semiconducting CNTs. We here used the products. Then, we used anthocyanin dye as the dye. First, we prepared a dye solution by mixing 0.4 mg of the dye and 0.2 mL of pure water. Then, we prepared the semiconducting paint by mixing the dye solution and the semiconducting CNT dispersion. Next, we painted the semiconducting paint to the paper substrate and dried it at 100°C. After that, we used a container of the same size as the paper substrate to prevent the semiconducting paint with low viscosity from flowing down off the paper substrate. Figure 3(b) shows the obtained sample by this way. Finally, we wired the paper substrate using conductive silver paste.

This semiconducting electrode tended to have lower conductivity than the metallic one. Generally, its decrease caused a drop in the power generating efficiency of the DSC. One of the reasons for this decrease in conductivity is the movement of the excited electrons into and out of the dyes. When generating power, the electrons must move to the connected external circuit via the semiconducting electrode. However, when the conductivity is low, many electrons cannot reach the circuit. Thus, the conductivity must be high for efficient power generation. In the original DSC, the transparent electrode is used and collects the electrons. But our aiming DSC does not have the transparent electrode. To handle this problem, we designed an
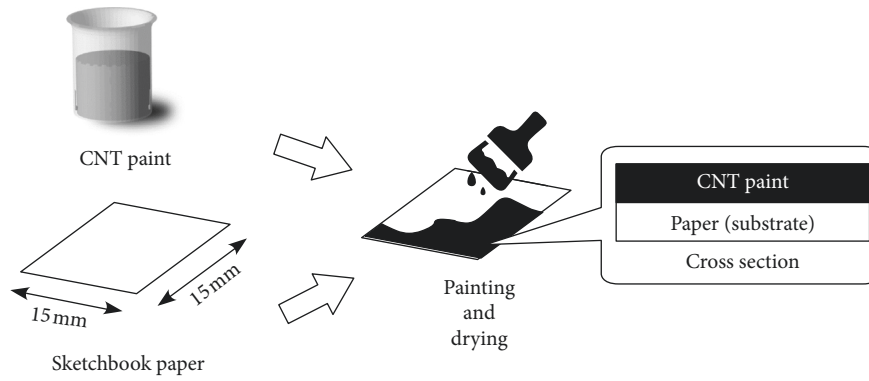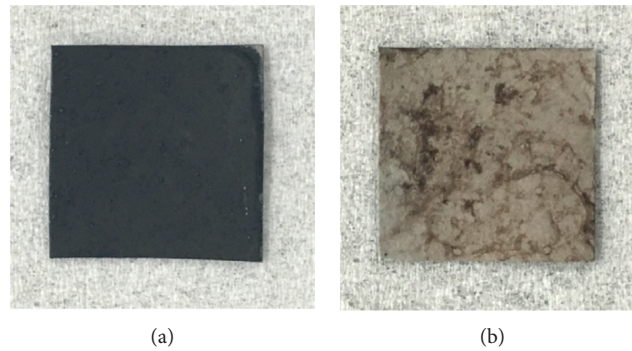
FIGURE 2: Schematic of making method for electrodes.

(a)   (b)

FIGURE 3: (a) Metallic electrode obtained by painting our metallic CNT paint and (b) semiconducting electrode obtained by painting our semiconducting CNT paint. Both electrodes have area of 15 by 15 mm^2.

additional construction. Specifically, we painted conductive paint, such as the metallic CNT paint as described above and conductive silver paste, as an undercoat for the semiconducting CNT paint. We here used conductive silver paste as the undercoat paint for simplicity as shown in Figure 4.

3.4. Measurements for Our Paint-Type Dye-Sensitized Solar Cell. We prepared the electrolyte by mixing 100 mL of ethylene glycol, 1.3 g of iodine, and 8.3 g of potassium iodide. We put the semiconducting electrode over the metallic electrode as shown in Figure 5 and dripped 200 μL of the electrolyte between the two electrodes as same as the original DSC as shown in Figure 1. Finally, we irradiated the electrodes with artificial sunlight (1000 W/m^2) and measured the DSC's I-V characteristics using a semiconductor characterization system (Keithley Instruments, Inc., 4200-SCS).

3.5. Evaluation of Our Paint-Type Dye-Sensitized Solar Cell. Generally, I-V characteristics (Figure 6) are checked for the evaluation of solar cells [38]. A short-circuit current (I_{SC}) is defined as the current when the voltage in the cell is 0 V. An open-circuit voltage (V_{OC}) is defined as the voltage when the current in the cell is 0 A. I_{max} is defined as the current when the generated power is as its maximum (P_{max}). V_{max} is defined as the voltage at that time.
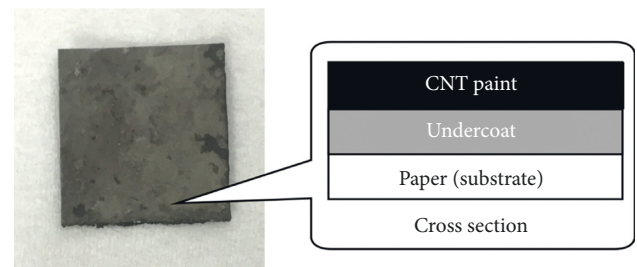

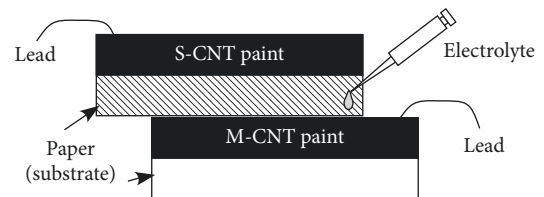FIGURE 4: Modified semiconducting electrode with undercoat using conductive silver paste.

FIGURE 5: Schematic of paint-type DSC for test. S-CNT indicates semiconducting CNT and M-CNT indicates metallic CNT.

By checking the I-V characteristics, the conversion efficiency (η), which is the ratio of the energy of the sunlight to the energy generated by the solar cell, can be found [38]. The conversion efficiency and the energy of the sunlight (P_{in}) are expressed respectively as
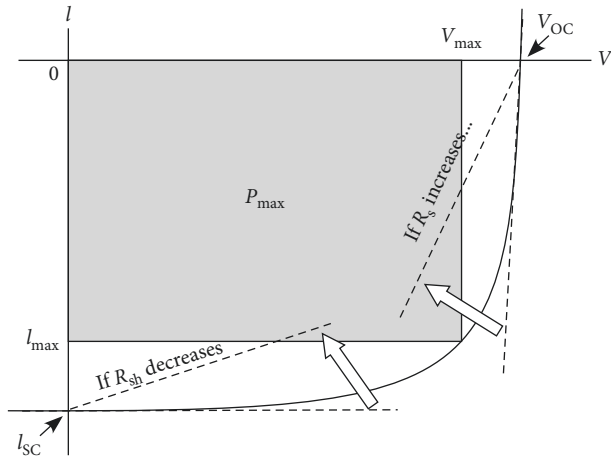
FIGURE 6: Ideal I-V characteristics of DSC.

$$\eta = \frac{P_{\max}}{P_{\mathrm{in}}} \times 100 \ [\%], \tag{1}$$

P_{in} = irradiance power × light receiving area.

The fill factor (FF) is defined as the ratio of the obtained I_{SC} and V_{OC} to the actual $P_{\max}$ (i.e., $I_{\max} \times V_{\max}$ as shown in Figure 6) [38]. FF is an index of the quality of the I-V characteristics of solar cells, which is expressed in equation (2), and has a value between 0 and 1:

$$\mathrm{FF} = \frac{P_{\max} (\mathrm{actual})}{P_{\max} (\mathrm{ideal})} = \frac{I_{\max} \times V_{\max}}{P_{\max} (\mathrm{ideal})}. \tag{2}$$

To obtain high FF, it is known that equivalent series resistance (R_{s}) of the solar cells including DSCs must be low (ideally 0) and equivalent shunt resistance (R_{sh}) must be high (ideally ∞) [38]. Figure 7 shows an equivalent circuit of solar cells, and it has resistances R_{s} and R_{sh}. It is known that R_{s} is caused by internal resistance of electrodes and electrolyte, and R_{sh} is caused by leakage current. Therefore, we should decrease the internal resistance and the leakage current.

4. Results and Discussion

We examined the characteristics of the following two kinds of samples: a DSC without an undercoat and a DSC with an undercoat. The resistance value was 24.6 Ω/□ (Ω/square) for the metallic electrode with 1 mL of the dispersion and 156 Ω/□ with 0.25 mL of the dispersion, 471 kΩ/□ for the semiconducting electrode without the undercoat, and 13 mΩ/□ for the semiconducting electrode with the undercoat, respectively. These results show that using a large amount of paint (dispersion) was effective to improve the conductivity of the metallic electrode and using the undercoat was also effective to improve the conductivity of our semiconducting electrode.

The I-V characteristics of our DSCs without and with the undercoat are shown in Figure 8. The conversion efficiency, FF, V_{OC}, and I_{SC} for the samples are shown in Table 1. We determined the power generated from the fabricated sample without the undercoat first (we used 24.6 Ω/□ of the sample
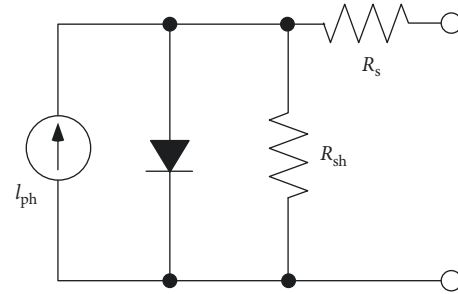

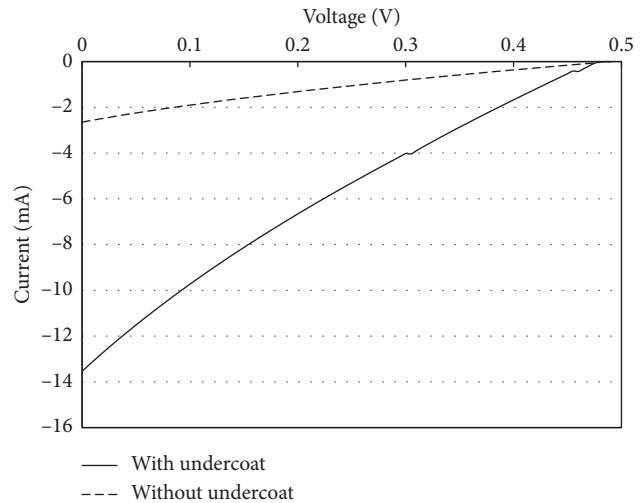FIGURE 7: Schematic of equivalent circuit of solar cell.

FIGURE 8: Measured I-V characteristics of our samples.

TABLE 1: Performance of our DSCs.

Sample	Type A	Type B
η (%)	0.12	0.60
FF	0.21	0.20
I_{SC} (mA)	2.64	13.5
V_{OC} (V)	0.48	0.49

Type A indicates semiconducting electrodes without the undercoat and type B indicates those with the undercoat.

for the metallic electrode because of its low resistance). We found that its conversion efficiency was 0.12%. Although this value was weak, our paint-type DSC had definitely generated power. We think our DSC can significantly improve its performance if we upgrade its structure. In fact, we found the semiconducting electrode with the undercoat effectively improved the performance of our DSC. Comparing the two types of our DSCs, that with the semiconducting electrodes without an undercoat and that with an undercoat, the I_{SC} of our DSC with the undercoat was about 5.1 times greater than of that without the undercoat. In other words, the undercoated DSC showed better performance than the DSC without the undercoat.

However, we were unable to improve the FF. Normally, the FF improves when the resistance value (R_{s}) of a sample decreases as described above. In this paper, the semiconducting electrode with the undercoat was painted only

on the one side that was irradiated with artificial sunlight. This meant that the other side (the back surface) that faced the metallic electrode consisted of just the semiconducting CNT and the dye. Currently, we assume that when the electrons of the dye on the surface of the electrode are excited, they move to the semiconducting CNT and reach the back surface before they reach the wire. Then, the electrons can move in reverse to the electrolyte. We think that because the resistance value of the back surface of the semiconducting electrode is poor, the electrons are not able to move into the semiconducting electrode and back to the electrolyte. That is, high leakage current may flow (R_{sh} may be small) and high internal resistance may be constructed (R_s may be large) in our DSC. We consider this to be the reason for the low FF. If we improve the FF of the sample with the undercoat to 0.5, maintaining I_{SC} and V_{OC}, we can obtain a conversion efficiency of 1.47%. If we improve the FF to 0.7, the conversion efficiency should be 2.06%. For this, we will determine a suitable painting process in the near future because our paint-type DSC can be made using any combination of the paints as described previously. For aiming high FF, we prepared aluminium foil as a substrate instead of the paper substrate for the semiconducting electrode as a new test. That is, we painted our semiconducting CNT paint with dye on the aluminium foil. Then, we prepared the metallic CNT electrode based on the paper substrate and put new semiconducting electrode on the metallic electrode. After that, we irradiated the electrodes with artificial sunlight and measured the DSC's I-V characteristics. Figure 9 shows the result. From this result, we confirmed this sample obtained 0.40 of FF. We consider there are two reasons for this FF improvement. One is the conductivity of the substrate (electrode). Therefore, one of our approaches, that is, the undercoat, is expected to be effective. Second is flatness of the substrate surface. Comparing with the paper substrate, the aluminium foil substrate surface is so smooth. For this, we will study the details in near future.

5. Conclusion

We proposed a paint-type DSC. It has two electrodes made through painting. Our DSCs are made by using CNT paints. First, we confirmed that power was generated from our fabricated DSCs. Although the first sample showed power generation, its efficiency was not very high. Aiming to improve it, we used a semiconducting electrode with an undercoat. We found this type of our DSC had better efficiency. Finally, we considered the difference between the efficiency of our DSCs' two structures. The DSC without the undercoat exhibited much higher performance, particularly in I_{SC}, than that with the undercoat. This was because the sample with the undercoat had a resistance value much smaller than that of the sample without the undercoat. However, we were unable to obtain high FF. The reason may be that high leakage current may flow and high internal resistance may be constructed in our DSC. To take a hint to overcome the problems, we prepared and tested new sample that used the aluminium foil for the semiconducting electrode. Then, the sample showed higher FF than the previous
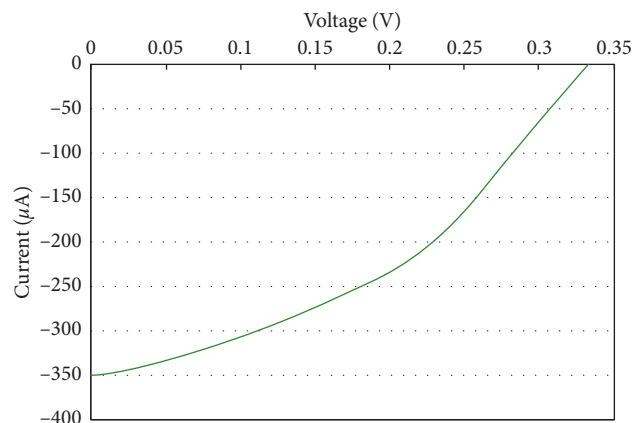

Figure 9: Measured I-V characteristics of new sample.

ones. In the near future, we will be able to overcome the problems. Moreover, our paint-type DSC will be able to be applied to any objects or places, such as wallpaper, window glass, and furniture, where paint can be used.

Data Availability

The data described in the manuscript are available from the corresponding author on reasonable request.

Conflicts of Interest

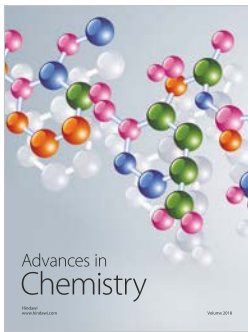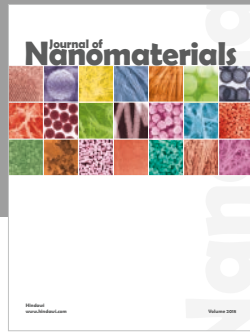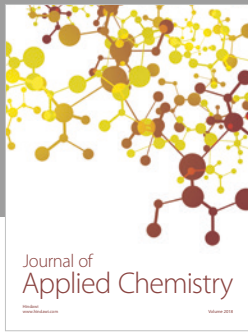The authors declare that they have no conflicts of interest.

Acknowledgments

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