# Dye-Sensitized Solar Cells Using Natural Dyes and Nanostructural Improvement of $TiO_2$ Film

**Shoji Furukawa**

**Abstract** The characteristics of the dye-sensitized solar cells using natural dyes, such as those of red-cabbage, curcumin, and red-perilla, and synthesized dyes, such as NKX-2553, NKX-2677, and D149, in which a precious metal is not contained, will be explained. The largest conversion efficiency obtained is over 1% for the dye-sensitized solar cell using the dye of red-cabbage. This value is relatively small. However, the cost performance (defined by [conversion efficiency]/[cost of dye]) is more than 50 times greater than that of the dye-sensitized solar cell using Ruthenium complex. Therefore, when the cost of FTO and ITO substrates, oxide semiconductor, electrolyte solution, and opposite electrode becomes very low, dye-sensitized solar cells fabricated using natural dyes may become more pervasive, although the physical device becomes larger than one using a Ruthenium complex. The effects of pH of the dye solution on the characteristic of the dye-sensitized solar cells will be also described. The conversion efficiencies of the dye-sensitized solar cells using the dye of red-cabbage, red-perilla, NKX-2553, and NKX-2677 become larger when the pH value is low. It is expected that this technique will be used in future dye-sensitized solar cell systems.

## 1 Introduction

Clean energy has become a focal point for many researchers. Among the clean energy, solar cells are quite promising device because their cost is relatively low. However, traditional crystalline silicon solar cells require much energy in order to

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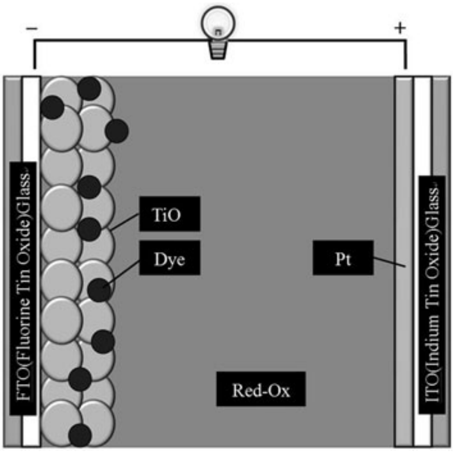


**Fig. 1** Fundamental structure of dye-sensitized solar cell

fabricate the wafers. Therefore, recently, dye-sensitized solar cells are expected as cheap energy conversion devices [1, 2].

Most researchers working in the field of dye-sensitized solar cells are using a Ruthenium complex as the dye because such devices can have a high conversion efficiency. To date, the highest conversion efficiency obtained experimentally is over 12%. However, Ruthenium is a rare metal, so its cost is very high.

Our group has fabricated various kinds of dye-sensitized solar cells using natural dye, such as those of red-cabbage, curcumin, and red-perilla [3–5]. The cost of these natural dyes is very low. The largest conversion efficiency of dye-sensitized solar cells using these dyes is about 1%, which is smaller than solar cells fabricated using a Ruthenium complex. However, the cost performance (defined by [conversion efficiency]/[cost of dye]) of the dye-sensitized solar cell using the dye of red-cabbage is more than 50 times larger than that of the dye-sensitized solar cell fabricated using a Ruthenium complex. Therefore, when the cost of FTO substrate, oxide semiconductor, electrolyte solution, and opposite electrode becomes very low, dye-sensitized solar cells using natural dyes may be a more cost effective solution, although such devices will be physically larger than those using a Ruthenium complex.

In this chapter, the characteristics of the dye-sensitized solar cells using natural dyes and synthesized dyes in which precious metal is not contained are explained. The effects of pH of the dye solution on the characteristic of the dye-sensitized solar cell are also described.

## 2 Structure of Dye-Sensitized Solar Cell

Figure 1 shows the fundamental structure of a dye-sensitized solar cell. A glass sheet coated by the transparent and electrically conductive film (FTO) is used as the cathode. On the cathode, an oxide semiconductor film, such as $TiO_2$, is coated and the dye is adsorbed onto its surface. The anode is prepared by sputtering Pt film onto another ITO substrate. Then, the electrolyte solution is inserted between the cathode and anode.

When sunlight irradiates the cathode substrate, most of it propagates to the dye. This is because glass is transparent to visible light, and the band gap of the oxide semiconductor is large. Then, the electronic states of the electrons in the dye molecules jumps to higher levels due to energy absorbed from the sunlight. The excited electrons transfer to the conduction band of the oxide semiconductor, then move through the transparent oxide to the cathode. After arriving at the conducting film on the anode, the electrons move to the dye surface through the iodine ions in the electrolyte solution and back to the dye, creating electric current and completing the circuit.

## 3 Fabrication Process for Dye-Sensitized Solar Cells

In order to fabricate the dye-sensitized solar cells, an oxide semiconductor paste was prepared [6–11]. In our laboratory, two kinds of $TiO_2$ were used in fabrication of the paste: P-25, with an average particle size of 26 nm, and PC-101, with an average particle size of 20 nm. The two kinds of $TiO_2$, P-25 (0.14 g) and PC-101 (0.06 g), were first placed in a shaker with $HNO_3$ (0.6 ml). Then, polyethylene glycol (PEG) was also added to the shaker in order to obtain a porous oxide semiconductor film. The characteristics of the porous oxide semiconductor film depend on the molecular weight of PEG. In our work, two kinds of PEGs were used: PEG with a molecular weight of 500,000 and PEG with a molecular weight of 2,000,000. The $TiO_2$ paste was coated onto the FTO glass substrate, which had been cleaned by ethyl alcohol and acetone. Then, the substrate was placed in an electric heater, and annealed at 450°C for 1 h.

After depositing the porous oxide semiconductor film onto the FTO glass substrate, the dye was adsorbed onto the oxide semiconductor film surface by dipping the substrate into the dye solution. In our study, six types of dyes were used. They were dyes of red-cabbage, curcumin, red-perilla, NKX-2553, NKX-2677, and D149. The former three dyes are natural and the latter three are synthetic. All dyes contain no precious metal. The chemical structures of the natural dyes are shown in Fig. 2, and the molecular structures of NKX-2553, NKX-2677, and D149 are shown in Fig. 3. The molar density of the red-cabbage, curcumin, and red-perilla dye solutions was 0.5 mM. The solvent of the solutions of red-cabbage and red-perilla was water, and the solvent for the curcumin solution was ethyl alcohol.

The electrolyte solution was prepared using $I_2$, LiI, 1-propyl-2,3 dimethylimidazolium iodide (DMPImI), 4-*tert*-butyl pyridine (TBP), and propylene carbonate (PC). The weights of $I_2$, LiI, DMPImI, TBP, and PC were 0.0317, 0.330, 0.342, 0.189, and 3.00 g, respectively. The electrolyte solution was inserted between the two electrodes.

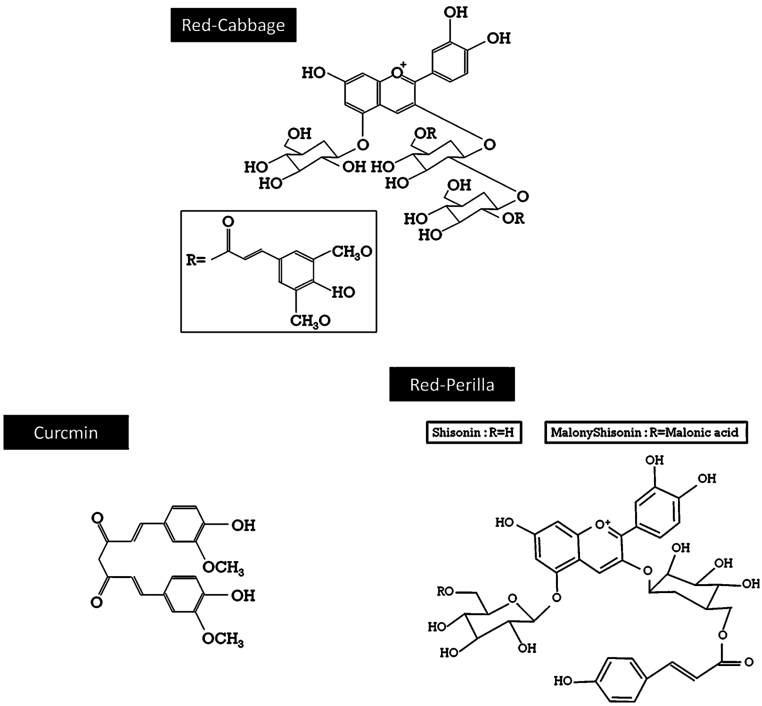


**Fig. 2** Chemical structures of the dyes from red-cabbage, curcumin, and red-perilla

Photocurrent-voltage characteristics of the dye-sensitized solar cells were measured using a halogen lamp as a light source. The power of the light source was 50 mW/cm$^2$. The conversion efficiency obtained using the halogen lamp was about 5% larger than that obtained while using the standard light source that was constructed in our laboratory. In our standard light source system, visible and infrared components were partly eliminated by an optical filter.

## 4 Characteristics of Dye-Sensitized Solar Cells Using Natural Dyes

### *4.1 Characteristics of Dye-Sensitized Solar Cells using Dye of Red-Cabbage*

Figure 4 shows the photocurrent-voltage characteristics of the dye-sensitized solar cells using the dye of red-cabbage. Five solar cells have been fabricated, and the numbers in Fig. 4 correspond to the sample number. In this case, the oxide semiconductor paste was prepared using the PEG with the molecular weight of 500,000.

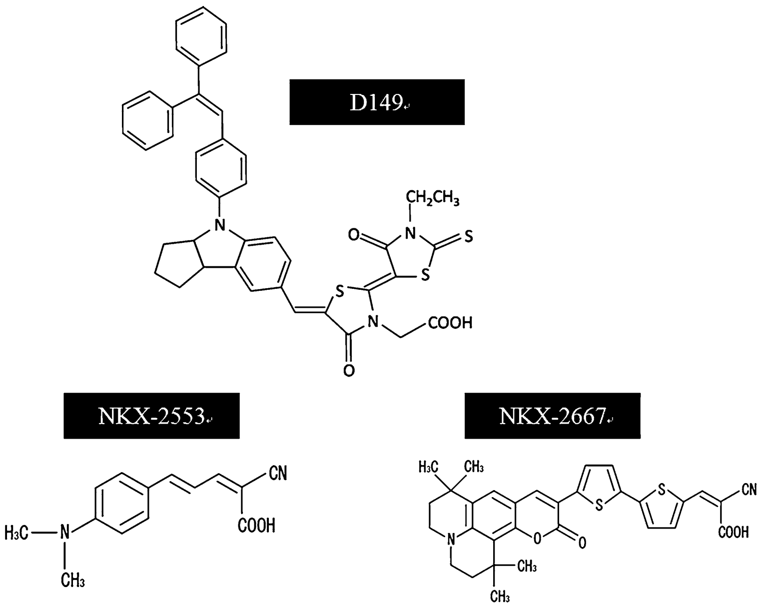


**Fig. 3** Molecular structures of NKX-2553, NKX-2677, and D149

**Fig. 4** Photocurrent-voltage characteristics of the dye-sensitized solar cells using dye of red-cabbage. The numbers correspond to the sample number. The oxide semiconductor paste was fabricated using the PEG whose molecular weight was 50,000

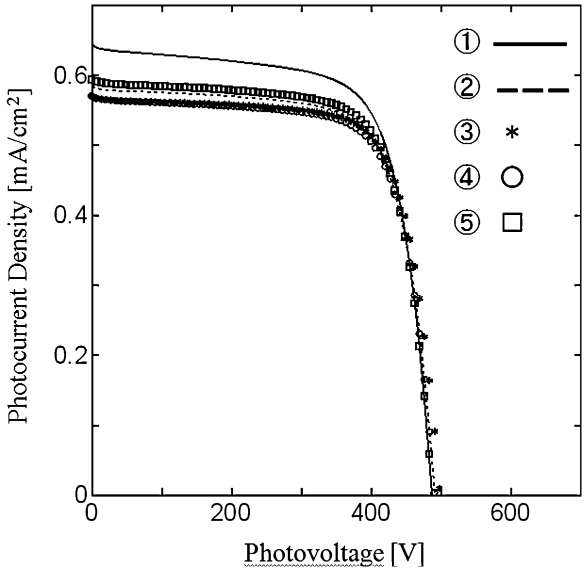


Table 1 shows the summary of the characteristics of the solar cells fabricated using the dye of red-cabbage (molecular weight of PEG is 500,000). The average fill factor was 0.72, and the average open circuit voltage was 0.49 V. In this case, the average conversion efficiency was 0.42%.

**Table 1** Summary of the characteristics of the solar cells fabricated using dye of red-cabbage and the PEG with the molecular weight of 500,000

| Sample number | $J_{SC}$ (mA/cm$^2$) | $V_{OC}$ (V) | $P_{MAX}$ (mW) | F.F. | $\eta$ (%) |
|---|---|---|---|---|---|
| Sample 1 | 0.64 | 0.48 | 0.22 | 0.70 | 0.44 |
| Sample 2 | 0.59 | 0.49 | 0.20 | 0.71 | 0.41 |
| Sample 3 | 0.57 | 0.50 | 0.20 | 0.72 | 0.41 |
| Sample 4 | 0.57 | 0.49 | 0.20 | 0.72 | 0.40 |
| Sample 5 | 0.59 | 0.48 | 0.21 | 0.72 | 0.42 |
| Average | 0.59 | 0.49 | 0.21 | 0.72 | 0.42 |

$J_{SC}$ short circuit current density; $V_{OC}$ open circuit voltage; $P_{MAX}$ maximum power; *F.F.* fill factor; $\eta$ conversion efficiency

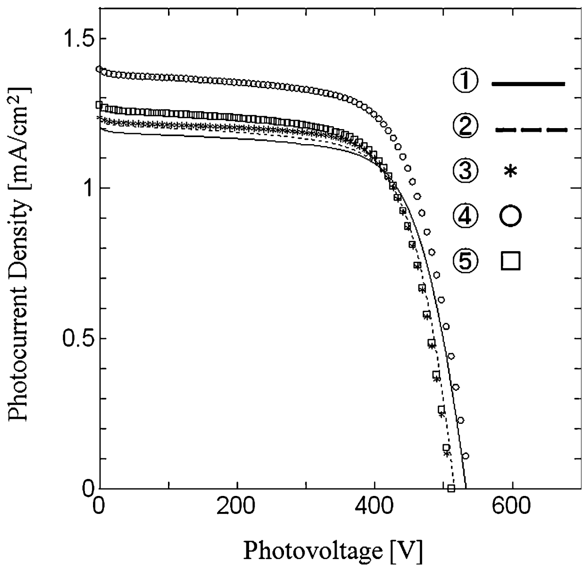


**Fig. 5** Photocurrent-voltage characteristics of the dye-sensitized solar cells using dye of red-cabbage. The numbers written in the figure are the sample numbers. In this case, the oxide semiconductor paste was fabricated using the PEG whose molecular weight was 2,000,000

Similar to Figs. 4, 5 show the photocurrent-voltage characteristics of the dye-sensitized solar cells using the dye of red-cabbage. In this case, the oxide semiconductor paste was fabricated using the PEG whose molecular weight was 2,000,000. Five solar cells were fabricated and the numbers written in Fig. 5 correspond to the sample number.

Table 2 shows the summary of the characteristics of the solar cells fabricated using the dye of red-cabbage (molecular weight of PEG is 2,000,000). In this case, the average short circuit current density is approximately twice that of the device fabricated with the PEG with the smaller molecular weight. The average conversion efficiency also doubled to 0.90%.

In order to investigate the difference in conversion efficiencies between the two devices, the oxide semiconductor surfaces were characterized by scanning electron microscopy (SEM). This study showed that the pores were uniformly dispersed in the oxide semiconductor film prepared using the PEG with a molecular weight of

**Table 2** Summary of the characteristics of the solar cells fabricated using dye of red-cabbage and the PEG whose molecular weight is 2,000,000

| Sample number | $J_{SC}$ (mA/cm$^2$) | $V_{OC}$ (V) | $P_{MAX}$ (mW) | F.F. | $\eta$ (%) |
|---|---|---|---|---|---|
| Sample 1 | 1.2 | 0.53 | 0.44 | 0.68 | 0.87 |
| Sample 2 | 1.2 | 0.51 | 0.44 | 0.69 | 0.87 |
| Sample 3 | 1.2 | 0.50 | 0.44 | 0.70 | 0.88 |
| Sample 4 | 1.4 | 0.53 | 0.50 | 0.67 | 1.00 |
| Sample 5 | 1.3 | 0.51 | 0.44 | 0.68 | 0.89 |
| Average | 1.3 | 0.52 | 0.45 | 0.69 | 0.90 |

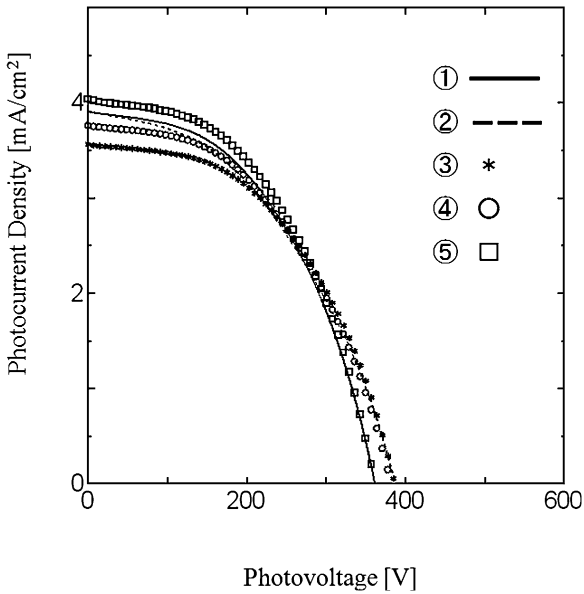


**Fig. 6** Photocurrent-voltage characteristics of the dye-sensitized solar cells using dye of red-cabbage with an improved electrolyte solution

2,000,000. On the other hand, some large particles were observed in the oxide semiconductor film prepared using the PEG with a molecular weight of 500,000. This change in morphology is hypothesized to be the cause of the difference in efficiencies between the two devices.

In order to improve the characteristics of the dye-sensitized solar cells using the dye of red-cabbage, the electrolyte solution was changed [12]. The photocurrent-voltage characteristics of the solar cells fabricated using the dye of red-cabbage are shown in Fig. 6. Table 3 shows the summary of the characteristics of these devices (the pH is controlled by adding citric acid) with the improved electrolyte solution. The highest conversion efficiency was approximately 1.4%. The improved electrolyte solution is considered to be the cause of the improved performance.

**Table 3** Summary of the characteristics of the dye-sensitized solar cells fabricated using red-cabbage dye solutions (whose pH is controlled by adding citric acid) with improved electrolyte solution

| Sample number | $J_{SC}$ (mA/cm$^2$) | $V_{OC}$ (V) | $P_{MAX}$ (mW) | F.F. | $\eta$ (%) |
|---|---|---|---|---|---|
| Sample 1 | 3.9 | 0.39 | 0.66 | 0.44 | 1.3 |
| Sample 2 | 4.1 | 0.36 | 0.71 | 0.49 | 1.4 |
| Sample 3 | 3.6 | 0.39 | 0.67 | 0.49 | 1.3 |
| Sample 4 | 3.9 | 0.36 | 0.68 | 0.48 | 1.4 |
| Sample 5 | 3.8 | 0.38 | 0.68 | 0.47 | 1.4 |
| Average | 3.9 | 0.37 | 0.68 | 0.47 | 1.4 |

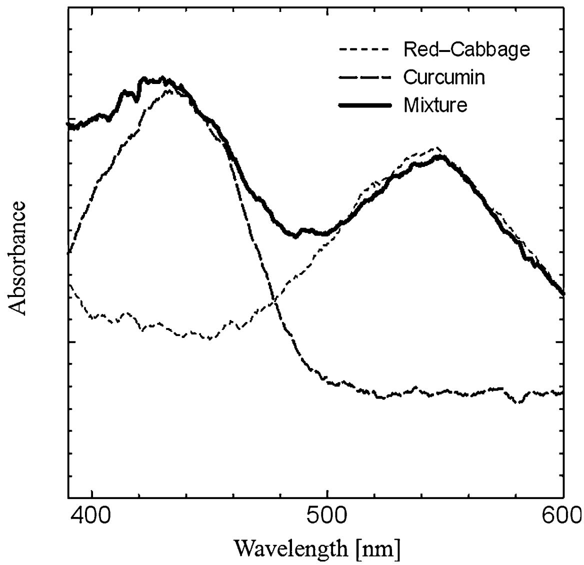


**Fig. 7** Optical absorption spectra of the red-cabbage, curcumin, and mixed dye of red-cabbage and curcumin solutions

## *4.2 Characteristics of Dye-Sensitized Solar Cells Using Mixed Dye of Red-Cabbage and Curcumin*

Figure 7 shows the optical absorption spectra of the red-cabbage, curcumin, and the mixture of the two dyes in solution. As shown in Fig. 7, the dyes of red-cabbage and curcumin have absorption peaks at about 550 and 440 nm, respectively. When the both dyes are mixed, optical absorption occurs in wide band, as shown in Fig. 7. Therefore, it is expected that the conversion efficiency of the solar cell increases when using the dye mixture.

Figure 8 shows the photocurrent-voltage characteristics of the dye-sensitized solar cells fabricated using the dye of red-cabbage, curcumin, and mixed dye of red-cabbage and curcumin. As shown in Fig. 8, the characteristics of the dye-sensitized solar cell using the mixed dye of red-cabbage and curcumin is better than those fabricated using the dye of red-cabbage or curcumin.

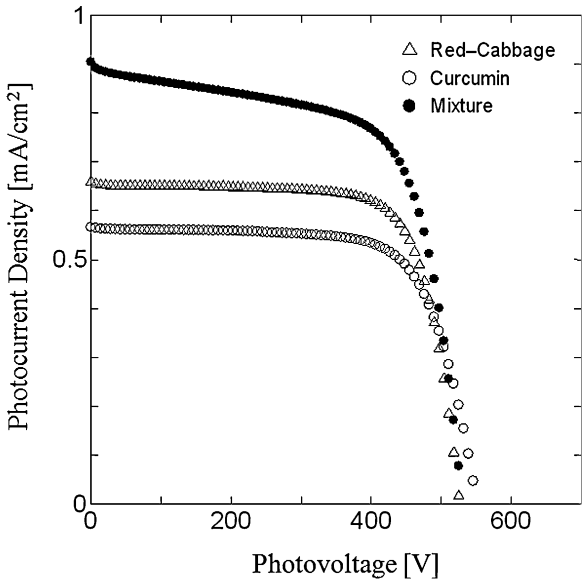


**Fig. 8** Photocurrent-voltage characteristics of the dye-sensitized solar cells using the dye of red-cabbage, curcumin, and mixed dye of red-cabbage and curcumin

**Table 4** Summary of the characteristics of the dye-sensitized solar cells fabricated using the dye of red-cabbage, curcumin, and mixed dye of red-cabbage and curcumin

| Red-cabbage:curcumin | $J_{SC}$ (mA/cm$^2$) | $V_{OC}$ (V) | $P_{MAX}$ (mW) | F.F. | $\eta$ (%) |
|---|---|---|---|---|---|
| 1:0 | 0.69 | 0.52 | 0.25 | 0.70 | 0.50 |
| 100:1 | 0.73 | 0.52 | 0.27 | 0.71 | 0.53 |
| 90:1 | 0.78 | 0.50 | 0.27 | 0.69 | 0.54 |
| 80:1 | 0.78 | 0.50 | 0.27 | 0.69 | 0.54 |
| 70:1 | 0.81 | 0.53 | 0.30 | 0.69 | 0.60 |
| 60:1 | 0.74 | 0.52 | 0.27 | 0.71 | 0.54 |
| 50:1 | 0.74 | 0.50 | 0.25 | 0.69 | 0.51 |
| 0:1 | 0.53 | 0.53 | 0.20 | 0.72 | 0.41 |

Table 4 shows the summary of the characteristics of the dye-sensitized solar cells using the dyes of red-cabbage, curcumin, and the mixture. The conversion efficiency of the dye-sensitized solar cell using the mixed dye of red-cabbage and curcumin becomes greater than those of the solar cells fabricated using the dye of red-cabbage or curcumin.

Our recent data showed a large conversion efficiency of about 1.3% for the dye-sensitized solar cell fabricated using the dye of curcumin with the improved electrolyte solution [12].

## *4.3 Characteristics of Dye-Sensitized Solar Cells Using Dye of Red-Perilla*

Table 5 shows the characteristics of the dye-sensitized solar cells using the dye of the red-perilla. The average conversion efficiency was 0.17%, which is smaller

**Table 5** Characteristics of the dye-sensitized solar cells fabricated using the dye of red-perilla

| Sample number | $J_{SC}$ (mA/cm$^2$) | $V_{OC}$ (V) | $P_{MAX}$ (mW) | F.F. | $\eta$ (%) |
|---|---|---|---|---|---|
| Sample 1 | 0.20 | 0.48 | 0.06 | 0.63 | 0.12 |
| Sample 2 | 0.27 | 0.48 | 0.09 | 0.68 | 0.17 |
| Sample 3 | 0.33 | 0.48 | 0.10 | 0.65 | 0.21 |
| Sample 4 | 0.24 | 0.53 | 0.09 | 0.70 | 0.18 |
| Sample 5 | 0.26 | 0.50 | 0.09 | 0.68 | 0.18 |
| Average | 0.26 | 0.49 | 0.09 | 0.67 | 0.17 |

**Table 6** Summary of the characteristics of the dye-sensitized solar cells using NKX-2553

| Sample number | $J_{SC}$ (mA/cm$^2$) | $V_{OC}$ (V) | $P_{MAX}$ (mW) | F.F. | $\eta$ (%) |
|---|---|---|---|---|---|
| Sample 1 | 4.2 | 0.63 | 1.5 | 0.56 | 3.0 |
| Sample 2 | 4.3 | 0.63 | 1.5 | 0.54 | 2.9 |
| Sample 3 | 4.3 | 0.63 | 1.5 | 0.54 | 2.9 |
| Sample 4 | 4.3 | 0.63 | 1.5 | 0.55 | 3.0 |
| Sample 5 | 4.7 | 0.62 | 1.6 | 0.54 | 3.2 |
| Average | 4.4 | 0.63 | 1.5 | 0.55 | 3.0 |

than that of the solar cell fabricated using the dye of the red-cabbage. As shown in Fig. 2, the chemical structure of the main part of the red-perilla dye is the same as that of the red-cabbage dye. However, the chemical structure of other part is different. The OH groups in the dyes are believed to react with the OH groups on the oxide semiconductor film surface. This reaction causes $H_2O$ and covalent bonding between the dye and the oxide semiconductor via oxygen atom. The number of OH groups of the dye of red-perilla is smaller than that of the dye of red-cabbage (see Fig. 2). This may cause a lower conversion efficiency of the solar cell using the dye of red-perilla.

## 5 Characteristics of Dye-Sensitized Solar Cells Using Synthetic Dyes

In this section, the characteristics of dye-sensitized solar cells using the synthetic dyes containing no precious metal, such as Ruthenium, will be presented.

### *5.1 Characteristics of Dye-Sensitized Solar Cells Using NKX-2553*

Table 6 shows the summary of the characteristics of the dye-sensitized solar cells using NKX-2553. Five solar cells were fabricated. The average conversion efficiency obtained was 3.0%, which is larger than that of the solar cells fabricated

**Table 7** Summary of the characteristics of the dye-sensitized solar cells using NKX-2677

| Sample number | $J_{SC}$ (mA/cm$^2$) | $V_{OC}$ (V) | $P_{MAX}$ (mW) | F.F. | $\eta$ (%) |
|---|---|---|---|---|---|
| Sample 1 | 5.9 | 0.61 | 1.8 | 0.51 | 3.7 |
| Sample 2 | 5.7 | 0.61 | 1.8 | 0.51 | 3.5 |
| Sample 3 | 5.8 | 0.61 | 1.8 | 0.51 | 4.0 |
| Sample 4 | 6.0 | 0.60 | 1.7 | 0.47 | 3.4 |
| Sample 5 | 5.4 | 0.61 | 1.8 | 0.55 | 3.7 |
| Average | 5.8 | 0.61 | 1.8 | 0.51 | 3.6 |

using natural dyes. However, the cost of the synthesized dyes is high, so the cost performances of the dye-sensitized solar cells fabricated using synthetic dyes is smaller than those of the solar cells fabricated using natural dyes.

### *5.2 Characteristics of Dye-Sensitized Solar Cells Using NKX-2677*

Table 7 shows the summary of the characteristics of the dye-sensitized solar cells using NKX-2677. Five solar cells were fabricated. The average conversion efficiency obtained was 3.6%, again larger than those found in solar cells fabricated using natural dyes or NKX-2553. Similar to the solar cell using NKX-2553, the cost performance is smaller than those of the solar cells fabricated using natural dyes.

### *5.3 Characteristics of Dye-Sensitized Solar Cells using Mixture of NKX-2553 and D149*

Figure 9 shows the optical absorption spectra of NKX-2553, D149, and the mixed dye of NKX-2553 and D149. The ratio of the mixture is 1:1. The vertical axis indicates normalized absorbance.

As shown in Fig. 9, NKX-2553 and D149 have absorption peaks at about 450 nm and 540 nm, respectively. By mixing NKX-2553 and D149, the absorption in the range from about 400 to 500 nm increases, suggesting an increase of conversion efficiency of the solar cell.

In our study, the FTO substrate on which the oxide semiconductor film was fabricated was dipped into the dye solution, and various dye-sensitized solar cells were fabricated by changing dipping time. Figure 10 shows the relation between conversion efficiency and dipping time obtained at 12, 40, 60, and 80°C. The ratio of the mixture of NKX-2553 and D149 is 1:1. As shown in Fig. 10, the maximum conversion efficiency was obtained at a short dipping time when the dipping temperature was high. However, the largest conversion efficiency was obtained at a low temperature of 12°C. The largest conversion efficiency was 5.4%.

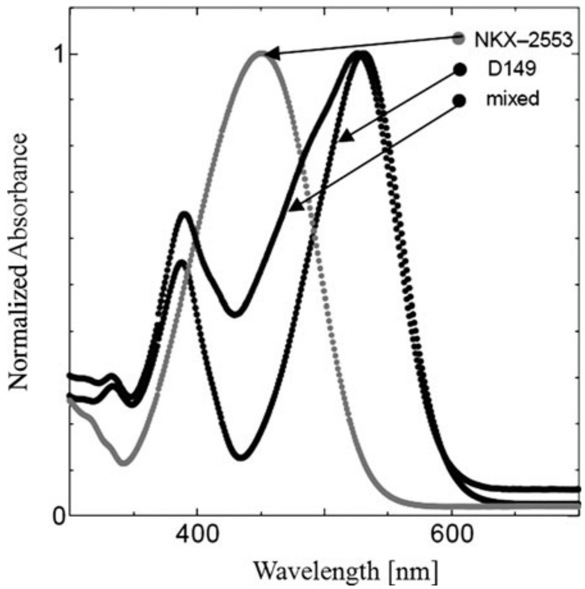


**Fig. 9** Optical absorption spectra of NKX-2553, D149, and the mixed dye of NKX-2553 and D149. The ratio of the mixture is 1:1. The vertical axis indicates normalized absorbance

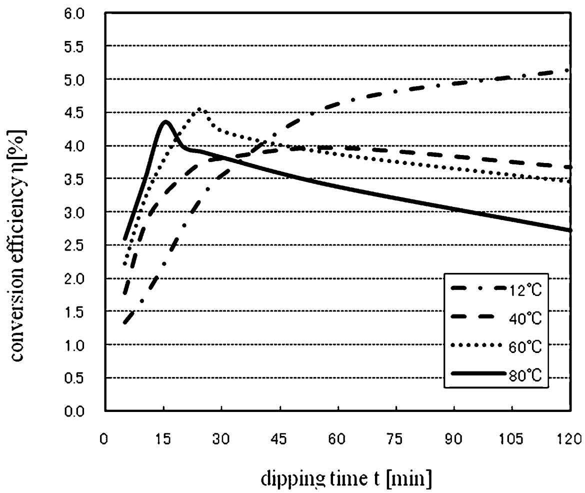


**Fig. 10** Relation between conversion efficiency and dipping time obtained at 12, 40, 60, and 80°C. The ratio of the mixture of NKX-2553 and D149 is 1:1

# 6 Effects of pH of Dyes on Characteristics of Dye-Sensitized Solar Cells

## *6.1 Effects of pH of Dye on Characteristics of Dye-Sensitized Solar Cells Using Dye of Red-Cabbage*

Figure 11 shows the optical absorption spectra of various red-cabbage dye solutions in which the pH is controlled by adding citric acid or sodium hydroxide.

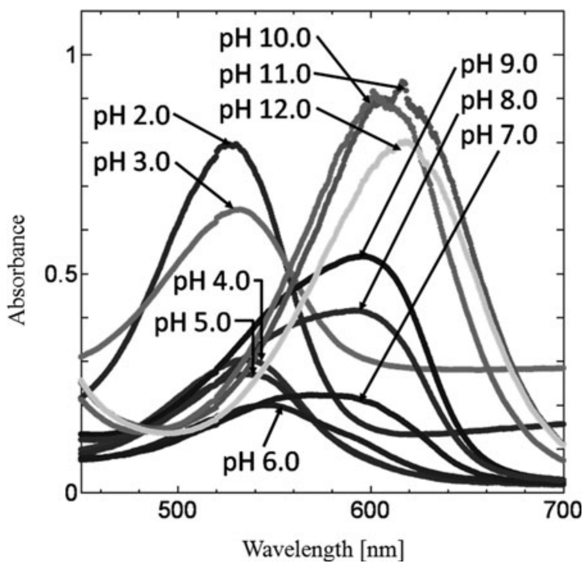


**Fig. 11** Optical absorption spectra of various red-cabbage dye solutions whose pH is controlled by adding citric acid or sodium hydroxide

**Table 8** Summary of the characteristics of the dye-sensitized solar cells fabricated using various red-cabbage dye solutions in which the pH is controlled by adding citric acid or sodium hydroxide

| pH | $J_{SC}$ (mA/cm$^2$) | $V_{OC}$ (V) | $P_{MAX}$ (mW) | F.F. | $\eta$ (%) |
|---|---|---|---|---|---|
| 2.0 | 2.7 | 0.38 | 0.56 | 0.56 | 1.1 |
| 3.0 | 2.4 | 0.39 | 0.52 | 0.57 | 1.0 |
| 4.0 | 2.1 | 0.39 | 0.49 | 0.59 | 0.97 |
| 5.0 | 2.0 | 0.40 | 0.44 | 0.55 | 0.89 |
| 6.0 | 1.4 | 0.42 | 0.34 | 0.59 | 0.68 |
| 7.0 | 0.92 | 0.42 | 0.22 | 0.57 | 0.45 |
| 8.0 | 0.91 | 0.42 | 0.22 | 0.57 | 0.43 |
| 9.0 | 0.96 | 0.42 | 0.25 | 0.61 | 0.50 |
| 10.0 | 0.93 | 0.44 | 0.26 | 0.63 | 0.51 |
| 11.0 | 0.38 | 0.47 | 0.09 | 0.50 | 0.19 |
| 12.0 | 0.15 | 0.44 | 0.03 | 0.45 | 0.06 |

In the case of pH = 7.0, no addition was made. As shown in Fig. 11, the strength of the absorption peak became increased when pH was 2.0, 3.0, 4.0, 5.0, 8.0, 9.0, 10.0, 11.0, and 12.0. When pH was low, the strength of the peak at about 530 nm increased, whereas when pH was high, the strength of the peak at about 610 nm increased. Therefore, it is expected that the conversion efficiency will increase by adding citric acid or sodium hydroxide into the dye solution.

Table 8 shows the summary of the characteristics of the dye-sensitized solar cells fabricated using various red-cabbage dye solutions whose pH is controlled by adding citric acid or sodium hydroxide. The largest conversion efficiency (1.1%) was obtained when pH was 2.0.

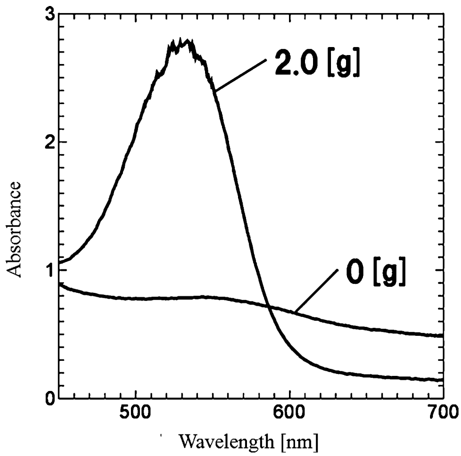


**Fig. 12** Optical absorption spectra of the red-perilla dye solutions with unmodified pH and a pH controlled by adding citric acid (2 g)

According to the chemical structure shown in Fig. 2, the basic structures of the dyes of red-cabbage and red-perilla are both anthocyanin. As shown in Fig. 2, there are three OH groups in the fundamental anthocyanin structure. However, one of the OH groups changes to oxygen (double bond) under neutral conditions. As discussed in "Nanomorphology and charge transport in organic solar cells", it is considered that these OH groups in the dyes react with the OH groups which terminate the oxide semiconductor film surface. This reaction causes the strong covalent bond between the dye and the oxide semiconductor. In the acidic case, there are three OH groups in the fundamental structure. This may cause an increase in the number of the strong covalent bond between the dye and the oxide semiconductor, resulting in an increase of the conversion efficiency.

## 6.2 *Effects of pH of Dye on Characteristics of Dye-Sensitized Solar Cells Using Dye of Red-Perilla*

Figure 12 shows the optical absorption spectra of the red-perilla dye solutions with unmodified pH and a pH modified by adding citric acid (2 g). The strength of the absorption peak at around 530–540 nm is very much increased by adding citric acid.

Table 9 shows the summary of the characteristics of the dye-sensitized solar cells fabricated using red-perilla dye solutions with modified pH. The average conversion efficiency is 0.27% which is ~60% larger than that of solar cells fabricated using the red-perilla dye solution with an unmodified pH (0.17%, see Table 5). This increase of the conversion efficiency is proposed to be caused by an increase of the optical absorption strength shown in Fig. 12.

**Table 9** Summary of the characteristics of the dye-sensitized solar cells fabricated using red-perilla dye solutions whose pH is controlled by adding citric acid (2 g)

| Sample number | $J_{SC}$ (mA/cm$^2$) | $V_{OC}$ (V) | $P_{MAX}$ (mW) | F.F. | $\eta$ (%) |
|---|---|---|---|---|---|
| Sample 1 | 0.39 | 0.50 | 0.13 | 0.68 | 0.26 |
| Sample 2 | 0.40 | 0.51 | 0.14 | 0.67 | 0.27 |
| Sample 3 | 0.40 | 0.51 | 0.14 | 0.67 | 0.27 |
| Sample 4 | 0.43 | 0.48 | 0.14 | 0.68 | 0.28 |
| Sample 5 | 0.40 | 0.48 | 0.13 | 0.69 | 0.26 |
| Average | 0.40 | 0.50 | 0.13 | 0.68 | 0.27 |

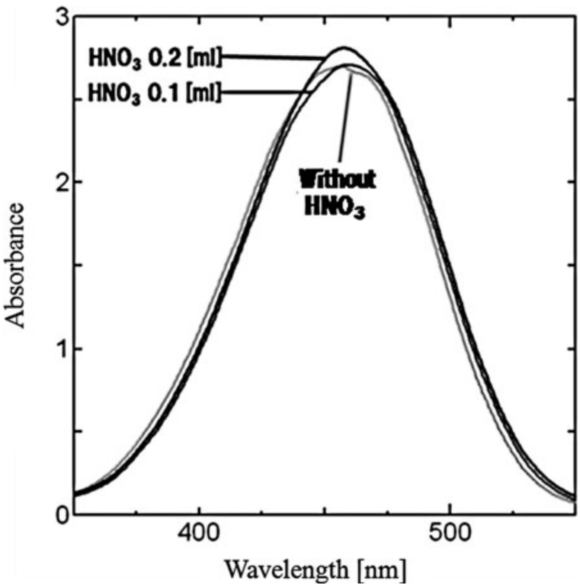


**Fig. 13** Optical absorption spectra of NKX-2553 solutions with and without $HNO_3$. The former specimen was prepared by adding $NHO_3$ (0.1 or 0.2 ml) into NKX-2553 solution (25 ml)

## *6.3 Effects of pH of Dye on Characteristics of Dye-Sensitized Solar Cells Using NKX-2553*

Figure 13 shows the optical absorption spectra of NKX-2553 solutions with and without $HNO_3$. The former solutions were prepared by adding $NHO_3$ (0.1 or 0.2 ml) into NKX-2553 solution (25 ml). As shown in Fig. 13, the strength of the absorption peak at about 460 nm increased slightly by adding 0.2 ml of $HNO_3$.

Table 10 shows the summary of the characteristics of the dye-sensitized solar cells fabricated using NKX-2553 with and without $HNO_3$ (0.1 ml). Five solar cells were fabricated. The average conversion efficiency is 3.3% which is larger by 0.3 point (about 9%) than that of the solar cells fabricated using NKX-2553 without $HNO_3$ (see Table 6). The reason for this increase of the conversion efficiency is not clear. However, it may be related to a small increase of the optical absorption strength at about 460 nm shown in Fig. 13.

**Table 10** Summary of the characteristics of the dye-sensitized solar cells fabricated using NKX-2553 with and without $HNO_3$ (0.1 ml)

| Sample number | $J_{SC}$ (mA/cm$^2$) | $V_{OC}$ (V) | $P_{MAX}$ (mW) | F.F. | $\eta$ (%) |
|---|---|---|---|---|---|
| Sample 1 | 4.7 | 0.62 | 1.6 | 0.55 | 3.2 |
| Sample 2 | 4.9 | 0.64 | 1.7 | 0.54 | 3.3 |
| Sample 3 | 4.9 | 0.64 | 1.7 | 0.54 | 3.4 |
| Sample 4 | 4.3 | 0.64 | 1.5 | 0.55 | 3.0 |
| Sample 5 | 5.0 | 0.62 | 1.7 | 0.55 | 3.4 |
| Average | 4.8 | 0.63 | 1.6 | 0.55 | 3.3 |

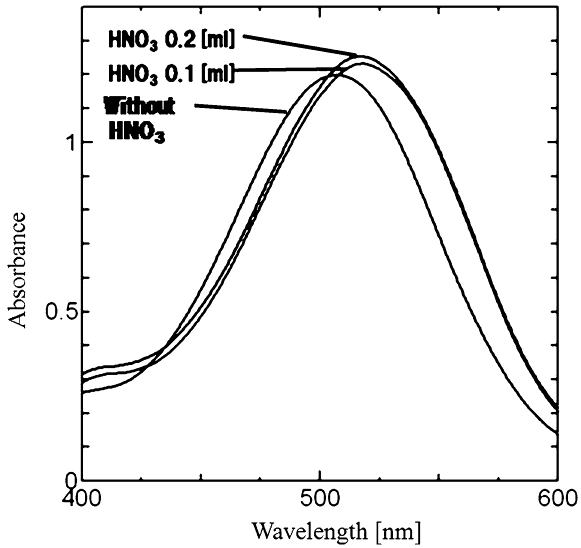


**Fig. 14** Optical absorption spectra of NKX-2677 solutions with and without $HNO_3$. The former specimen was prepared by adding $NHO_3$ (0.1 or 0.2 ml) into NKX-2677 solution (25 ml)

## *6.4 Effects of pH of Dye on Characteristics of Dye-Sensitized Solar Cells Using NKX-2677*

Figure 14 shows the optical absorption spectra of NKX-2677 solutions with and without $HNO_3$. The former specimen was prepared by adding $NHO_3$ (0.1 or 0.2 ml) into NKX-2677 solution (25 ml). As shown in Fig. 14, the strength of the absorption peak at about 520 nm was increased by adding $HNO_3$ (0.1 or 0.2 ml).

Table 11 shows the summary of the characteristics of the dye-sensitized solar cells fabricated using NKX-2677 with and without $HNO_3$ (0.1 ml). Five devices were fabricated. The average conversion efficiency is 4.1% which is larger by 0.5 point (about 15%) than that of the solar cells fabricated using NKX-2677 without $HNO_3$ (see Table 7). The reason of this increase of the conversion efficiency is considered to be an increase of the optical absorption strength at about 520 nm shown in Fig. 14.

**Table 11** Summary of the characteristics of the dye-sensitized solar cells fabricated using NKX-2677 with and without $HNO_3$ (0.1 ml)

| Sample number | $J_{SC}$ (mA/cm$^2$) | $V_{OC}$ (V) | $P_{MAX}$ (mW) | F.F. | $\eta$ (%) |
|---|---|---|---|---|---|
| Sample 1 | 6.5 | 0.60 | 2.0 | 0.50 | 4.0 |
| Sample 2 | 6.7 | 0.60 | 2.0 | 0.49 | 4.0 |
| Sample 3 | 7.0 | 0.61 | 2.0 | 0.48 | 4.1 |
| Sample 4 | 7.2 | 0.61 | 2.1 | 0.47 | 4.1 |
| Sample 5 | 7.4 | 0.61 | 2.1 | 0.47 | 4.2 |
| Average | 7.0 | 0.61 | 2.0 | 0.48 | 4.1 |

# 7 Summary

In this chapter, the characteristics of the dye-sensitized solar cells using natural dyes, such as those found in red-cabbage, curcumin, and red-perilla, and synthesized dyes, such as NKX-2553, NKX-2677, and D149, with no precious metal have been discussed.

The obtained largest conversion efficiency is greater than 1% for the dye-sensitized solar cell using the dye of red-cabbage. This value is small. However, the cost performance is more than 50 times larger than that of the dye-sensitized solar cell using Ruthenium complex. Therefore, when the cost of FTO and ITO substrates, oxide semiconductor, electrolyte solution, and opposite electrode becomes very low, dye-sensitized solar cells fabricated using natural dyes may become the most cost effective type of solar cell available.

The effects of the dye solution's pH on the characteristics of the dye-sensitized solar cells have been also described. The conversion efficiencies of the dye-sensitized solar cells using the dyes of red-cabbage, red-perilla, NKX-2553, and NKX-2677 become larger when the pH value is low. It is expected that this technique will be used for future dye-sensitized solar cell systems.

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