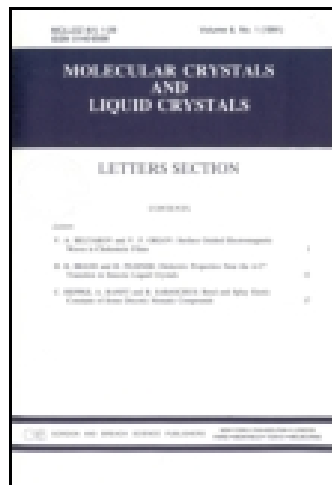


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# Enhancing Performance of Dye-Sensitized Solar Cell Influenced by Phosphor $\text{ZnGa}_2\text{O}_4$

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*Generally, N-719 dye absorbs only visible light in the wavelength range 400–700 nm; therefore, it does not permit utilization of most of the solar ultraviolet radiation. Ultra-violet radiation may be converted to visible light by conversion luminescence, and may be reabsorbed by the dye in a dye-sensitized solar cell (DSSC), and thus, more solar radiation may be utilized. Thus, we used a phosphor ( $\text{ZnGa}_2\text{O}_4$ ) in DSSCs to improve the harvesting of the incident light. As a result, the phosphor-mixed  $\text{TiO}_2$  electrode showed better performance than did the bare cells. When the amount of  $\text{ZnGa}_2\text{O}_4$  added was 7 wt%, the light-to-electric energy conversion efficiency of the DSSCs reached 4.97% and increased by a factor of 1.23 relative to those without  $\text{ZnGa}_2\text{O}_4$ .*

**Keywords** DSSCs;  $\text{TiO}_2$ ; phosphor; phosphor-mixed DSSC

## Introduction

Considerable effort has been devoted to the study of dye-sensitized solar cells (DSSCs) since they were first reported by Gratzel and O'Regan in 1991 [1]. As a promising alternative to solid-state photovoltaic devices for the next generation, the DSSC has attracted attention because of its low cost production, environmentally friendly components, and relatively high conversion efficiency [2, 3]. Moreover, enhanced dye-sensitized solar cell efficiency would provide enormous economical advantages [4–8]. While great progress on the DSSC has occurred over the past decade [9–11], enhancing its efficiency and reducing its production costs are still crucial problems. Their efficiency could not be increased further because of the inability of commonly employed sensitizers to efficiently absorb light over a sufficiently broad region of the solar spectrum [10, 11]. Thus, new sensitizers are being designed and synthesized to achieve highly efficient DSSCs. Generally, N-719 absorbs only visible light in the wavelength range 400–700 nm; therefore, it does not allow utilization of most of the solar ultraviolet (UV) radiations [9]. In order to enhance the efficiency of DSSCs by up to 15%, they should utilize both visible and UV radiations. If UV radiation can be converted to the visible light by conversion luminescence and then reabsorbed by the dye in DSSCs, more solar radiation can be utilized, effectively enhancing the DSSC photocurrent. Using

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phosphors in DSSCs is an effective method to improve the harvesting of the incident light [12-14]. Therefore, we used a phosphor-mixed  $\text{TiO}_2$  working electrode in DSSCs, and observed that the efficiency of DSSCs with phosphors was higher than that of the bare cell. This paper described the efficient performance and characterization of phosphor-mixed DSSCs.

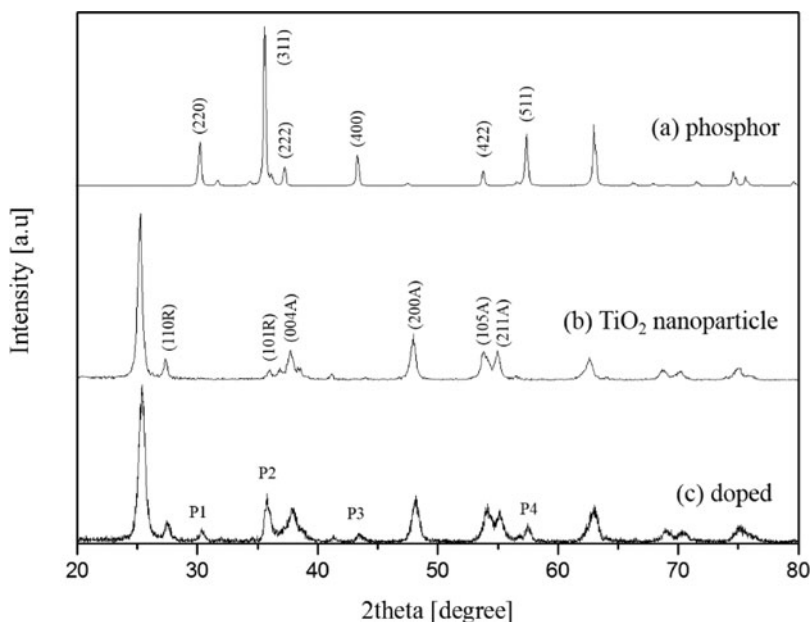
## Experimental

Phosphors were prepared by the precipitation method. The starting materials for  $\text{ZnGa}_2\text{O}_4$  phosphors were zinc sulfate heptahydrate ( $\text{ZnSO}_4 \cdot 7\text{H}_2\text{O}$ , 99.9%, Aldrich) and gallium sulfate hexahydrate ( $\text{Ga}_2(\text{SO}_4)_3 \cdot 6\text{H}_2\text{O}$ , 99.995%, Aldrich). The starting solutions were prepared by dissolving zinc sulfate and gallium sulfate at a Zn/Ga metal atom ratio of 1:2 in distilled water. The solutions were then added directly to an aqueous  $\text{NH}_3$  solution with stirring; the temperature was controlled at  $90^\circ\text{C}$  with stirring for 20 h to cause precipitation. The precipitates were separated by using a membrane filter; they were then washed with distilled water and ethanol, and dried in an oven at  $60^\circ\text{C}$  in air. The obtained powder was sintered at  $1000^\circ\text{C}$  for 2 h in an alumina crucible placed in a box furnace.

Titanium(IV) isopropoxide (TTIP, Sigma Aldrich), ethyl alcohol, nitric acid, and de-ionized water were used as the precursors for the  $\text{TiO}_2$  particles. The phosphor-mixed  $\text{TiO}_2$  paste was prepared by the sol-gel method. The phosphor-mixed  $\text{TiO}_2$  film was prepared by coating it on an FTO plate by the doctor blade technique. The phosphor and  $\text{TiO}_2$ -mixed cell were immersed in the dye (N719) solution for 24 h at room temperature. A counter electrode was prepared by spin-coating a  $\text{H}_2\text{PtCl}_6$  solution onto the FTO glass and subsequent heating at  $450^\circ\text{C}$  for 30 min. The  $\text{TiO}_2$  electrode with the adsorbed dye and the Pt counter electrode were assembled into a sandwich-type cell; the assembly was then sealed with a  $60\text{-}\mu\text{m}$  thick hot-melt sealant. An electrolyte solution was introduced through a drilled hole in the counter electrode, which was then sealed with a cover glass. The morphology of the prepared phosphor was investigated using field-emission scanning electron microscopy (FE-SEM, model S-4700, Hitachi). Photoluminescence spectra were obtained using a FP-6200 spectrofluorometer with 150 W Xe lamp (Jasco). The active area of the resulting cell exposed to light was approximately  $0.25\text{ cm}^2$  ( $0.5\text{ cm} \times 0.5\text{ cm}$ ). The absorption spectra of the electrode films were measured using an ultraviolet-visible (UV-vis) spectrometer (UV-vis 8453, Agilent). The conversion efficiency of the fabricated DSSCs was measured using an I-V solar simulator (Solar Simulator, McScience).

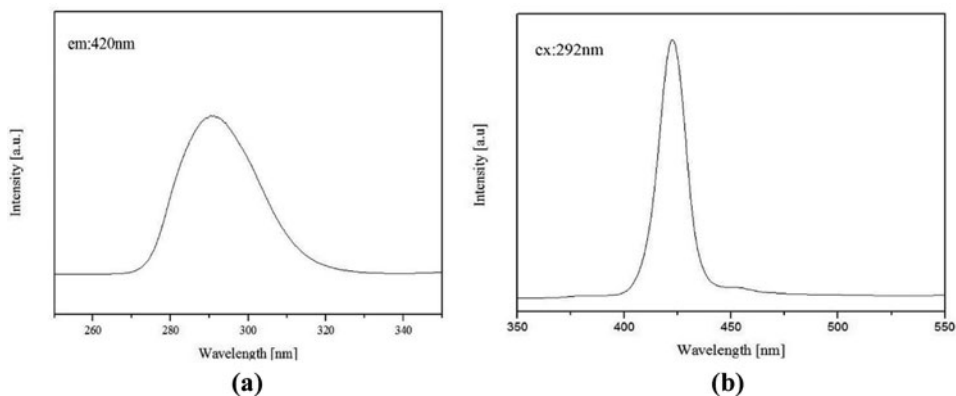
## Results and Discussion

Structural characterization of the pure  $\text{TiO}_2$  and phosphor-mixed  $\text{TiO}_2$  powders was performed by X-ray diffraction (XRD). Figure 1 shows the XRD patterns for pure  $\text{TiO}_2$  and phosphor-mixed  $\text{TiO}_2$  powder,  $\text{ZnGa}_2\text{O}_4$  phosphor samples. Fig. 1(a) shows the XRD patterns of the  $\text{ZnGa}_2\text{O}_4$  phosphor sintered at  $1000^\circ\text{C}$ . The XRD patterns of  $\text{ZnGa}_2\text{O}_4$  phosphor are consistent with the samples composed of a single phase of JCPDS 38-1240. Fig. 1(b) shows the XRD pattern of the  $\text{TiO}_2$  nanoparticles sintered at  $500^\circ\text{C}$ . It indicates a mixture of the anatase and rutile phases. As a result, Fig. 1(c) showed that phosphor-mixed  $\text{TiO}_2$  sintering at  $500^\circ\text{C}$  had polycrystalline structures consisting of  $\text{TiO}_2$  and phosphor phase. Figure 2(a) shows the excitation spectra of the  $\text{ZnGa}_2\text{O}_4$  phosphor powders sintered at  $1000^\circ\text{C}$  obtained by monitoring the emission at 420 nm. The excitation band at wavelength 280–300 nm is the most intense band. The emission spectrum of the  $\text{ZnGa}_2\text{O}_4$  phosphor powder sintered at  $1000^\circ\text{C}$  is shown in Fig. 2(b). The emission spectrum of  $\text{ZnGa}_2\text{O}_4$



**Figure 1.** XRD patterns of (a)  $\text{ZnGa}_2\text{O}_4$  phosphor and (b) pure  $\text{TiO}_2$ , (c) phosphor-mixed  $\text{TiO}_2$ .

shows a narrow, self-activated blue emission band around 420 nm, which can be attributed to the presence of  $\text{Ga}^{3+}$  at the octahedrally coordinated site [15]. As a result, the powder samples had an emission band at 420 nm under excitation at 292 nm. Figure 3 shows the SEM images of the  $\text{ZnGa}_2\text{O}_4$  phosphor at  $1000^\circ\text{C}$ . Uniform, spherical nanoparticles of the  $\text{ZnGa}_2\text{O}_4$  phosphor with the homogeneous structure were obtained by precipitation at  $1000^\circ\text{C}$ . The mean size of the particles at  $1000^\circ\text{C}$  measured from the SEM image was  $<50$  nm. Figure 4 shows the Nyquist plot of DSSCs with seven types of mixed phosphors at open-circuit voltage. Electrochemical impedance spectroscopy (EIS) are a useful method for analysis of charge transport process and internal resistances [16]. The results

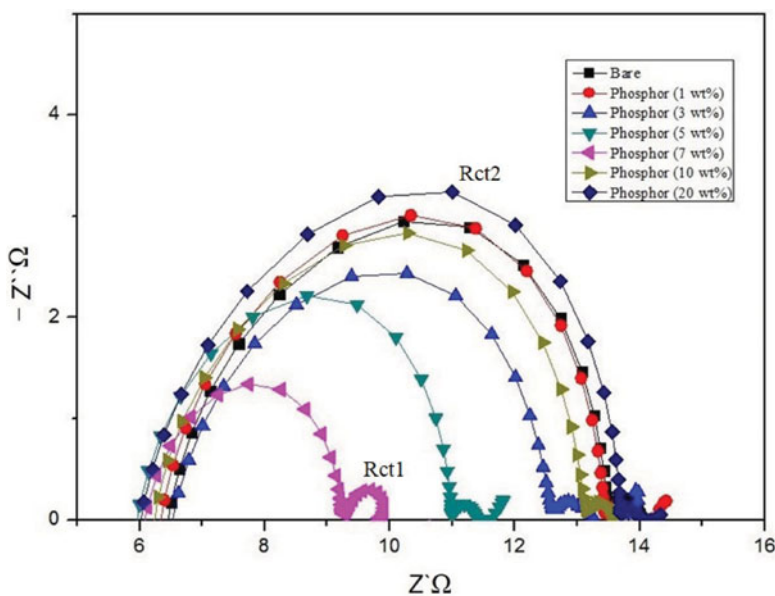


**Figure 2.** (a) Excitation spectrum of powder samples prepared at  $1000^\circ\text{C}$ . (b) Emission spectra of powder samples prepared at  $1000^\circ\text{C}$ .

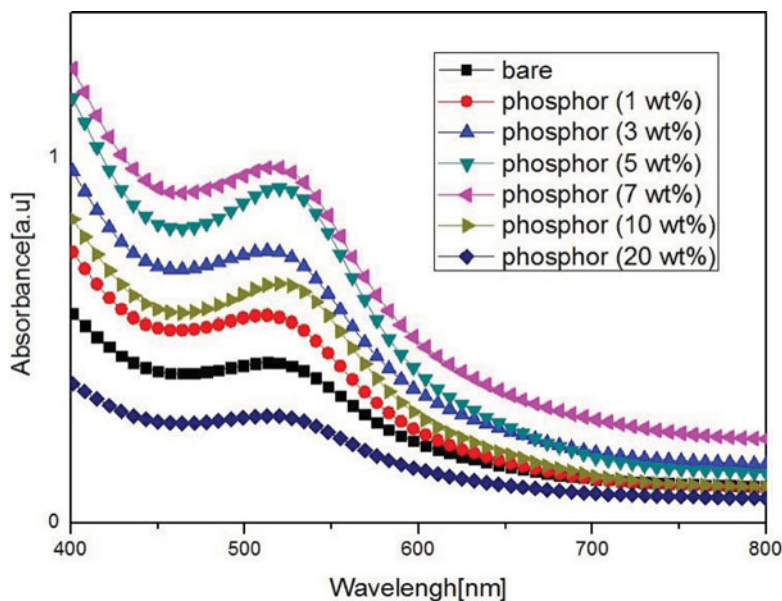


**Figure 3.** FE-SEM images of  $\text{ZnGa}_2\text{O}_4$  phosphor prepared at  $1000^\circ\text{C}$ .

indicate that the  $R_{ct2}$  values gradually decreased as the amount of the mixed phosphors was increased. However, the  $R_{ct2}$  values decreased when the amount applied became more than 10 wt%. The result indicates that DSSCs with the phosphor showed the highest cell efficiency (4.97%) compared with the results of the other DSSCs. The phosphor-mixed DSSC produced higher  $J_{sc}$  values. Apparently, the phosphor-mixed DSSCs showed much better performance than did the DSSCs without any phosphor.

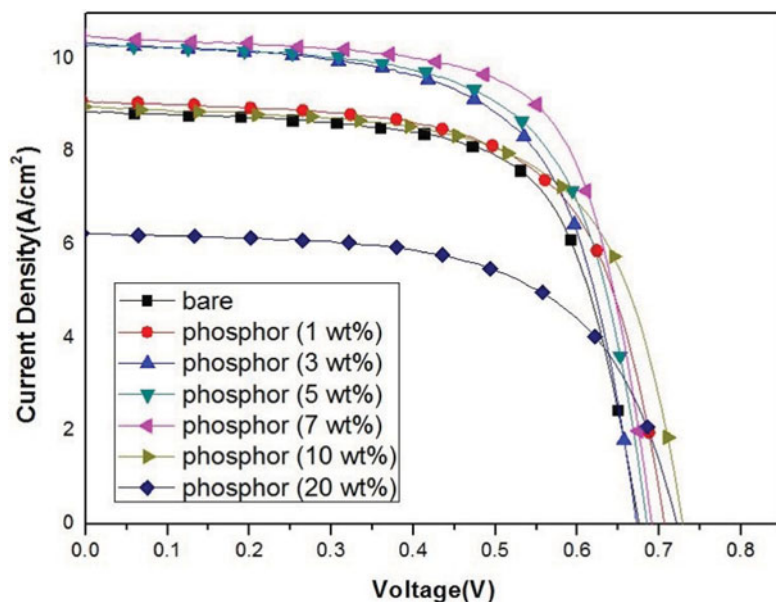


**Figure 4.** Nyquist plot of phosphor mixed DSSC at open-circuit voltage.



**Figure 5.** UV-vis absorbance of phosphor-mixed DSSCs.

Figure 5 shows the UV-vis absorbance of the phosphor-mixed DSSCs. The absorption spectrum of  $\text{ZnGa}_2\text{O}_4$  phosphor has a peak at ultraviolet ray area [17]. And N-719 only absorb visible light in the wavelength range 400–800 nm [9]. The sample doped with 7wt% showed the highest absorbance over the wavelength range 400–800 nm. Obviously, this is



**Figure 6.** Photovoltaic parameters of DSSC.

**Table 1.**  $J_{sc}$ ,  $V_{oc}$ , FF, and Efficiency

Sample	$J_{sc}$ (mA/cm <sup>2</sup> )	$V_{oc}$ (V)	FF (%)	Efficiency ( $\eta$ )
Bare	9.38	0.676	63.77	4.04
Phosphor (1 wt%)	9.10	0.706	64.94	4.17
Phosphor (3 wt%)	10.34	0.672	64.39	4.48
Phosphor (5 wt%)	10.30	0.685	65.57	4.62
Phosphor (7 wt%)	10.50	0.691	68.52	4.97
Phosphor (10 wt%)	8.98	0.728	64.49	4.22
Phosphor (20 wt%)	6.25	0.721	61.36	2.76

due to the conversion luminescence of phosphor from reabsorption by dye in the DSSC. Evidently, effective conversion luminescence of the phosphor via the phosphor-mixed TiO<sub>2</sub> electrode occurred.

Figure 6 shows the current–voltage photovoltaic performance curves of DSSCs and phosphor-mixed DSSCs under AM 1.5 illumination (100 mW/cm<sup>2</sup>). The equivalent values based on the weight ratio of the phosphor are summarized in Table 1. When the amount of ZnGa<sub>2</sub>O<sub>4</sub> added was 7 wt%, the light-to-electric energy conversion efficiency of the DSSC reached 4.97% and increased by a factor of 1.23 compared with that of the DSSC without ZnGa<sub>2</sub>O<sub>4</sub>. However, the efficiency decreased when the amount of ZnGa<sub>2</sub>O<sub>4</sub> became more than 10 wt%. This is mostly because of the crystal defects produced by the large amount of the phosphor. The phosphor-mixed DSSC exhibited a slightly higher photocurrent density than did the bare cell because of light harvesting by the ZnGa<sub>2</sub>O<sub>4</sub> phosphor in the DSSC. Therefore, the optimum condition for high conversion efficiency was the addition of 7 wt% ZnGa<sub>2</sub>O<sub>4</sub>.

## Conclusions

The photovoltaic performance of the phosphor-mixed DSSCs (weight ratio) was compared. The UV radiation could be converted to the visible light by conversion luminescence and then reabsorbed by the dye in the DSSCs, so that more solar irradiation could be utilized. Therefore, ZnGa<sub>2</sub>O<sub>4</sub> improves light harvesting and the photocurrent by conversion luminescence. When the amount of ZnGa<sub>2</sub>O<sub>4</sub> added was 7 wt%, the light-to-electric energy conversion efficiency of the DSSCs reached 4.97% an increase by a factor of 1.23 compared with the bare DSSCs. Therefore, increasing the amount of the light absorbed by DSSCs by using a phosphor may be an effective method to enhance their efficiency.

## Acknowledgments

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