Reliability performance of up-scaling DSSC into sub-module in series design using hermetic sealing

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Abstract. Dye-sensitized solar sub-module has been prepared as a practical application of DSSC. TiO$_2$ paste was deposited onto the 3.3 cm $\times$ 5 cm FTO glass by screen printing method consisting of two active cells with a dimension of 1 cm $\times$ 5 cm for each cell. The sub-module was assembled using two parts (part A and part B) hermetic sealing compound as a sealant. The performance of the sub-module was then observed using two independent measurements in the span of six days using sun simulator. Photovoltaic effect was properly occurred as indicated by the I-V curve for both of sub-module. First test showed that the sub-module with hermetic sealant has good performance with the highest efficiency of 2.9%. Meanwhile, the second test showed significant decrease of the sub-module performance with the highest efficiency of 1.43%.

1. Introduction
Several alternative energy sources have been developed to replace fossil energy source. Solar energy, as one of renewable energy source which is abundant, clean, safe, and able to generate energy in remotes areas, become the main candidate energy source to replace fossil fuel [1]. To date, silicon solar cells are the main commercial photovoltaic technology. Unfortunately, this technology has high cost and is not environmentally friendly. To overcome these problems, researchers have been developing new photovoltaic energy based on dyed-TiO$_2$ semiconductor, known as dye-sensitized solar cells (DSSC). One study, conducted by Gratzel, et al [2], obtained cell efficiency of 11% [3].

Scaling up the cells into DSSC module is the most important part for practical application. Module designs in two different groups of series and parallel can be chosen according to its necessity. Serial connection consists of monolithic and W/Z interconnection schemes that has high output voltage and low current. Gratzel disclosed that performance of DSSC in module scale had reached efficiency of 7% [3].

In the fabrication of DSSC module, the performance stability is the main focus of investigation. Some factors such as materials, assembly process, and environmental condition can affect the performance of DSSC particularly on photoconversion efficiency and stability. One of the requirements to meet the criteria is good sealing between photoelectrode and counter electrode which could avoid the leakage of electrolyte liquid thermoplastic sealant is one of the best sealant materials for cells. It is not suitable for module due to its difficulty in the assembly process. One of many materials for sealant is a hermetic seal-based materials, which has semi-rigid seal for DSSC enclosure.
In this work, two-part hermetic sealing compound from Dyesol was employed as sealing material for DSSC sub-module using series interconnection. The hermetic compound can be applied easily by screen printing technique since it is in its paste form. In addition, the hermetic sealant does not require high temperature for drying which is suitable for dyed DSSC. Two sets of hermetic compound were prepared to obtain the optimum sealant based on its composition. The scope of investigation include performance optimization, as well as sealing endurance which affects its lifetime stability.

2. Experiments

2.1. Preparation of photoelectrode and counter electrode
The screen printing method was used in the fabrication process of DSSC sub-modules. Photoelectrode was prepared on a 3.3 cm x 5 cm area of FTO glass which was divided into two active areas of 1 cm x 5 cm for each area. Then, the pattern for sub-module screen (figure 1) consisted of active area, circuit contact, and sealant area was created using photolithography process. Meanwhile, FTO glass as a conductive substrate was prepared for photoelectrode and counter electrode by scribing the surface into two parts. These substrates were then cleaned with DI water using ultrasonic cleaner and immersed in ethanol solution for 1 hour. For photoelectrode preparation, TiCl$_4$ pretreatment was applied by dipping the cleaned FTO glass in TiCl$_4$ solution (0.04 M) for 30 minutes with temperatures of 70 °C. After the pretreatment process, TiO$_2$ paste (Dyesol) was deposited onto the FTO glass by screen-printing technique to make TiO$_2$ active layer. This film was dried in vacuum oven with temperatures of 100 °C for 10 minutes and sintered in infrared furnace at 450 °C for 45 minutes. TiCl$_4$ post-treatment was applied as well to the sintered film, using the same process as pretreatment. After rinsing in ethanol solution, the film was sintered once more with the same process as before and immersed in dye solution of N907 for 24 hours.

Counter electrode was prepared with an area equal to the photoelectrodes. Platinum (Pt) was deposited onto the substrate using sputtering process for 2 hours. The area of the electrode was divided into two parts by scribing the Pt layer as the area of photoelectrode substrate.

2.2. Assembly process and performance measurement
The assembly process of DSSC sub-module was preceded by the sealing preparation, which uses two parts hermetic sealing compound (part A and part B) from Dyesol. These parts were combined in two different compositions that can be seen in table 1. On sealant area of TiO$_2$ photoelectrode, hermetic compound was subsequently deposited by screen printing method. Silver conductive paste (Ag) was deposited as well on circuit contact area. Counter electrode was directly stacked on the photoelectrode to assemble them into sandwich construction in Z-type series interconnection. The sub-module was heated in temperature of 100 °C for 5 min to dry the sealant and Ag paste so that both electrodes attached to each other. The scheme of the interconnection was described in figure 2. Electrolyte liquid
was injected to the assembled sub-module through a pre-drilled tiny hole on the counter electrode surface.

![Interconnection scheme of DSSC sub-module.](image)

**Figure 2.** Interconnection scheme of DSSC sub-module.

**Table 1.** Composition ratio of hermetic sealing compound (Dyesol).

| Sealant Compound | Part A | Part B |
|------------------|--------|--------|
| Hermetic A       | 1      | 0.75   |
| Hermetic B       | 1      | 0.5    |

To investigate photovoltaic performance of the sub-module, I-V measurement was applied by illuminating the sub-module using sun simulator at 500 watt/m² intensity. Aside from measuring performance of the sub-module, this work also observed the quality of the compound of hermetic sealant in the stability aspect. Therefore, the I-V measurement was taken twice in the range of six days.

### 3. Results and discussion

In the fabrication of DSSC sub-modules, some aspects need to be observed including photoelectrode and counter electrode preparation, assembly process, and sealing material. In the preparation of TiO₂ photoelectrode, TiCl₄ treatment was applied to improve the inter-particle connectivity and make TiO₂ layer more compact [4]. Application of hermetic compound for the sub-modules becomes an attempt to find out the optimum sealing materials. With sufficient ratio between part A and part B, hermetic sealing compound should be able to carry out the high reliability of DSSC sub-module so that the submodule can perform with long-term stability which is important for practical application.

![Sample of DSSC sub-module](image)

**Figure 3.** Sample of DSSC sub-module (a) with hermetic A sealant compound and (b) with hermetic B sealant compound.

TiO₂ has been used as a semiconductor material for the DSSC sub-module. Characteristics of TiO₂ as main material is essential for photovoltaic performance which can be obtained by its morphology or crystalline phase. Figure 4 shows XRD pattern of TiO₂ layer deposited on FTO glass. It can be seen from the pattern that strong peak was in position of 2θ with intensity of 25.3° which is confirmed as
anatase phase of TiO₂. Chemical and physical treatment in the fabrication process did not change the phase composition of TiO₂.

![XRD pattern of TiO₂](image)

**Figure 4.** XRD pattern of TiO₂.

Performance of solar sub-module was investigated by I-V measurement using sun simulator. Figure 5 shows the I-V curve from the first test that describes photovoltaic performance of the sub-module. Both of sub-module using hermetic A and hermetic B sealant showed the photovoltaic effect confirms the proper work as a solar cells. Photoelectrodes had good performance which was affected by the treatment in the preparation process in spite of manual process in assembly.

![I-V curve of DSSC sub-module sample from the first I-V test](image)

**Figure 5.** I-V curve of DSSC sub-module sample from the first I-V test.

Composition between two parts of hermetic sealant compound obviously affected the sub-modules performance as well. It can be seen from table 2 that higher performance of photovoltaic was achieved by DSSC sub-module that uses hermetic B as sealant, with the value of open-circuit voltage (Voc),
short-circuit current density ($J_{sc}$), fill factor (FF), and photoconversion efficiency ($\eta$) of 1.427 V, 26.38 mA/cm$^2$, 0.38, and 2.9%, respectively. Hermetic B might have been a more effective compound which formed an effective hermetic seal that can make stronger bond with glass and minimize electrolyte leakage. The result also indicates that both cells of each sub-module had good series interconnection which could affect the value of open circuit voltage.

| Sealant Compound | $V_{oc}$ (V) | $J_{sc}$ (mA/cm$^2$) | FF | $\eta$ (%) |
|------------------|--------------|----------------------|----|----------|
| Hermetic A1      | 1.386        | 18.92                | 0.37 | 1.9          |
| Hermetic B1      | 1.427        | 26.38                | 0.38 | 2.9          |

Second I-V test of sample was taken six days after the first test with same criteria. The result of the second test can be seen in figure 6. Both of I-V test of samples show curves that were less ideal compared with the curves from the first I-V test. Nevertheless, the effect of photovoltaic still appears which means that both hermetic sealants were able to insulate the interconnection of the cells and avoid unwanted mass transport between electrolytes of adjacent cells.

![I-V curve of DSSC sub-module sample from the second I-V test.](image)

Figure 6. I-V curve of DSSC sub-module sample from the second I-V test.

Poor performance of the samples in the second test can also be seen from the quantity parameters, mainly for sub-module with hermetic B which yielded more significant decrease. The result shows that performance of sub-module with hermetic A as sealant was higher than sub-module with hermetic B, with best performance of $V_{oc}$, $J_{sc}$, FF, and $\eta$ respectively are 1.427 V, 18.29 mA/cm$^2$, 0.28, and 1.43%. In this case, sub-module with hermetic A was able to remain more stable after six days from the first measurement. Performance degradation of sub-module with hermetic A was 24% for the photoconversion efficiency while sub-module with hermetic B was 71.3%.

Hermetic A initially demonstrated good performance as sealant in terms of the ability to avoid the leakage and minimize the reaction between electrolytes. However, hermetic A failed to maintain its
performance in long-term stability. Hermetic B, which had lower performance in the first test, became relatively stable according to the second test. Hermetic B was considered to have less reaction with electrolyte and less effect on surroundings in terms of temperature and humidity.

| Sealant Compound | Voc (V) | Jsc (mA/cm²) | FF | η (%) |
|------------------|---------|--------------|-----|-------|
| Hermetic A2      | 1.427   | 18.29        | 0.28| 1.43  |
| Hermetic B2      | 1.467   | 8.968        | 0.32| 0.83  |

**Table 3.** Photovoltaic parameters of DSSC sub-module from the second I-V test.

### 4. Conclusion
In summary, hermetic sealing compound was employed as sealant in the fabrication of DSSC sub-module. The optimized composition of sealant was hermetic compound with ratio part A : part B = 1 : 0.5 as the DSSC sub-module received the higher photoconversion efficiency of 2.9% in the first I-V test. On the contrary, in terms of obtaining the stability higher performance was received by sub-module with sealant containing hermetic compound part A : part B = 1 : 0.75 in the second I-V test. Initially, hermetic B sealant compound had better quality compared with hermetic A but hermetic A is more recommended for long term use.

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