Mechanical Stacking Multi Junction Solar Cells Using Transparent Conductive Adhesive

Shinya Yoshidomi, Jun Furukawa, Masahiko Hasumi, Toshiyuki Sameshima *

Tokyo University of Agriculture and Technology, 2-24-16 Naka-cho, Koganei, Tokyo 184-8588, Japan

Abstract

We report a mechanical stacking technology with transparent conductive adhesive as intermediate conductive layer for multi-junction-solar cells. Transparent adhesive jell dispersed with Indium-Tin-Oxide (ITO) particles ranging from 5 to 10 wt% was developed for stacking solar cell substrates. A connecting resistivity below 2.0 $\Omega \text{cm}^2$ was achieved in a condition of solidifying the adhesive gel under a gas pressure from 4.0x10^5 to 7.0x10^5 Pa. The connecting resistivity was kept lower than 3.0 $\Omega \text{cm}^2$ when samples was exposed at a high temperature at 80°C and a high humidity of 85% humidity atmosphere in the case of solidification pressure at 7.0x10^5 Pa. The low connecting resistivity give a possibility of application our conductive adhesive to solar cell with an effective efficiency higher than 30%. Stacked solar cells was successfully achieved by pasting a InGaP/GaAs thin film epitaxial layer to a Ge solar cell using our intermediate adhesive layer.

© 2014 The Authors. Published by Elsevier Ltd. This is an open access article under the CC BY-NC-ND license (http://creativecommons.org/licenses/by-nc-nd/3.0/).
Peer-review under responsibility of The European Materials Research Society (E-MRS)

Keywords: Multi junction solar cells, Indium Tin Oxide, Mechanical Stacking

* Corresponding author. Tel.: +81-42-388-7109; fax: +81-42-388-7109.
E-mail address: tsamesim@cc.tuat.ac.jp
1. Introduction

Solar cell is an important device, which converts sunlight into electrical power [1-5]. Especially, multi junction solar cells with different band gaps are attractive for effectively collecting sunlight from ultraviolet to infrared region. The hetero-epitaxial growth method has been widely developed for forming layered structure crystalline semiconductors whose band gap energies decrease in the depth direction [6-17]. Fabrication of large area in low cost has been still important subject. We have proposed fabrication of multi junction solar cells by stacking individual solar cells using intermediate transparent adhesive layers including conductive indium tin oxide (ITO) particles [18-20]. Individually fabricated top and bottom cells with a wide and narrow band gap, respectively, are electrically and optically bonded as stacked cells using the transparent and conductive adhesive. We have developed adhesive including ITO particles. Our method may have the advantage of high yield in cell fabrication at low cost. Lattice matching of top and bottom cells, atomic flat surfaces are not required for this method. Therefore, it is possible to stack fragile or no-heat resistant materials with a large area by soft adhesion, which solidifies at room temperature. Important subjects are establishment of optimum conditions of low connecting resistivity, temperature and humidity resistivity properties of the adhesive, and demonstration of multi junction cells using this method.

In this paper, we discuss the condition of the low connecting resistivity for solar cells with high effective efficiencies. We also report experimental demonstration the low connecting resistivity lower than that condition. Moreover, temperature and humidity resistivity properties of adhesive are also reported. Finally, application of our adhesive layer to solar cell was demonstrated by pasting thin film InGaP/GaAs epitaxial layer to a Ge solar cell.

2. Low connecting resistivity condition

We set a target value of connecting resistivity of intermediate adhesive layer. To estimate the target value, the influence of resistivity to conversion efficiency of solar cell was estimated by numerically solving eq.1 with one PN diode and a series resistivity Rs. Self-consistent calculation was conducted.

\[
J = J_0 \left\{ \exp \left( \frac{q(V - R_s J)}{n k T} \right) - 1 \right\} + \frac{V - R_s J}{R_{sh}} J_{ph}
\]

where J, J_0, R_sh, n, and J_{ph} are current density, saturation current density, parallel resistivity, ideality factor, and photo current density, respectively. Multi junction solar cell characteristics with 30% conversion efficiency and 0 \( \Omega \text{cm}^2 \) series resistance ranging their open circuit voltage \( V_{oc} \) from 1.0 to 3.0 V were calculated. Then, connecting the value of connecting resistivity which decreases the effective conversion \( \eta \) from 30 to 29.5, 29.0, 28.5 and 28% were calculated. Figure 1 shows connecting resistivity as a function of \( V_{oc} \) with effective \( \eta \) of 28.0, 28.5, 29.0 and 29.5%, respectively. Connecting resistivity increased as \( V_{oc} \) increased with every effective \( \eta \) case. Connecting resistivity below 2.0 \( \Omega \text{cm}^2 \) is required to achieve 29.0% for \( V_{oc} \) at 1.5 V. Solar cell with high \( V_{oc} \) allows high connecting resistivity. In this report, we set our target value of connecting resistivity as 2.0 \( \Omega \text{cm}^2 \).
3. Experimental

ITO particles with a diameter of 20-25 μm were prepared. ITO particles ranging from 5 to 10 wt% were mechanically dispersed in Cemedine adhesive. The Cemedine gel including ITO particles was sandwiched with two substrates. To solidify the intermediate layer, samples were set in the pressure proof chamber under nitrogen gas environment with a pressure at ranging from $3.0 \times 10^5$ to $7.0 \times 10^5$ Pa for 1.5 h at room temperature. Fragile Ge substrates with a diameter of 4 inch and thickness of 150 μm were stacked by Cemedine adhesive including ITO particles. Then, the intermediate adhesive layer was solidified in nitrogen gas environment at $6.0 \times 10^5$ Pa.

Cemedine gel with ITO/Cemedine weight ratios ranging from 5 to 10 wt% was sandwiched by two n-type Si substrates with a resistivity of 0.001 Ωcm and 20 cm² area. The intermediate layers were solidified by setting samples in the pressure proof chamber under nitrogen gas environment with a pressure at ranging from $3.0 \times 10^5$ to $7.0 \times 10^5$ Pa for 1.5 h. Current with a bias voltage (J-V) characteristics were measured with Al electrodes. To calibrate the connecting resistivity of intermediate layer, contact resistivity was estimated by measuring J-V characteristics of Si substrate with Al electrodes. We also prepared 100 n-type 0.001 Ωcm Si pieces with a size of 0.6 x 0.6 cm² mechanically stacked on the 4-inch n-type 0.001 Ωcm Si substrate. The distributions of 100 J-V characteristics were measured for applying voltage individually to all Si pieces.

We investigated temperature and humidity resistivity properties of adhesive. Samples of Si/7wt%-ITO dispersed Cemedine/Si were fabricated by hydrostatic pressing with $7.0 \times 10^5$ Pa. The samples were placed in 80°C and 85% humidity conditions for 1100 h. Samples were directly faced to high humidity at high temperature.

To investigate the possibility for application of our adhesive gel, stacking of InGaP/GaAs cell and Si substrate was demonstrated. InGaP/GaAs/Ge three junction solar cell with a size of 8x4 cm² was prepared. Then, the rear electrode of solar cell was removed by mechanical etching. The solar cell and silicon substrate were pasted by Cemedine gel including ITO particles at 6 wt%. The adhesive layer was solidified with a pressure at $4.0 \times 10^5$ Pa. Solar cell characteristics were measured using a AM1.5 solar cell simulator at 1 sun. Moreover, stacking of InGaP/GaAs thin film solar cell and Ge cell was also demonstrated. InGaP/GaAs tandem solar cell formed on 500 μm-thick GaAs substrate was prepared. Top metal electrodes were formed on n-GaAs contact layer by photolithography and etching method. Conformal etching of the n-GaAs layer was carried out to form light windows. Then, underlying doped GaAs substrate of the InGaP/GaAs thin film solar cells, formed with the epitaxial method, was polished and etched up to 20 μm. They were then subsequently stacked on Ge cells, using the transparent conductive adhesive with the ITO particles. Solar cell characteristics were measured using a AM1.5 solar cell simulator at 1 sun.

4. Results and discussion

Figure 2 shows picture of sample with the structure of 150 μm-thick Ge/ Cemedine-ITO layer/ Ge with a diameter of 4 inch. The fragile substrates were successfully stacked by intermediate adhesive layer including ITO particles with nitrogen gas press at $6.0 \times 10^5$ Pa. This result indicates that stacking of fragile substrates with large area was successfully demonstrated by gas press.

Figure 3 shows electrical current density as a function of voltage applied to the samples with the structure of Si/ Cemedine-ITO layer/ Si with an area of 20 cm². The amount of ITO particles was set to be 6 wt% of Cemedine adhesive. The adhesive sandwiched with silicon substrates was solidified in nitrogen gas at the pressure ranging from $3.0 \times 10^5$ to $7.0 \times 10^5$ Pa for 1.5 h. All samples showed good ohmic characteristics as shown in Fig. 3. A high current density of 0.05 A/cm² was obtained at the applied voltage of 0.034 V for the sample solidified at $5.0 \times 10^5$ Pa. This means that the calibrated connecting resistivity of the intermediate Cemedine-ITO layer was 0.1 Ωcm², which was lower than our target value. On the other hand, in the case of sample formed at $3.0 \times 10^5$ Pa, the connecting resistivity was high of 4.1 Ωcm². The gas pressure is important to achieve intermediate adhesive layer with a low connecting resistivity. Figure 4 shows the connecting resistivity as a function of gas pressure with ITO/Cemedine weight ratios ranging from 5 to 10 wt%. The connecting resistivity lower than 2.0 Ωcm² was achieved for any conditions of sample fabrication, except for a gas pressure of $3.0 \times 10^5$ Pa and ITO amounts of 6 and 9 wt%. The lowest value was 0.04 Ωcm² for the condition of gas pressure of $4.0 \times 10^5$ Pa and ITO amount of 9 wt%. 
Figure 5 shows distribution of the connecting resistivity plotted by a step of 0.08 Ωcm² for 100 n-type Si pieces over the 4-inch n-type Si substrate. The pieces were bonded at 4.0x10⁵ Pa with ITO/Cemedine adhesive of 6 wt% ITO particles dispersed in the Cemedine. The connecting resistivity was distributed from 0.36 to 1.00 Ωcm² probably because of non-uniformity of ITO dispersion. The average connecting resistivity was 0.52 Ωcm². This result indicates that our mechanical stacking technique has a capability of low connecting resistivity at any points.
Figure 6 shows solar cell characteristics of the initial commercial InGaP/GaAs/Ge cell (dashed curve) and the cell stacked on the n-type Si substrate. Both I-V curves show similar and typical solar cell characteristics. The short circuit current density $J_{sc}$, fill factor $FF$, open circuit voltage $V_{oc}$, and conversion efficiency $Eff$ were 14.8 mA/cm$^2$, 0.85, 2.51 V, and 31.6% for the initial cell. They were 14.8 mA/cm$^2$, 0.84, 2.53 V, and 31.4%, respectively for the mechanically stacked cell. The effective efficiency was slightly lower than that initial cell because the connecting resistivity of the intermediate layer 0.8 $\Omega$cm$^2$, which was estimated from the solar cell characteristics around $V_{oc}$ point. This result demonstrates that our technique has a possibility of fabrication of stacked type multi junction solar cell with a high efficiency and a large area.

![Graph showing solar cell characteristics](image1)

Fig.6. Solar cell characteristics when InGaP/GaAs/Ge cell and Si substrate were stacked by intermediate layer. Dashed curve shows solar cell characteristics of initial cell.

We also demonstrate the mechanical stacking of thin film InGaP/GaAs cell to a Ge cell, as shown by solar cell characteristic in Fig. 7. The $J_{sc}$, FF, $V_{oc}$, and $Eff$ were obtained as 12.6 mA/cm$^2$, 0.60, 2.13 V, and 16.0%, respectively. The solar cell characteristic shows that our intermediate layer electrically and optically connected between the top InGaP/GaAs and Ge cells. The intermediate adhesive layer is applicable to stacking of thin-film multi junction solar cells.

![Graph showing solar cell characteristics](image2)

Fig.7. Solar cell characteristics of InGaP/GaAs/Cemedine-ITO layer /Ge cell.
Figure 8 shows changes in the connecting resistivity with time when the samples with a structure of n-type Si/ Cemedine-ITO adhesive layer/ n-type Si were placed at 80°C in 85% humidity atmosphere for 1100 h. The samples with intermediate adhesive layers solidified at 4x10^5 and 7x10^5 Pa by nitrogen gas press were prepared. Measurement of the connecting resistivity of the sample formed at 7.0x10^5 Pa kept at room temperature in dry atmosphere for 1100 h was also carried out for comparison. Although the connecting resistivity of samples treated at 80°C and 85% humidity gradually increased from 1.7 to 7.2 Ωcm² as elapse time increased for sample formed at 4.0x10^5 Pa, it increased only to 3.0 Ωcm² for sample formed at 7.0x10^5 Pa. On the other hand, connecting resistivity of sample which was kept in room temperature and dry atmosphere for 1100 h was 1.7 Ωcm². The connecting resistivity value of sample formed at 7.0x10^5 Pa and kept at 80°C and 85% humidity for 1100 h was close to that for sample kept in room temperature and dry atmosphere. Gas pressure for solidifying adhesive was an important condition for keeping low resistivity. Hard contact with a high pressure can by one of methods to realize mechanical staking with a low connecting resistivity in atmosphere with changeable temperature and humidity.

5. Conclusion

We reported achievements in fabrication of intermediate layer including ITO particles for mechanical stacking multi junction solar cells. According to numerical calculation of PN diode formula, the target connecting resistivity was set to be 2.0 Ωcm². 20 cm² size n-type Si substrates were stacked by our intermediate Cemedine adhesive. The ratio of ITO in Cemedine adhesive was ranged from 5 to 10 wt%. The samples were solidified in nitrogen gas environment with a pressure at ranging from 3.0x10^5 to 7.0x10^5 Pa for 1.5 h at room temperature. According to J-V characteristics measurement, the connecting resistivity of samples bonded with various ITO/Cemedine weight ratios achieved below 2.0 Ωcm², except for the samples solidified at 3.0x10^5 Pa. Moreover, 100 n-type Si pieces mechanically stacked on the 4-inch n-type Si substrate bonded at 4.0x10^5 Pa with ITO/Cemedine adhesive of 6 wt% ITO particles dispersed in the Cemedine revealed that the connecting resistivity was distributed from 0.36 to 1.00 Ωcm². Moreover, changes in connecting resistivity by heating the samples with a structure of n-type Si/ Cemedine-ITO adhesive layer/ n-type Si at 80°C in 85% humidity atmosphere for 1100 h was investigated. In the case of sample solidified at 7.0x10^5 Pa, the connecting resistivity was kept below 3.0 Ωcm² after 1100 h high temperature and high humidity treatment. We believe that it is enough low connecting resistivity for fabricating multi junction solar cells with high V_{oc}. Finally, stacking of InGaP/GaAs/Ge cell and Si solar cell, InGaP/GaAs and Ge cells revealed that our intermediate layer electrically connects top cell and bottom cells. We believe that the intermediate adhesive layer is applicable to stacking of thin-film multi junction solar cells.
Acknowledgements

This work was supported by the New Energy and Industrial Technology Development Organization (NEDO) under the Ministry of Economy, Trade and Industry, Japan (METI).

References