CHALCOPYRITE THIN-FILM TANDEM SOLAR CELLS WITH 1.5 V OPEN-CIRCUIT-VOLTAGE

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ABSTRACT

We have fabricated two-terminal mechanical stacked chalcopyrite-based tandem devices to identify the technical issues required to achieve high performance solar cells in the near future. A tandem device with a $Ag(In_{0.2}Ga_{0.8})Se_2$ (AIGS) top cell and a $Cu(In,Ga)Se_2$ (CIGS) bottom cell showed an open-circuit-voltage (V_{oc}) of 1.46 V. It was, however, found that the short-circuit current (J_{sc}) of a two-terminal tandem device was limited by the low J_{sc} of the filtered bottom cell because of a current mismatch between the top and bottom cells. In order to improve the J_{sc} of the filtered bottom cell, we have tried two process options, replacing ITO back contacts with high-mobility (Mo-doped In_2O_3) IMO and the use of low-Ga content CIGS bottom cells. The J_{sc} of the filtered bottom cells increased significantly using these techniques. As a result, an 8 % total-area ($0.5cm^2$) efficiency 2-terminal mechanical stacked tandem device was achieved using a CIGS (15% Ga) bottom cell with an AIGS top cell fabricated with an IMO back contact.

INTRODUCTION

The Japanese PV Roadmap until 2030 calls for a target efficiency for CIGS solar cell of 25% in FY2020 [1]. It would appear that this target will be difficult to achieve for single-junction CIGS devices as has been predicted by the Colorado State university group using computer simulations [2]. Therefore, a tandem device is a more likely candidate to realize a 25% efficiency device at present. The objectives of this work are to identify the technical issues that must be resolved before a 25% efficiency CIGS-based tandem thin film solar cells can be realized as well as to explore the possibility for super high-efficiency solar cells.

The NREL group has reported that the optimum bandgaps for a series-connected two-junction tandem cell were 1.72 eV for the top and 1.14 eV for the bottom cells [3]. These bandgaps are ideally matched to $CuGaSe_2$ (CGS) (Eg=1.68eV) and $Cu(In_{0.7}Ga_{0.3})Se_2$ (Eg=1.15eV) solar cells. It is well known that one of the most important issues for realizing CIGS-based tandem solar cells is to achieve high-efficiency top cells fabricated on a transparent back contact. However, CGS solar cells fabricated with a transparent conducting oxide (TCO) back contact showed an efficiency of 6.8% [4], although over 15% efficiency is required for the top cell. We thus have investigated a novel wide-gap (Eg=1.7eV) material, $Ag(In_{0.2}Ga_{0.8})Se_2$ (hereafter AIGS) as an alternative top cell [5]. In this paper, we discuss the technical issues facing two-terminal mechanically-stacked tandem devices using AIGS-based top cells and CIGS-based bottom cells. Two process options for the optimization of cell performance with respect to TCO back contacts and Ga content of bottom CIGS cells are presented.

FABRICATION AND PERFORMANCE OF AIGS TOP CELLS

Typically, 1.5-μm thick AIGS thin films were deposited by three-stage process using a molecular beam epitaxy (MBE) system. 200 nm-thick Indium tin oxide (ITO) films with a sheet resistance of 7 ohm/sq were used as a standard back contact. AIGS thin film solar cells with a ZnO:Al/ZnO/CBD-CdS/AIGS/ITO/SLG structure were fabricated by a process similar to that for CIGS device fabrication. A 60-nm thick CBD-CdS buffer layer was deposited onto the AIGS absorber layer. 60 nm-thick non-doped ZnO and 500 nm-thick ZnO:Al thin films were then subsequently deposited using rf magnetron sputtering at room temperature. The solar cells were air-annealed at 200 ºC for 2 hours to improve cell performance. Current-voltage characteristics of the devices were measured under AM1.5, $100mW/cm^2$ illumination at 25°C.

Table 1 shows the cell performance of the chalcopyrite top cells fabricated on the TCO back contacts. We have achieved a 9.6% total-efficiency solar cell using a Mo back contact. However, the cell performance decreased for the devices using an ITO back contact. This is due in part to the highly-resistive Ga_2O_3 thin layer at the ITO/AIGS interface formed during the AIGS deposition process.

Table 1 Cell performance of chalcopyrite top cells fabricated on TCO back contacts

Absorber /TCO	Eg (eV)	V_{OC} (mV)	J_{SC} (mA/cm^2)	FF	Efficiency (%)	Institute
AIGS/ITO	1.7	883	15.4	0.538	7.3	AGU
AIGS/IMO	1.7	891	15.7	0.441	6.2	AGU
CGS/FTO	1.68	864	15.4	0.513	6.8	NREL [4]
CGS/ITO	1.68	700	12.8	0.485	4.3	HMI [7]

EBIC analysis revealed the depletion layer in the AIGS device is significantly wider than that of other wide-gap devices such as $CuGaSe_2$ [5]. This suggests that the junction structure is different from that of Cu-chalcopyrite solar cells. We also fabricated AIGS top cell on a Mo-doped In_2O_3 (IMO) back contact. The efficiency was not essentially different from that of an AIGS cell made using an ITO back contact.

IDENTIFYING TECHNICAL ISSUES FOR 2-TERMINAL CIGS-BASED TANDEM CELLS

As mentioned above, it is well known that one of the most important issues for realizing CIGS-based tandem solar cells is to achieve high-efficiency top cells. However, other technical issues for CIGS-based tandem solar cells have not been fully understood yet. We thus have investigated the cell performance of tandem devices using a ZnO:Al/ZnO/CdS/AIGS/ITO top cell and a ZnO:Al/ZnO/CdS/CIGS(30%Ga)/Mo bottom cell in order to identify these other technical issues.

Table 2 Cell performance of a two-terminal mechanical stacked tandem cell, together with that of an AIGS top cell fabricated on ITO back contact and filtered CIGS (30% Ga) bottom cell.

Cell	V_{OC} (V)	J_{SC} (mA/cm^2)	FF	Efficiency (%)
Tandem	1.462	6.0	0.748	6.6
Top	0.912	13.7	0.538	6.7
Filtered bottom	0.548	5.6	0.688	2.1
Bottom	0.607	30.7	0.713	13.3

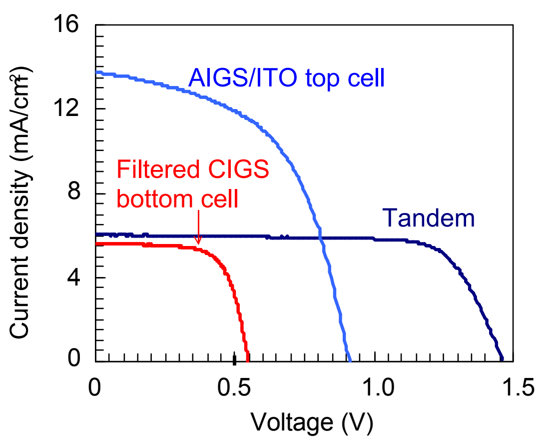

Fig. 1. J-V characteristics of a two-terminal mechanical stacked tandem cell, an AIGS top cell fabricated on ITO back contact, and a CIGS (30%Ga) bottom cell underneath the AIGS top cell.

Figure 1 shows the J-V characteristics of a two-terminal tandem cell, an AIGS top cell fabricated on ITO, and a CIGS (30%Ga) bottom cell underneath the AIGS device (namely, a filtered CIGS bottom cell with an AIGS top cell). The cell performance of the tandem cell is listed in Table 2, together with that of the top and filtered bottom cells. As can be seen in this table, a 6.6% total-area (0.5cm^2) efficiency was obtained with an open-circuit-voltage (V_{oc}) of 1.46 V which is the highest voltage reported so far among CIGS-based tandem solar cells. This high V_{oc} value is mainly attributable to the considerably higher V_{oc} (912 mV) of the AIGS top cell. It is clear that the J_{sc} of a 2-terminal tandem device is limited by the low current of a filtered CIGS bottom cell because of the current mismatch between top and filtered bottom cells, resulting in a relatively low efficiency tandem device.

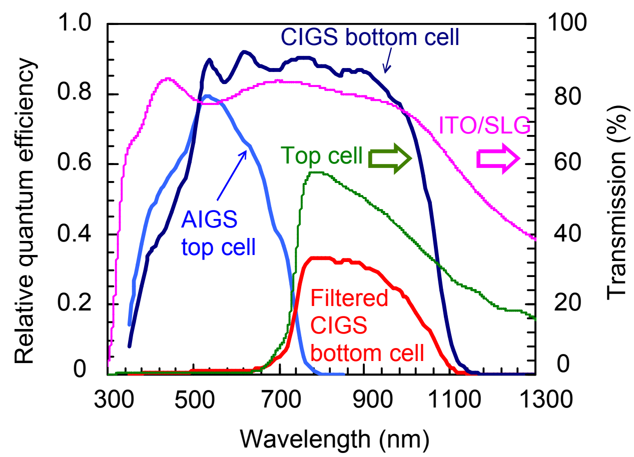

Fig. 2 QE curves of an AIGS top cell and a filtered CIGS (30% Ga) bottom cell and optical transmission curves of the top cell and ITO/soda-lime glass (SLG).

One of the possible reasons behind the low current of the filtered bottom cell is a low quantum efficiency (QE) at long wavelengths. Another reason for the low QE is the wider bandgap of the CIGS bottom cell (30% Ga) used in this preliminary experiment. Figure 2 shows the QE curves of an AIGS top cell and a filtered CIGS (30%Ga) bottom cell as well as the optical transmission curves of the top cell and ITO/soda-lime glass (SLG). The QE curve of the CIGS bottom cell fabricated on a Mo back contact is also included for reference. It is obvious that the QE of the filtered CIGS bottom cell drops at long wavelengths as compared to that of the CIGS device. On the other hand, a remarkable drop in the optical transmission of the AIGS top cell is observed at long wavelengths. This is mainly due to the use of ITO back contacts that exhibit large free carrier absorption as shown in Fig. 2. Therefore, further improvement of the QE or J_{sc} of the filtered bottom cell is expected by replacing ITO with other high-mobility TCO's.

IMPACT OF HIGH MOBILITY IMO BACK CONTACT TOP CELLS

Transparent conducting Mo-doped In_2O_3 (IMO) thin films were deposited on SLG substrates by RF co-sputtering using non-doped In_2O_3 and metallic Mo targets. 200-nm thick IMO thin films showed a mobility of 94 cm^2/Vs, a carrier concentration of 4.4×10^{20} cm^{-3}, and a resistivity of 1.5×10^{-4} ohm·cm. Details will be reported elsewhere [7]. The AIGS top cell was fabricated using high mobility IMO back contacts. Figure 3 shows the QE curves of the AIGS/IMO top cell, the filtered CIGS bottom cell, and the CIGS (30%Ga) bottom cell. The optical transmission curves of the top cell and IMO/SLG have been included for comparison. It should be noted that the QE of the filtered CIGS bottom cell improved at long wavelengths. This is mainly due to the high transmission of high-mobility IMO back contact, which results in high optical transmission for the top cell.

Table 3 Cell performance of a two-terminal mechanical stacked tandem cell, an AIGS top cell fabricated on an IMO back contact and a filtered CIGS (30% Ga) bottom cell.

Cell	V_{OC} (V)	J_{SC} (mA/cm^2)	FF	Efficiency (%)
Tandem	1.218	9.0	0.601	6.6
Top	0.896	16.3	0.426	6.2
Filtered bottom	0.520	8.5	0.639	2.8
Bottom	0.589	35.8	0.656	13.8

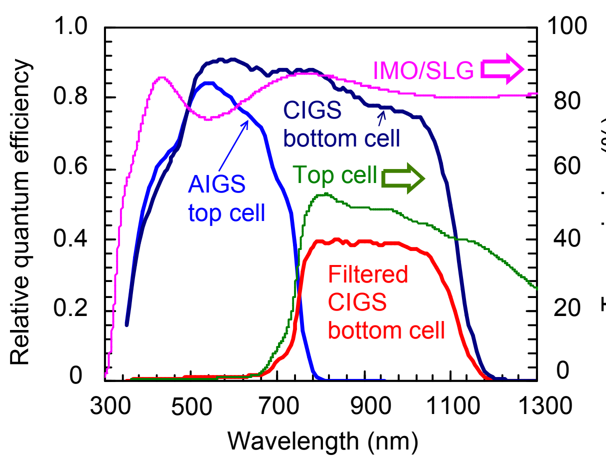

Fig. 3 QE curves of an AIGS/IMO top cell, a filtered CIGS bottom cell, and a CIGS (30% Ga) bottom cell. The optical transmission curves of the top cell and IMO/SLG have been included for reference.

The cell performance of the tandem cell is listed in Table 3, together with that of the top and filtered bottom cells. As can be seen in this table, the J_{sc} of the filtered CIGS (30% Ga) bottom cell increased. As a result, the J_{sc} of the tandem cell increased to 9 mA/cm^2. These results indicate that high-mobility TCO's such as IMO are very effective to increase the J_{sc} of the filtered bottom cell. As for a 4-terminal tandem device structure, 9 % efficiency was achieved as can be seen in the table. The reported efficiency is almost the same as that of a tandem device with a CGS top cell on a SnO_2:F back contact in conjunction with a CIS bottom cell [4].

EFFECT OF LOW-GA CONTENT BOTTOM CELLS

As mentioned above another reason for the low QE of the filtered CIGS bottom cell is due to the relatively large Ga content of 30% in this preliminary experiment. Therefore, the QE of the filtered bottom cell can be increased by replacing the CIGS (30% Ga) layer with low-Ga content CIGS or CIS. We thus have fabricated tandem solar cells using CIGS bottom cells with different Ga content and AIGS top cells fabricated with IMO back contacts.

Figure 4 shows the QE curves of the filtered CIGS bottom cells fabricated with 0, 15 and 30 % Ga content. QE curves of a CIGS bottom cell fabricated on a Mo back contact have also been included. It is obvious that the fall-off at long wavelengths in the QE curves shifted toward long wavelengths as the Ga content decreased, since the bandgap energy becomes narrower. Table 4 shows the cell performance of a tandem device together with that of an AIGS top cell fabricated on an IMO back contact and a filtered CIS (0% Ga) bottom cell. As can be seen in this table, the J_{sc} of the filtered CIS bottom cell increased when CIGS (30%Ga) was replaced with CIS (0% Ga). As a result, the J_{sc} of the tandem cell increased to 9.4 mA/cm^2. However, the V_{oc} value became slightly less. We thus fabricated a low Ga content CIGS (15% Ga) bottom cell to improve both the V_{oc} and FF of the tandem device.

Table 4 Cell performance of a tandem device together with that of an AIGS top cell fabricated on an IMO back contact and a filtered CIS (0% Ga) bottom cell.

Cell	V_{OC} (V)	J_{SC} (mA/cm^2)	FF	Efficiency (%)
Tandem	1.176	9.4	0.666	7.3
Top	0.859	15.3	0.464	6.1
Filtered bottom	0.391	9.5	0.656	2.4
Bottom	0.425	35.1	0.654	9.8

Tables 5 and Fig. 5 shows the cell performance of the tandem cell together with that of the AIGS top cell fabricated on an IMO back contact and a filtered CIGS (15% Ga) bottom cell. The V_{oc} and FF of the filtered CIGS (15% Ga) cell increased when the CIS bottom cell was replaced by a 15% Ga CIGS cell, although J_{sc} decreased slightly. As a result, an 8% total-area efficiency 2-terminal tandem device was achieved. Further improvement in the cell performance of chalcopyrite-based tandem devices can be expected by current matching due to using thickness control and by optimizing the deposition conditions of the AIGS top cells.

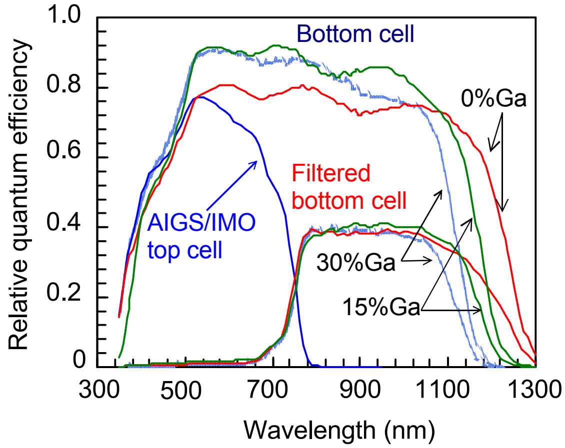

Fig. 4. QE curves of filtered CIGS bottom cells fabricated with 0, 15 and 30 % Ga content. The QE curves of a CIGS bottom cell fabricated on a Mo back contact have also been included.

Table 5 Cell performance of a tandem cell together with that of an AIGS top cell fabricated on an IMO back contact and a filtered CIGS (15% Ga) bottom cell.

Cell	V_{OC} (V)	J_{SC} (mA/cm^2)	FF	Efficiency (%)
Tandem	1.305	9.1	0.675	8.0
Top	0.823	13.7	0.473	5.3
Filtered bottom	0.490	9.2	0.713	3.2
Bottom	0.524	37.1	0.711	14.3

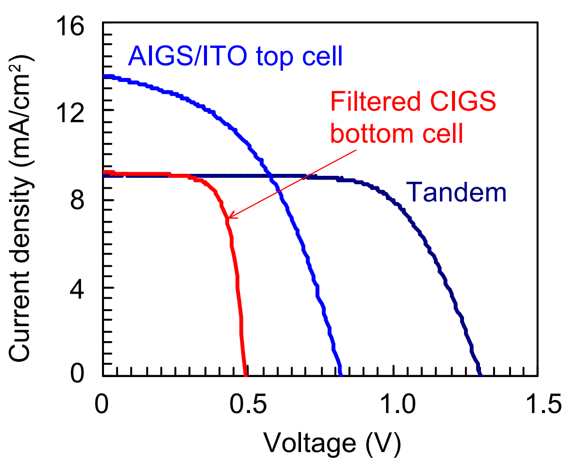

Fig. 5. The J-V characteristics of the tandem cell together with that of the AIGS top cell fabricated on an IMO back contact and a filtered CIGS (15% Ga) bottom cell.

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