ULTRA-THIN, TRIPLE-BANDGAP GaInP/GaAs/GaInAs MONOLITHIC TANDEM SOLAR CELLS

National Renewable Energy Laboratory (NREL), Golden, CO 80401

INTRODUCTION

The performance of state-of-the-art, series-connected, lattice-matched (LM), triple-junction (TJ), III-V tandem solar cells could be improved substantially (10-12%) by replacing the Ge bottom subcell with a subcell having a bandgap of ~1 eV. For the last several years, research has been conducted by a number of organizations to develop ~1-eV, LM GaInAsN to provide such a subcell, but, so far, the approach has proven unsuccessful. Thus, the need for a high-performance, monolithically integrable, 1-eV subcell for TJ tandems has remained.

In this paper, we present a new TJ tandem cell design that addresses the above-mentioned problem. Our approach involves inverted epitaxial growth to allow the monolithic integration of a lattice-mismatched (LMM) ~1-eV GaInAs/GaInP double-heterostructure (DH) bottom subcell with LM GaAs (middle) and GaInP (top) upper subcells. A transparent GaInP compositionally graded layer facilitates the integration of the LM and LMM components. Handle-mounted, ultra-thin device fabrication is a natural consequence of the inverted-structure approach, which results in a number of advantages, including robustness, potential low cost, improved thermal management, incorporation of back-surface reflectors, and possible reclamation/reuse of the parent crystalline substrate for further cost reduction.

Our initial work has concerned GaInP/GaAs/GaInAs tandem cells grown on GaAs substrates. In this case, the 1-eV GaInAs experiences 2.2% compressive LMM with respect to the substrate. Specially designed GaInP graded layers are used to produce 1-eV subcells with performance parameters nearly equaling those of LM devices with the same bandgap (e.g., LM, 1-eV GaInAsP grown on InP).

Previously, we reported preliminary ultra-thin tandem devices (0.237 cm²) with NREL-confirmed efficiencies of 31.3% (global spectrum, one sun) (1), 29.7% (AM0 spectrum, one sun) (2), and 37.9% (low-AOD direct spectrum, 10.1 suns) (3), all at 25°C. Here, we include recent results of testing similar devices under the concentrated AM0 spectrum, and also present the first demonstration of a high-efficiency, ultra-thin GaInP/GaAs/GaInAs tandem cell processed on a flexible kapton handle.

APMOVPE GROWTH PARAMETERS

The GaInP/GaAs/GaInAs tandem structures discussed here were grown using atmospheric-pressure metalorganic vapor-phase epitaxy (APMOVPE) in a home-built system at NREL. Trimethylindium, triethylgallium, trimethylgallium, trimethylaluminum, arsine, and phosphine were used as the primary reactants, with hydrogen selenide, disilane, carbon tetrachloride, and diethylzinc used as the doping precursors. Growth on GaAs substrates was performed at temperatures ranging from 600 to 700°C in a purified hydrogen ambient.
The TJ tandem devices investigated in our preliminary tests have been grown on GaAs substrates, but similar devices could also be fabricated using Ge substrates. As illustrated in Fig. 1, from the substrate up, the tandem structure consists of the following components: a LM n-GaInP etch-stop layer, a LM n-GaAs contact layer, a LM n/p-GaInP/AlInP DH subcell, a LM p'/?-GaAs tunnel junction, a LM n/p-GaAs/GaInP DH subcell, a LM p'?/?-GaAs tunnel junction, a LMM n-GaInP compositionally step-graded layer, a LMM n/p-GaInAs/GaInP DH subcell, and a LMM p'-GaInAs contact layer. Further details of the tandem structure, and a general processing sequence for handle-mounted, ultra-thin devices, have been published previously (1).

Some of the important advantages of our new approach are listed below:

1) The handle material can be chosen to have a wide range of advantageous characteristics (e.g., mechanical strength, flexibility, specific electrical/optical parameters, high thermal conductivity, low cost, etc.).
2) Thermal management can be optimized since the ultra-thin device layers can be placed directly on a heat sink.
3) A back-surface reflector can be easily incorporated on the back side of the LMM bottom subcell, which is grown last.
4) Reuse/reclamation of the parent substrate is also possible, resulting in substantially reduced cost.
5) Effective co-generation of heat and electric power is possible since the new TJ tandem cells do not absorb photons with energies less than ~1 eV.
6) Monolithically interconnected module (MIM) devices are easily realizable by mounting the ultra-thin tandems on...
an electrically insulating material.

7) The basic concept can be expanded to include numerous subcells for increased performance (NREL patent pending (4)).

Additionally, the key advantages of including a back-surface reflector on the back side of the LMM bottom subcell are as follows:
1) The GaInAs/GaInP DH subcell is grown to 1/2 the usual thickness, which translates to less growth time and lower $J_0$ due to the "narrow diode" effect (~20 mV improvement in $V_{oc}$).
2) The back-surface reflector reflects away sub-bandgap photons leading to a reduced operating temperature.
3) Photon escape due to radiative recycling is also reduced, which also lowers $J_0$.

SEMI-REALISTIC TANDEM PERFORMANCE MODELING

We have performed semi-realistic modeling calculations, based on a rigorous approach for series-connected tandem subcells (5), to serve as a guide for the choice of the bottom subcell bandgap, and to predict potential performance, under operating conditions relevant to space. We assume that the bottom subcell quantum efficiency is 0.95 (spectrally independent) in the calculations. Also, the top and middle subcells are fixed to be GaInP (1.87 eV) and GaAs (1.42 eV), respectively. For space applications, we modeled for the AM0 spectrum at one sun, 25°C, and obtained an optimum bottom subcell bandgap of 1.02 eV, with a tandem conversion efficiency of ~33% (current state of the art is ~30%). At 10 suns concentration, the modeled efficiency increases to ~36%.

PROPERTIES OF ~1-eV GaInAs/GaInP DH LMM SUBCELLS

The characteristics of the LMM bottom subcells are of particular interest due to the potential deleterious impact of crystalline defects in the active subcell layers. Cross-sectional transmission electron microscopic characterization of the transparent GaInP graded region and GaInAs/GaInP subcell layers shows that misfit and threading dislocation networks are present within the GaInP compositionally step-graded layers, but are not visible in the active subcell layers. The coherence between the top of the grade and the active layers is quite apparent. Plan-view cathodoluminescence images reveal active threading dislocations in the GaInAs layers, with an average areal density of 2x10^6 cm^-2. Typical device performance data for the 1-eV subcells show that the losses due to the dislocations are quite small. Internal quantum efficiency data range from 95 to 100% for photon energies ranging from the band edge (~1 eV) to the bandgap of GaAs (1.42 eV), respectively. Additionally, open-circuit voltages of 0.56–0.58 V are routinely observed for photocurrent densities of 15–20 mAcm^-2, which compares favorably with ~0.60 V that we obtain for LM, 1-eV GaInAsP/InP cells tested under similar photoexcitation.

GaInP/GaAs/GaInAs TANDEM PERFORMANCE

In an initial effort, we have successfully grown, processed, and tested monolithic, series-connected, handle-mounted, ultra-thin GaInP/GaAs/GaInAs tandem solar cells. Performance data for the best device fabricated to date are included in this section of the paper. Quantum efficiency (QE) and reflectance (R) data are given in Fig. 2. The data generally show excellent carrier collection across a broad spectral range for all of the subcells. The R data, however, show that photocurrent gains are still possible at the far edges of the tandem response range. Improving the two-layer ZnS/MgF$_2$ ARC will be a focus of future work. Interference effects are also observed in the QE data for the 1.02-eV bottom subcell, which occur because the subcell is optically thin with a back-surface reflector, causing it to behave like a Fabry-Perot cavity. The interference effects are also evident in the R data over the response range of the bottom subcell. It is important to note that the QE for the bottom subcell is excellent despite its 2.2% LMM with respect to the GaAs substrate. We have also tested the new TJ tandem cells under mild AM0 solar concentration for the first time. The cells tested were designed for one-sun operation, but had sufficiently low resistance to allow peak performance at 4-10 suns concentration. Conversion efficiency data as a function of concentration ratio are shown in Fig. 3 for a TJ tandem cell with an area of 0.243 cm$^2$. The measurements were performed using a water-filtered Xe lamp source, a cell temperature of 25°C, and a concentration ratio based on one-sun data obtained from our X25 multi-source solar simulator (AM0 reference spectrum). As shown in Fig. 3, the efficiency rises rapidly from one to four suns, and then peaks at 31.4% at 8.9 suns. Thereafter, the efficiency drops quickly with increased concentration. The peak efficiency value of ~31-32% at such a low concentration ratio is particularly encouraging considering that the tandem cells are in an early stage of development and are far from being optimized. Also, the demonstrated high performance at low
concentration is well suited to low-concentration systems such as the Stretched Lens Array being developed by Entech, Inc.

The data shown in Fig. 4 elucidate the behavior of the data in Fig. 3. Here, open-circuit voltage ($V_{oc}$) and fill factor (FF) data for the same tandem cell are plotted as a function of the concentration ratio. The rapid increase in efficiency from one to four suns results from both $V_{oc}$ and FF rising strongly over this range. The FF peaks at ~4 suns then decreases sharply with increasing concentration due to resistive losses. The $V_{oc}$ shows a reduced rate of increase with concentration beyond ~4 suns, which we believe is due to a transition from $n=2$ to $n=1$ diode behavior, principally in the LMM bottom subcell. The net result of the above trends is that the efficiency peaks at ~9 suns.

Current-voltage data for one of the first the tandem cells processed on a flexible kapton handle are shown in Fig. 5. To our knowledge, the one-sun AM0 efficiency value of 26.5% represents a new record for a flexible solar cell. The high tandem $V_{oc}$ suggests that the quality of the subcell junctions is excellent. With continued development, we see no fundamental reason why the performance of flexible tandem cells will not equal that of their rigid counterparts.

CONCLUSION

We have described a new approach for ultra-high-efficiency tandem solar cells based on inverted III-V heteroepitaxial epistructures that combine both LM and LMM component subcells in a monolithic structure. The tandem epistructures are fabricated into handle-mounted, ultra-thin devices, which have many advantages, and potential realistic AM0 conversion efficiencies in the 33-36% range. In initial work, we have demonstrated ultra-thin GaInP/GaAs/GaInAs tandem cells with exciting performance levels processed using both rigid and flexible handles.

A number of research issues remain in order to move the new tandem cell technology from laboratory-scale demonstrations to potential commercial production. A cost-effective, high-yield processing scheme for large-area, handle-mounted, ultra-thin tandem devices must be explored and developed. Also, accurate performance testing in the laboratory is a difficult issue, particularly for series-connected tandem cells that have near-optimal subcell bandgaps. Testing under concentration adds an additional level of difficulty; a multi-source concentrator simulator may need to be developed. The tandem cell testing problem will only become more complicated as tandem cells with more than three bandgaps become available.

ACKNOWLEDGEMENTS
The authors gratefully acknowledge the support of the U. S. Department of Energy (under Contract DE-AC36-99GO10337) for this work. We also thank Charlene Kramer and Michelle Young for assistance with the epitaxial growth and device processing, respectively, of the ultra-thin GaInP/GaAs/GaInAs tandem devices.

REFERENCES


Figure 4. \( V_{OC} \) and FF as a function of AM0 concentration ratio for a TJ GaInP/GaAs/GaInAs tandem solar cell.

Figure 5. Current-voltage data for a TJ GaInP/GaAs/GaInAs ultra-thin tandem solar cell mounted on a kapton handle.