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Investigation of Polycrystalline Thin-Film CuInSe$_2$ Solar Cells Based on ZnSe Windows

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ABSTRACT

Investigations of ZnSe/CIS solar cells are being carried out in an effort to improve the efficiencies CIS cells and to determine if ZnSe is a viable alternative to CdS as a window material. MOCVD growth of ZnSe is accomplished in a SPIRE 500XT reactor housed in the Electronic Materials Laboratory at WSU Tri-Cities by reacting a zinc adduct with H_2Se. Conductive n-type ZnSe is grown by using iodine as a dopant. Ethyliodide was mixed with helium and installed on one of the gas lines to the system. ZnSe films have been grown on CIS substrates at 200°C to 250°C. ZnO is also being deposited by MOCVD by reacting tetrahydrofuran (THF) with a zinc adduct. ZnSe/CIS heterojunctions have been studied by growing n-ZnSe films onto 2 cm x 2 cm CIS substrates diced from materials supplied by Siemens and then depositing an array of aluminum circular areas 2.8 mm in diameter on top of the ZnSe to serve as contacts. Al films are deposited with a thickness of 80 to 100 Å so that light can pass through the film, thus allowing the illuminated characteristics of the ZnSe/CIS junction to be tested. Accounting for the 20 to 25 % transmittance through the Al film into the ZnSe/CIS structure, current devices have estimated, active-area AM1.5 efficiencies of 14 %. Open circuit voltages > 500 mV are often attained.

1. INTRODUCTION AND BACKGROUND MATERIAL

This report concerns work carried out during the first year of a three year effort to investigate $CuInSe_2$ (CIS) and $CuIn_{1-x}Ga_xSe_2$ (CIGS) solar cells based on ZnSe windows. Background information, program objectives and the technical approach are discussed in the remainder of this section, and technical progress made during the first year is discussed in subsequent sections.

1.1 Background Material

The key objective of this effort is to determine if ZnSe represents a viable alternative to CdS for the wide bandgap, n-type window layer in CIS cells. An alternative to CdS is desirable for several reasons: (1) Cd is toxic and thus presents problems concerning safety; (2) an alternative heterojunction may be required in order to improve upon values of FF and V_{oc} exhibited by CdS/CIS and CdS/CIGS cells; (3) the lattice constant of ZnSe (5.6676 Å) is more appropriate for heterojunctions formed with wider bandgap CIGS materials.

Only two studies of ZnSe/CIS cells appear to have been reported. Nouhi, et al, reported on ZnSe/CIS solar cells fabricated by depositing ZnSe films onto CIS substrates supplied by industrial laboratories [1]. ZnSe films were deposited by reactive magnetron co-sputtering of Zn and In dopant in Ar/H_2Se. The ZnSe films were relatively low in resistivity, say, 20 ohm-cm. Efficiencies of 7 to 8.5% were obtained with ARCO supplied CIS substrates. The performance of these cells is encouraging. The study did not investigate a very large range of processing variables, however. Thus, a more extensive experimental study of ZnSe/CIS solar cells is clearly justified.

The other reported work was by Yoo, et al. [2]. This study involved fabrication of cells with a thin insulating layer of ZnSe between the base CIS layer and the CdS emitter. Cells were fabricated by depositing ZnSe onto CIS substrates provided by ARCO. The ZnSe films were deposited by physical vapor deposition. Cells exhibited low efficiencies primarily due to the ZnSe layers being highly resistive. The study particularly emphasized dark and light I-V analyses. The resistive ZnSe layer caused photocurrent suppression, and poor fill factors. This study along with that of Nouhi, et. al. indicates that a conductive layer of ZnSe should be utilized for the emitter layer in a ZnSe/CIS heterojunction cell.

1.2 Program Objectives

The primary objectives of this program are to improve the performance of CIS-based and CIGS-based solar cells, and to provide an alternative to CdS for the window layer material for such cells by developing and testing the use of ZnSe window layers. The use of ZnSe window layers is expected to lead to solar cell structures with a conversion efficiency of 14 %. A discussion of the technical approach being used to meet these objectives follows.

1.3 Technical Approach

Ultimately, cells will be fabricated with a structure as described by Figure 1A. The appropriate electron band structure is shown in Figure 1B. In order to fabricate ZnSe/CIS cells, procedures must be developed for growing conductive ZnSe on CIS and to complete the device structure by depositing conductive ZnO. To attain these goals, the program is structured into three tasks: (1) CIS and CIGS cells with ZnSe windows; (2) Material and device characterization; and, (3) Device modeling. Task 1 comprises the major thrust of the program. This task involves acquisition and characterization of CIS substrates, MOCVD growth of ZnSe and ZnO and fabrication of ZnO/ZnSe/CIS solar cells. Task 2 includes T-I-V and photoresponse analyses of completed cells and Al/CIS Schottky barriers. Finally, Task 3 concentrates on device modeling to support experimental studies. In particular, the utility of PC-1D software for analysis of CIS solar cell performance is being evaluated. PC-1D utilizes a finite element numerical approach to solving the basic semiconductor equations.

Results obtained for each of these tasks are discussed in the remainder of this report.

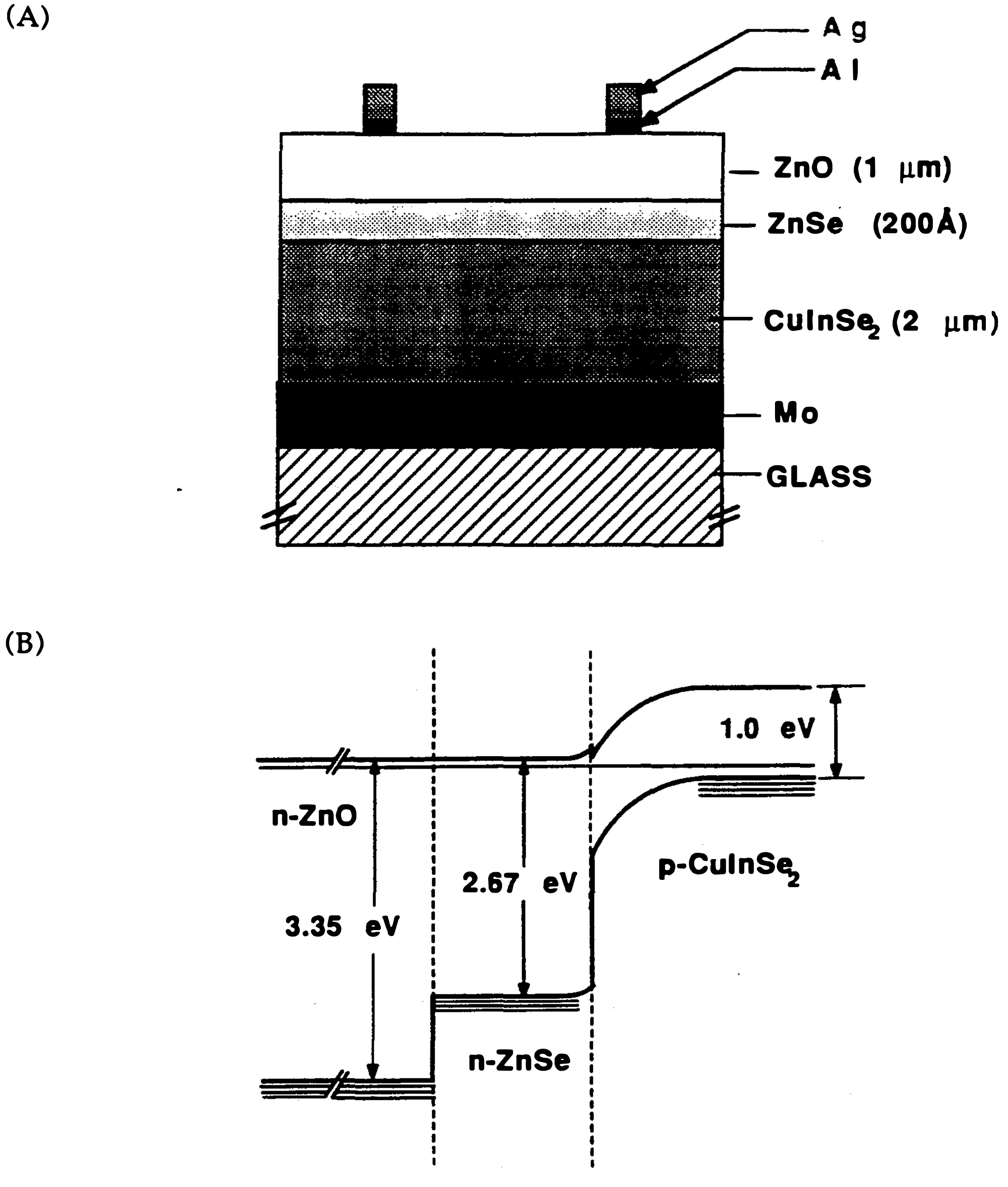

Figure 1. (A) Planned Structure of ZnO/ZnSe/CIS Cell; (B) Electron Band Diagram for Cell Structure.

2. CIS CELLS WITH ZnSe WINDOWS

Investigations of ZnSe/CIS heterojunction solar cells have involved four main activities, namely, substrate characterization, growth of the ZnSe/CIS heterojunction, ZnO deposition, and solar cell fabrication. Discussions of progress made the first year in each of these activities follows.

2.1 Substrate Characterization

CIS substrates were acquired from Siemens and ISET. Most of the work to date has been carried out with Siemens material, however. Thus, the work reported herein is based strictly on Siemens substrates. Siemens supplied 10 cm x 10 cm parts consisting of a CIS layer deposited onto Mo coated glass. These substrates were scribed and broken into nominally 2 cm x 2 cm die for ZnSe/CIS cell development.

Al/CIS Schottky barriers have been utilized in this program for diagnostic purposes. For example, Al/CIS barriers were fabricated to determine an approach to surface treatment prior to ZnSe deposition to form a heterojunction. Attempts to form an Al/CIS Schottky barrier by vacuum depositing aluminum on as-received CIS substrates resulted in very resistive interfaces. It was determined that etching the CIS substrate with either KBr/Br/H_2O or KCN (10 % by weight) aqueous solutions prior to Al deposition leads to consistent Al/CIS Schottky barrier properties. The use of these etches were discussed by McCandless and Birkmire [3], and Tuttle, et al. [4], respectively. Dark I-V curves for several small area barriers deposited onto substrate No. 120 are shown in Figure 2A. The individual diodes have areas of .06 cm^2. The current scale was chosen such that one division (0.5 mA) corresponds to a current density on the order of 10 mA/cm^2. This substrate was etched for 1 minute with 10% KCN solution. Etching with KBr,Br leads to similar results with the optimum etching time being approximately one minute. The dark characteristics for No. 120D are presented in Figure 2B on a semilog plot. These data were fit very well assuming two current mechanisms. Typical results of I-V analyses are discussed in Section 3.

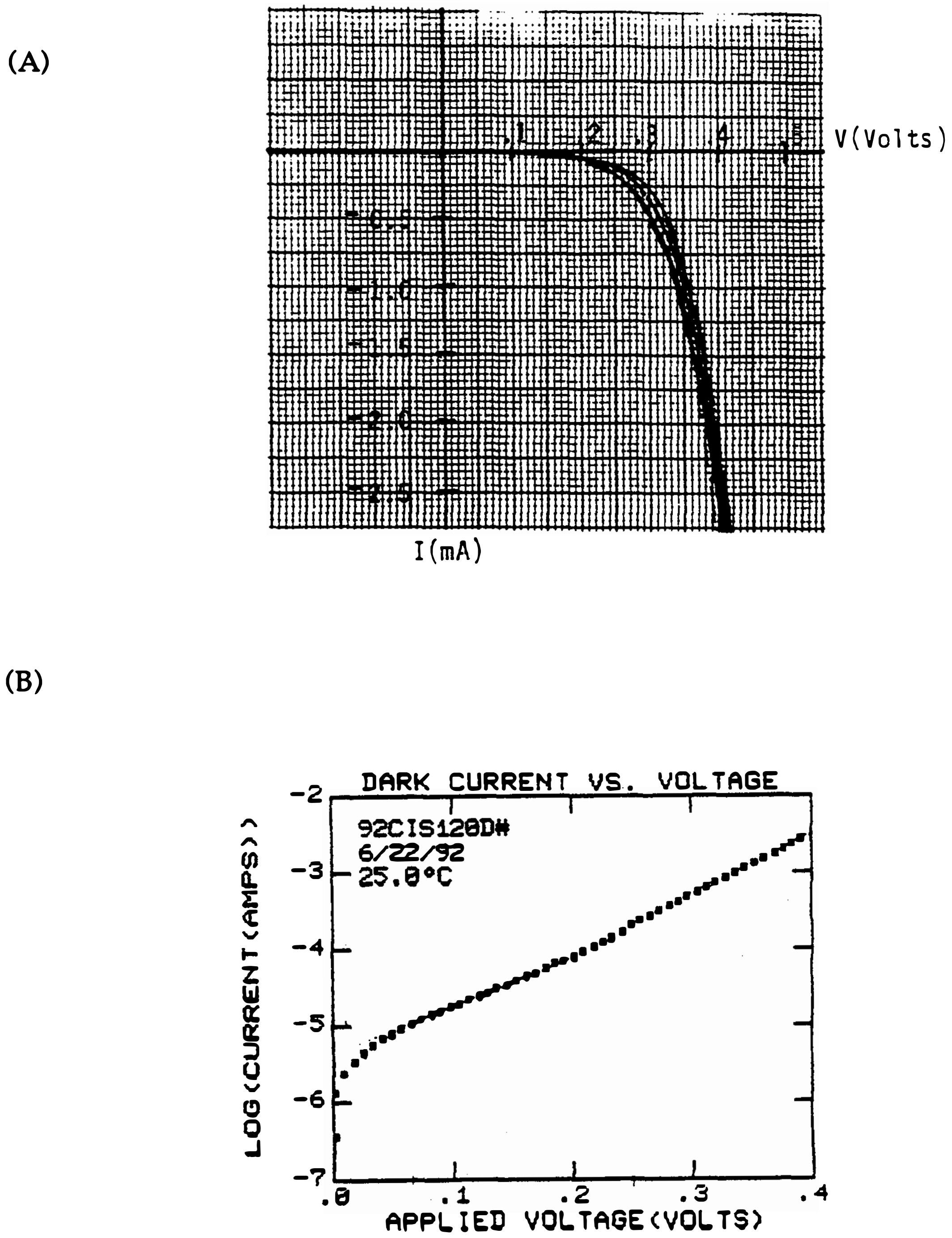

Figure 2. Dark I-V Characteristics for Al/CIS Schottky Barrier: (A) Data Taken with X-Y Recorder; (B) Digital Data Presented on a Semilog Plot.

2.2 ZnSe/CIS Heterojunctions

ZnSe/CIS heterojunctions are formed by depositing films of ZnSe (by MOCVD) on CIS substrates. MOCVD growth of ZnSe is accomplished in a SPIRE 500XT reactor housed in the Electronic Materials Laboratory by reacting a zinc adduct with H_2Se. The zinc adduct is formed by reacting dimethylzinc and triethylamine (TEN). It has been found that the use of the adduct reduces or eliminates the pre-reaction problems typical of ZnSe growth. The triethylamine was mixed to give a vapor pressure of 16 torr at 20°C. Growth rate of ZnSe is controlled by adjusting the flow of hydrogen through the metalorganic bubbler. The growth rate, in fact, varies linearly with the flow of hydrogen through the DMZn/TEN bubbler. Typical growth conditions that result in a ZnSe growth rate of 1 Å/s and a VI/II ratio of 5 are as follows: a total pressure af 65 torr, 6000 sccm of palladium-diffused hydrogen; 25 sccm hydrogen bubbled through the DMZn/TEN bubbler at 20°C; and 270 sccm hydrogen selenide (1% in H_2); and a substrate temperature of 250°C.

Procedures for growing n-type ZnSe by doping with iodine were also developed. Ethyliodide was mixed with helium (1000 ppm) and installed on one of the gas lines to the system. It is straight forward to grow ZnSe with a resistivity on the order of .05 ohm-cm on single crystal surfaces at 250°C. Iodine-doped ZnSe films grown on CIS have a much higher resistivity, but it is sufficiently low for heterojunction growth. The flow of the ethyliodide/helium mixture is usually set at 100 sccm.

ZnSe films grown on CIS substrates have a strong (111) orientation. Figure 3 gives X-ray diffraction results for a ZnSe film grown by MOCVD on a Siemens substrate at 200°C. Note the strong ZnSe (111) line intensity along with the CIS (112) line intensity.

ZnSe/CIS heterojunctions were studied by growing n-ZnSe films onto 2 cm x 2 cm CIS substrates and then depositing an array of aluminum circular areas 2.8 mm in diameter on top of the ZnSe to serve as a contacts. Al films are deposited with a thickness of 80 to 100 Å so that light can pass through the film. Figure 4 gives I-V characteristics for a diagnostic device illuminated by an ELH bulb simulator with the intensity adjusted to approximately

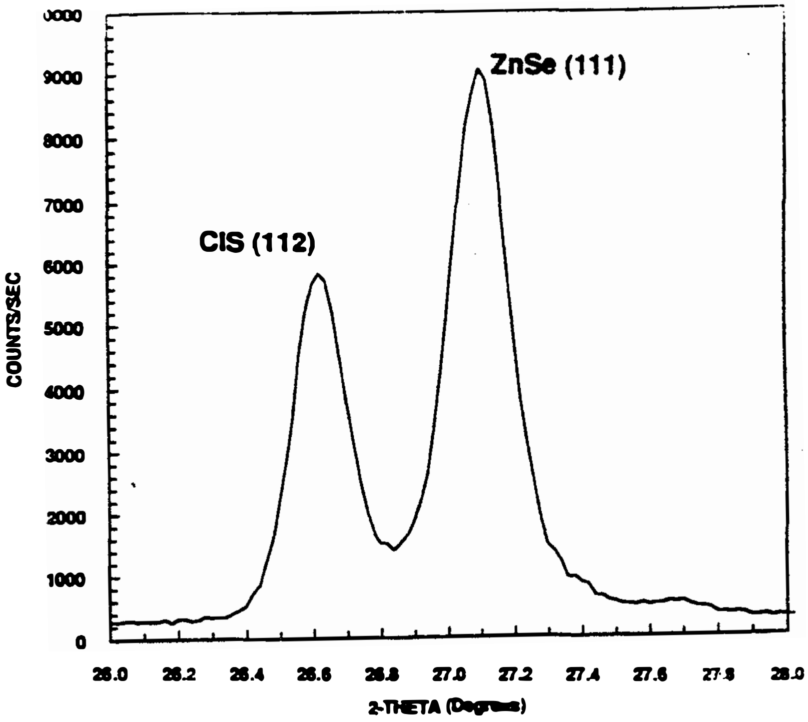

Figure 3. XRD Results for a 1 µm ZnSe Film Grown on a CIS Substrate

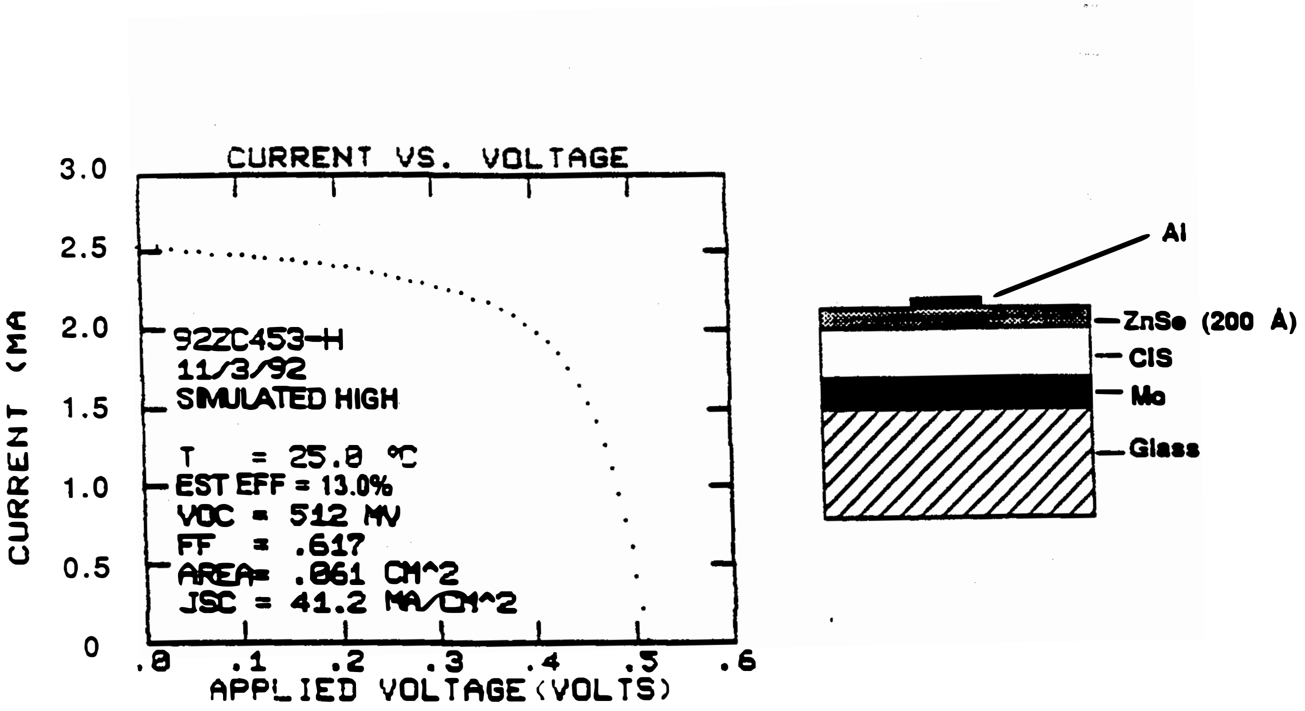

Figure 4. I-V Characteristics for a .06 cm^2 ZnSe/CIS Heterojunction with the Jsc adjusted to approximately 40 mA/cm^2.

compensate for the 20 to 25 % transmittance of the Al film. The insert depicts the device structure. The efficiency value is equivalent to an active-area number, since the light intensity is adjusted to achieve a value of Jsc of approximately 40 mA/cm^2. It is significant, however, that under conditions for which the photon flux entering the ZnSe/CIS structure is similar to that for AM1 illumination, the open-circuit voltage is 512 mV and the fill factor is 0.617. Once completed solar cell structures are fabricated which exhibit properties similar to 92ZC453-H, efficiencies greater than 12 % will be achieved.

2.3 ZnO Deposition

The WSU SPIRE 500XT reactor was modified for ZnO growth. Tetrahydrofuran (THF) was utilized as a source of oxygen, while the zinc adduct used for the growth of ZnSe was also utilized for ZnO growth. Thus, tetrahydrofuran (THF) is reacted with the zinc adduct to grow ZnO. The THF liquid is stored in a room temperature bubbler. Introduction of THF vapor into the reactor is controlled by a PFD Model 1004 gas flow control system which was modified for this purpose from its originally intended use of controling the flow of TCA vapor into an oxidation tube furnace. THF has been used with DMZ by Wright [5] and Wessels [6] to grow ZnO at substrate temperatures ≥ 300°C. THF was selected as a source of oxygen because of other film growth carried out in the WSU reactor. In particular, oxygen and H_2O were not considered as precursors since the WSU reactor is also used for GaAs and AlGaAs growth.

Three runs have been carried out to grow ZnO on substrates at 350°C. The ZnO films were not intentionally doped, but exhibited resistivities from .02 to 6 ohm-cm. Future work will involve further characterization of the system for growth of undoped and doped ZnO. It will be particularly important to investigate MOCVD growth of ZnO at temperatures below 350°C.

2.4 Solar Cell Fabrication

Although there is much to learn regarding the ZnSe/CIS heterojunction, some effort has been devoted to fabrication of a complete cell, that is, a structure as depicted by Figure 1. The remaining processing step required is the deposition of ZnO. Two approaches have been taken: (1) MOCVD growth of ZnO at WSU; (2) and to send ZnSe/CIS structures to Siemens for deposition of a film of low resistivity ZnO. As discussed above, procedures for MOCVD growth of ZnO are still being developed at WSU. However, preliminary results indicate that MOCVD grown ZnO with a resistivity on the order of 0.5 ohm-cm tends to short a CIS cell with a thin ZnSe window layer, whereas ZnO with a resistivity of > 5 ohm-cm does not short such a cell. One device configuration consisting of 1.5 μm of ZnO deposited by MOCVD on a ZnSe/CIS structure provided encouraging results. Figure 5 gives illuminated characteristics for a test device as described by the insert. Again, the light intensity was adjusted to compensate for the 20 to 25 % transmittance of the Al film. The ZnO film was approximately 1.5 μm

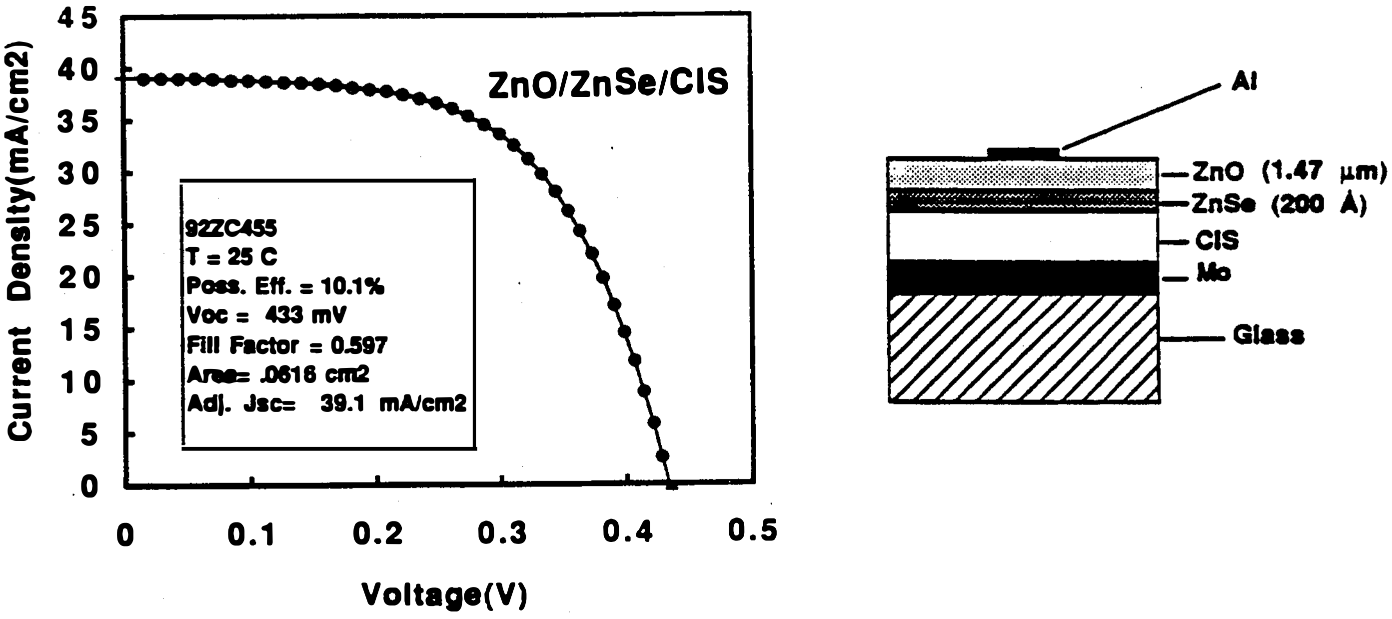

Figure 5. I-V Characteristics for a .06 cm^2 ZnO/ZnSe/CIS Device with the Jsc adjusted to approximately 40 mA/cm^2.

thick and had a resistivity of 6 ohm-cm. Measurement of I-V characteristics between two Al circular areas indicates that a small barrier exists between Al and ZnO. Thus, the illuminated I-V characteristics shown in Figure 5 are affected by contact resistance between Al and ZnO. We have verified that Al will make a reasonable contact with low resistance ZnO by measuring the resistance between two Al dots deposited onto ZnO. The properties of this test device are particularly significant since growth of the ZnO film onto Sample 92ZC455 was done at 350°C for one hour. Thus, it appears that the ZnSe/CIS interface is rather stable once it has been established. It is also significant that the reaction of THF and the zinc adduct during this particular growth run did not result in photocurrent suppression as observed when ZnO has been deposited on ZnSe/CIS structures by Siemens (see below). Much more effort will be required in order to develop procedures for MOCVD growth of ZnO, but encouraging results have been obtained.

Two sets of devices were sent to Siemens for ZnO deposition with their proprietary process. In both cases the ZnSe layers had a thickness (actually the thickness of ZnSe on a silicon witness) on the order of 200 Å. The first batch of samples was coated with low resistivity ZnO. These devices were essentially shunted. The second batch of devices was coated with MOCVD grown ZnO having a resistivity of 0.5 ohm-cm. Most of these devices were also shunted. Current voltage characteristics for a device that was not shunted are shown in Figure 6. In this case, the ZnO deposition process (apparently at Siemens) caused an effect resulting in a light-induced shunting mechanism. These results are very preliminary, and are not yet understood. At this point, however, it appears that the ZnSe/CIS junction will require a bilayer of ZnO for the top contact layer.

3. DEVICE CHARACTERIZATION

Device characterization has primarily involved dark and illuminated I-V analyses at room temperature. The approach utilized for the analyses is basically the same as previously described in Reference 7. Table 1 list results for a typical Al/CIS Schottky barrier, and a one of the best performing ZnSe/CIS heterojunctions. The I-V characteristics have been interpreted in

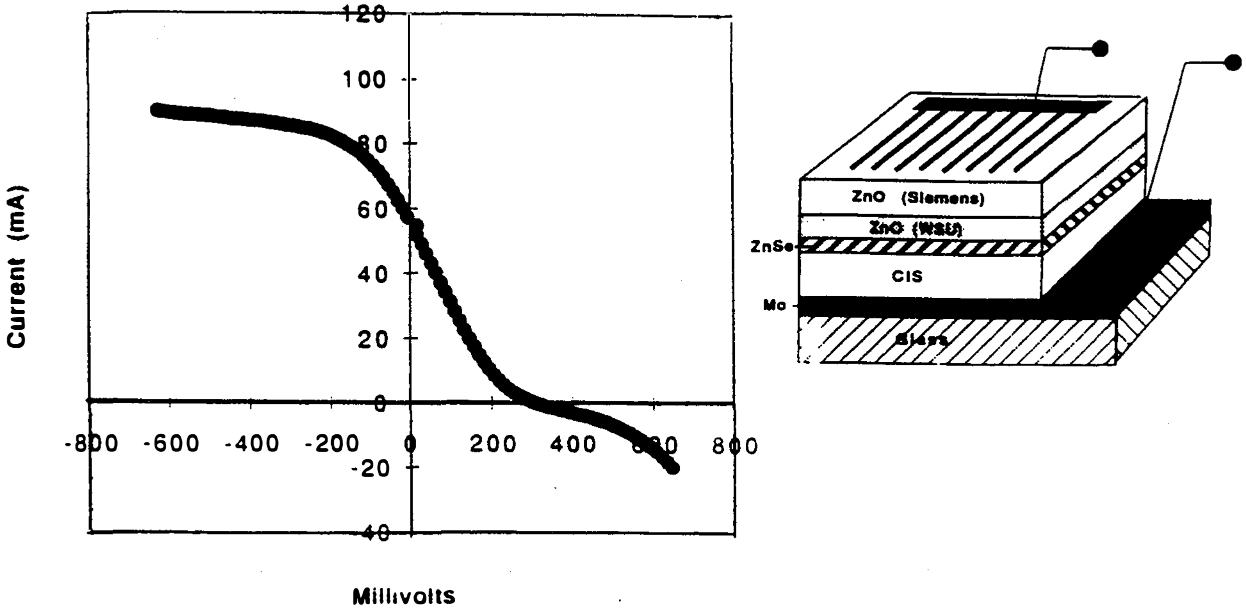

Figure 6. Illuminated I-V Characteristics for Cell 92ZC461 having a total area of 2.33 cm^2 under approximate simulated AM1 conditions.

terms of two loss mechanisms, one dominant in the low voltage range .15 to .35 Volts, and one at higher voltages, .35 to .60 Volts. The low voltage mechanism is probably due to multiple step tunneling. The high voltage mechanism for Al/CIS Schottky barriers appears to be due to depletion region recombination. Since the J_o for the Schottky barriers is two orders of magnitude higher than that observed for the ZnSe/CIS heterojunction, it appears that the recombination centers are much more numerous in the Schottky barriers. This result suggests that the recombination comprising the high voltage mechanism takes place near the interface. Thus, the relatively large open-circuit voltage observed for the ZnSe/CIS junctions may be a result of passivation of the CIS surface and underlying region provided by the ZnSe layer. The Al/CIS Schottky barrier may prove to be a valuable diagnostic device for investigating CIS solar cell current-voltage characteristics. It should be noted that if the Al/CIS Schottky barrier I-V characteristics were a result of thermionic emission, the A-value would be approximately 1.0 and the J_o value would be on the order of $1x10^{-12}$ A/cm^2,

TABLE 1 -- I-V PARAMETERS

Device	Rs•Area (Ohm•cm^2)	Low Voltage Range		High Voltage Range	
		J_o (A/cm^2)	A	J_o (A/cm^2)	A
Al/CIS(Dark)	0.3	6.2×10^{-4}	14.4	1.0×10^{-5}	1.8
ZnSe/CIS 453-H(Light)	.048	1.56×10^{-3}	9.4	1.2×10^{-7}	1.6

since the barrier height would be approximately 0.8 eV. Results concerning I-V analyses should be considered preliminary, since such properties as light biasing effects have not been investigated.

Physical characterization has primarily involved XRD studies. We expect to examine the physical structure of the ZnSe/CIS interface during the Phase II effort. In particular, SIMS profiles will be obtained in an effort to understand the effects of ZnO deposition on ZnSe/CIS interfaces.

4. DEVICE MODELING

Device modeling has involved a low level effort to evaluate the utility of PC-1D for modeling of CIS-based solar cells. This code has been used extensively by research groups investigating silicon and III-V devices. PC-1D utilizes a finite element numerical approach to solve the semiconductor equations [8]. Up to three regions of different material parameters can be used to define the device, each with its own doping profiles, electronic and optical properties. Recombination of electron-hole pairs can be defined in each region by S-R-H band to band transitions or through user-defined deep level transitions. Recombination at interfaces can also be taken into account. Property data files have been established for CIS and ZnSe to utilize in PC-1D. This computer code will eventually be used to examine the effect of the spatial location and position within the bandgap of an electron trap on depletion layer recombination. For example, dark current-voltage characteristics can be calculated and then compared to calculated results based on a simple, explicit expression for the dark current density, namely, $J = J_o \exp[V/AkT]$ with $V >> kT$. Then, one may be able to correlate A-values with trap locations. This procedure was utilized to discuss I-V characteristics of GaAs solar cells in Reference 9.

5. CONCLUSIONS

Investigations of ZnSe/CIS solar cells are being carried out in an effort to improve the efficiencies CIS cells and to determine if ZnSe is a viable alternative to CdS as a window material. MOCVD growth of ZnSe is accomplished in a SPIRE 500XT reactor housed in the Electronic Materials Laboratory at WSU Tri-Cities by reacting a zinc adduct with H_2Se. Conductive n-type ZnSe is grown by using iodine as a dopant. Ethyliodide was mixed with helium and installed on one of the gas lines to the system. ZnSe films have been grown on CIS substrates at 200°C to 250°C. ZnO is also being deposited by MOCVD by reacting tetrahydrofuran (THF) with a zinc adduct. ZnSe/CIS heterojunctions have been studied by growing n-ZnSe films onto 2 cm x 2 cm CIS substrates diced from materials supplied by Siemens and then depositing an array of aluminum circular areas 2.8 mm in diameter on top of the ZnSe to serve as contacts. Al films are deposited with a thickness of 80 to 100 Å so that light can pass through the film, thus allowing the illuminated characteristics of the ZnSe/CIS junction to be tested. Accounting for the 20 to 25 % transmittance through the Al film into the ZnSe/CIS structure, current devices have estimated, active-area AM1.5 efficiencies of 14 %. Open circuit voltages > 500 mV are often attained.

Based on characteristics of small area ZnSe/CIS and ZnO/ZnSe/CIS diagnostic devices, it appears that efficient ZnSe/CIS solar cells can be fabricated. Future studies will involve continued investigations of ZnSe/CIS heterojunctions, development of procedures for MOCVD growth of ZnO, and cell fabrication.

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16. Abstract (Limit: 200 words)

This report describes investigations of ZnSe/copper indium diselenide (CIS) solar cells being carried out to improve the efficiencies of CIS cells and to determine if ZnSe is a viable alternative to CdS as a window material. Metal-organic chemical vapor deposition (MOCVD) growth of ZnSe is accomplished in a Spire 500XT reactor by reacting a zinc adduct with H_2Se. Conductive n-type ZnSe is grown by using iodine as a dopant. Ethyliodide was mixed with helium and installed on one of the gas lines to the system. ZnSe films were grown on CIS substrates at 200°–250°C. ZnO was also deposited by MOCVD by reacting tetrahydrofuran (THF) with a zinc adduct. ZnSe/CIS heterojunctions were studied by growing n-ZnSe films onto 2-cm x 2-cm CIS substrates diced from materials supplied by Siemens, and then depositing an array of aluminum circular areas 2.8 mm in diameter on top of the ZnSe to serve as contacts. Al films were deposited with a thickness of 80–100Å so that light could pass through the film, thus allowing the illuminated characteristics of the ZnSe/CIS junction to be tested. Accounting for the 20%–25% transmittance through the Al film into the ZnSe/CIS structure, current devices have estimated, active-area air mass (AM)1.5 efficiencies of 14%. Open circuit voltages above 500 mW are often attained.

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