

Impact of Na on MoSe₂ formation at the CIGSe/Mo interface in thin-film solar cells on polyimide foil at low process temperatures

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Abstract

The aim of this work is to study the effect of Na on the formation of MoSe₂ at the absorber/Mo back contact interface of Cu(In,Ga)Se₂ (CIGSe) thin-film solar cells at low process temperatures using polyimide foil as substrate material. As reported previously, the presence of Na has been observed to modify the formation of the back interface, which may in part explain the different electronic properties of the completed device, as was determined by admittance spectroscopy and *I*–*V*–*T* measurements. In order to further study this interface formation, break-off experiments are performed and a lift-off technique is used to enable investigation of the different surfaces via X-ray photoelectron spectroscopy and Raman scattering. Both techniques confirm the dependence of the MoSe₂ layer formation at the back interface on the presence of Na. The experiments also reveal the relevance of the composition of the absorber layer to the development of the MoSe₂ layer during the Cu(In,Ga)Se₂ deposition process. Hence this work describes routines that may be employed to develop an “appropriate” CIGSe/Mo back interface for high-efficiency solar cells.

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1. Introduction

Cu(In,Ga)Se₂ (CIGSe) is one of the most promising materials for thin-film photovoltaic solar cell applications. Recently, an efficiency of 20.4% has been achieved in the laboratory using polyimide (PI) foil as substrate material with a low-temperature approach [1,2]. Lightweight and flexible CIGSe thin-film solar cell modules not only offer the opportunity to increase the range of terrestrial applications, but they are also attractive for space applications. In

contrast to metal foils, PI foils make monolithic module integration a viable option for solar cell interconnection. PI can only tolerate process temperatures *T* of up to ~420 °C, which is well below 600 °C, the temperature widely used for CIGSe absorber preparation on glass and metal foils. The fact that the best reported efficiency on glass is currently 20.3% [3] demonstrates that, compared to high-temperature approaches, low-temperature routines for CIGSe fabrication offer a comparable potential regarding deposition of the highest quality absorbers. Low-*T* processes are not only required for the use of PI as substrate material, but are also beneficial for the preparation of CIGSe devices on transparent contacts, on steel foils or for the projected preparation of possible future tandem devices. It is therefore necessary to identify and understand

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the effect of different deposition conditions on the absorber layer and interface formation at low process temperatures.

CIGSe solar cells for high-efficiency devices require the incorporation of a small amount of Na into the polycrystalline absorber layer to enhance its electrical properties [4–6]. The main effect of Na is, among others, the improvement in p-type conductivity due to an increase in the effective hole density and, as a consequence, an enhanced open circuit voltage (V_{oc}) and fill factor (FF). Güttler et al. [7] have reported the influence of Na incorporation at different stages of the CIGSe deposition process on the electronic and morphological properties of the final device. In addition, Na has been reported to have an impact on the formation of the CIGSe/Mo back interface, which may be of importance for the performance of the completed solar cell [8,9]. A barrier at the back interface, which may develop depending on a range of modifications of MoSe_2 formation due to the presence or absence of Na, has been associated with the rollover behaviour that is observed for particular CIGSe thin-film devices. Assuming an electronic barrier of 0.2 eV, Fig. 1 shows the effect of a varied charge carrier density N_A on the I – V characteristic of a complete $\text{ZnO}/\text{CdS}/\text{CIGSe}/\text{Mo}$ thin-film solar cell, using the software AfORShet, a 1-D device simulation tool [10]. A complete list of input parameters can be found in Ref. [11]. The charge carrier density is chosen to be located in an appropriate range, as previously observed [12]. The so-called rollover effect, a sharp kink in the course of the current density that is otherwise seen to steeply increase with the applied forward voltage, is observed for a wide range of charge carrier densities, when a barrier is placed at the back interface of the device. For high and low charge carrier densities,

however, the rollover behaviour disappears. While for a high value of N_A the number of charge carriers is simply too high for the back barrier to take effect, for very low values of N_A the front and back grading within the device overlap and the back barrier effectively disappears. A variation in height of the back barrier will shift the corresponding ranges for N_A , but the device behaviour will stay the same. This shows that, on the one hand, a barrier at the back interface can be made responsible for a rollover behaviour and that, on the other hand, the effect of Na in CIGSe can also act to impede it.

The formation of a MoSe_2 layer on the surface of Mo thin films in a Se-containing atmosphere [13–15] and at the back interface of CIGSe thin-film solar cells has also been studied in the literature for its impact on the device characteristics [16–18], but is still under discussion. One further aspect of the presence and formation dynamics of MoSe_2 at the CIGSe/Mo interface is the fact that it may also have significant influence regarding the adhesion of the CIGSe thin film to the underlying back contact. The latter is a critical aspect when working on PI substrates.

As shown in Fig. 1, and reported in Ref. [9], the modification of the back interface by the presence of Na with the formation of a MoSe_2 layer may explain in part the different electronic behaviour of the final devices. The objective of this work is to investigate the presence and formation of MoSe_2 at the back interface of the device, and how this depends on the presence/absence of Na and on the film composition during three-stage-based CIGSe thin-film growth. Two questions arise here. When is the MoSe_2 layer formed and how does this depend on the presence of Na? Here, we will try to answer those questions and describe some routines to develop a suitable CIGSe/Mo interface for high-performance devices. In the present work, CIGSe thin-film solar cells are deposited on PI foils. The use of PI as substrate material allows for an accurate investigation of the influence of Na, which is introduced here via evaporation of a NaF precursor layer before the absorber is deposited. The effect of Na on the chemical properties and the identification of the material phases present at the CIGSe/Mo interface are investigated by X-ray photoelectron spectroscopy (XPS) and Raman spectroscopy.

2. Experimental details

CIGSe absorber layers are grown on Mo-coated PI foils using a multistage co-evaporation process [19] based on the three-stage process [20]. The Mo layer was grown by DC sputtering under the same conditions for all the samples investigated. The nominal substrate temperature was kept at 330 °C during the first stage of the deposition process; during the second and third stages, this was increased up to 420 °C nominal maximum measured by a reference thermocouple. Break-off experiments at different points of the multistage deposition process were carried out to understand the formation of the MoSe_2 phase at the back side

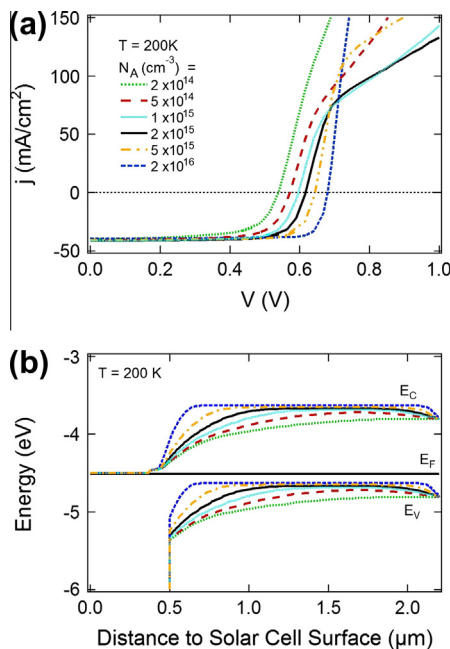


Fig. 1. Numerical simulation of the effect of a varied charge carrier density on the behaviour of a completed CIGSe thin-film solar cell: (a) I – V characteristic; (b) progression of E_C and E_V within the CIGSe thin film.

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