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Dielectric and diffusion barrier multilayer for Cu(In,Ga)Se_2 solar cells integration on stainless steel sheet

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ABSTRACT

For the fabrication of monolithically integrated flexible Cu(In, Ga)Se_2, CIGS modules on stainless steel, individual photovoltaic cells must be insulated from metal substrates by a barrier layer that can sustain high thermal treatments. In this work, a combination of sol–gel (organosilane-sol) and sputtered SiAlxOy forming thin diffusion barrier layers (TDBL) was prepared on stainless steel substrates. The deposition of organosilane-sol dielectric layers on the commercial stainless steel (maximal roughness, Rz = 500 nm and Root Mean Square roughness, RMS = 56 nm) induces a planarization of the surface (RMS = 16.4 nm, Rz = 176 nm). The DC leakage current through the dielectric layers was measured for the metal-insulator-metal (MIM) junctions that act as capacitors. This method allowed us to assess the quality of our TDBL insulating layer and its lateral uniformity. Indeed, evaluating a ratio of the number of valid MIM capacitors to the number of tested MIM capacitors, a yield of ~ 95% and 50% has been reached respectively with non-annealed and annealed samples based on sol–gel double layers. A yield of 100% was achieved for sol–gel double layers reinforced with a sputtered SiAlxOy coating and a third sol–gel monolayer. Since this yield is obtained on several samples, it can be extrapolated to any substrate size. Furthermore, according to Glow Discharge Optical Emission Spectroscopy and Time of Flight Secondary Ion Mass Spectroscopy measurements, these barrier layers exhibit excellent barrier properties against the diffusion of undesired atoms which could otherwise spoil the electronic and optical properties of CIGS photovoltaic cells.

1. Introduction

In recent years, the fabrication of efficient low-cost photovoltaic modules has become a major technological and economical challenge demanding the use of appropriate materials and manufacturing.

Organic photovoltaic technologies are in principle very cheap but their efficiency and lifetime are still insufficient. Thin inorganic layers such as CIGS (Cu(In, Ga)Se_2)absorbing layers can be economically attractive compared to silicon. This semiconductor allows to reach a high efficiency [1,2]. As an example, a conversion efficiency of 20.3% was reached with CIGS on soda lime glass substrate [1].

Stainless steel (SS) is one of the most suitable substrates with respect to the cost, flexibility and high temperature (>500 °C) required for CIGS growth [3]. SS substrates are also lightweight and compatible with a roll to roll process [4].

A major challenge for the fabrication of monolithic integration of CIGS cells on large flexible sheets is to prepare an appropriate insulated substrate using a dielectric layers. The dielectric layer called diffusion barrier layers (DBL) has to play two main roles: (1) inhibiting the diffusion of undesired atoms from the SS substrate to the absorber layer since some transition metal elements dramatically spoil the performances of CIGS absorbing layers [5] and (2) ensuring a good electrical insulation between the metal substrate and the monolithically interconnected photovoltaic cells [6].

In this study, the influence of thermal treatments on the surface morphology and leakage current of thin diffusion barrier layers (TDBL) on Cr-steel are reported. As this work deals with a dielectric multilayer for the monolithic integration of cells interconnected on the large area where a high voltage can be produced according to the size and the conversion efficiency of the modules, the voltage and time to breakdown are key parameters to determine. Metal-Insulating-Metal (MIM) capacitors were obtained by sputtering molybdenum pads of 10 mm × 10 mm and 50 mm × 50 mm onto a 100 mm × 100 mm stainless steel substrate covered with TDBL (Fig. 1). Finally, the ability of the TDBL to act against the diffusion of substrate elements is also investigated.

2. Experimental details

2.1. Preparation of substrate and thin barrier layer

All investigations reported in this work were performed on 0.5 mm-thick AISI-316-2RB stainless steel bare substrates. No polishing was applied to the substrate. Fig. 2 is a schematic representation of the

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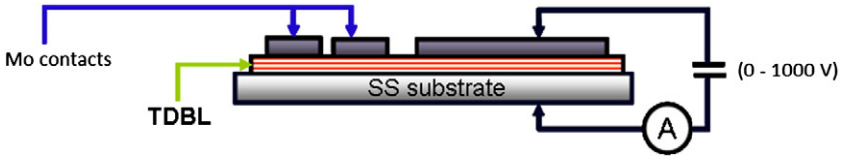

Fig. 1. Schematic diagram of a matrix of Mo/TDBL/Metal (MIM) capacitors used for measuring the DC leakage current.

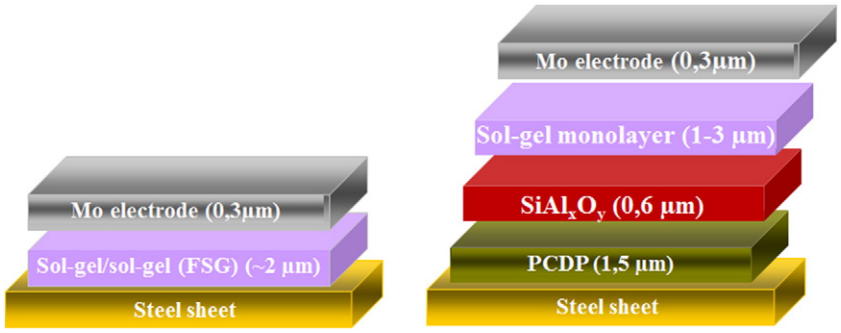

Fig. 2. Schematic of advanced substrate for CIGS application. a) Sol gel double layers, b) a combination of sol gel coatings with sputtered multilayer.

multilayer structure that could be used as a substrate for monolithic integration of CIGS photovoltaic cells. Two solutions are considered:

- Sol–gel bilayers based on organosilane-sol. The thickness of films is in the range of 1.5 to 2.5 µm;
- Sol–gel bilayer reinforced with a 600 nm SiO_xAl_y film deposited by magnetron sputtering.

Preparation of the surface is a crucial step to remove surface contamination, to ensure coverage of the whole surface and to enhance the adhesion of organosilane-sol-based layer on the SS substrate. Prior to the fabrication of the system described in Fig. 2, the surface of SS substrate was cleaned in successive ultrasonic baths with appropriate degreasing solvents followed by a drying with nitrogen.

Afterwards, SS substrates were bar coated with a hydrolyzed organosilane-sol layer. In our study, the bar coating process consist of depositing the wet hybrid-polymer by manually moving the coating bar on the surface of SS substrates. Rapid cross-linkage of organosilane-sol coatings is then obtained in a convection oven at 270 °C.

The second layer was deposited using the same procedure. Finally, samples were annealed at 550 °C with a conventional industrial gas mixture named HN_x (a 5% H2: 95% N2 gas mixture) in order to form a hard coating called PCDP (preceramic derived polymer).

SiO_xAl_y films were deposited using reactive DC magnetron sputtering. The target-to-substrate distance was fixed at 17 cm. The magnetron sputter source was supplied by a DC power of 300 W connected with a pulse generator with a frequency of 50 Hz.

Before Mo films deposition, the base pressure was 3.6×10^{-5} Pa. During deposition, the substrate holder was rotated around the central axis of the chamber at 20 rpm. Mixtures of argon and oxygen were used as sputtering gases; flows of O_2 and Ar were respectively 40 sccm and 72 sccm while the total pressure was kept constant at 0.86 Pa. 400 nm thick Molybdenum contacts were sputtered from a 3 in. Mo target onto all TDBL using the same sputtering system.

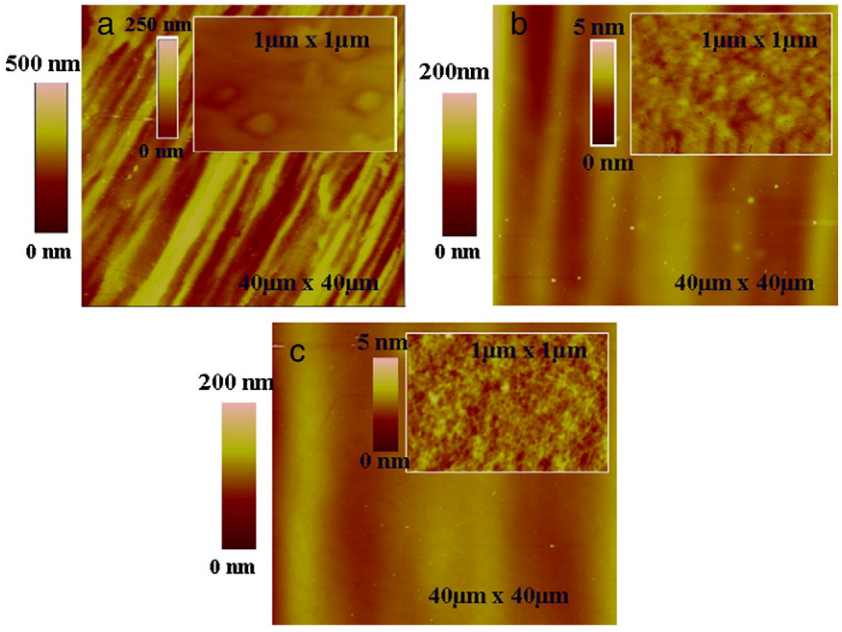

Fig. 3. AFM images of (a) uncoated SS surface, (b) sol–gel/sol–gel double layers and (c) sol–gel/sol–gel double layers annealed at 550 °C in HN_x.