

Nanocomposite of TiO₂ and CuInS₂: towards solid-state 3D solar cells

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The major drawback of the present generation of photovoltaic solar cells is their laborious, energy consuming, and costly production. Therefore, a completely new approach is desired. With the advent of dye-sensitized (Grätzel-type) solar cells, C60/polymer, and hybrid nanocrystal / polymer bulk heterojunctions, challenging alternatives are offered, although serious complications are still to be eliminated. A major concern in these alternatives is their poor stability if operating in full sunlight. The cells must be sealed rigorously to avoid reaction with oxygen and water. All-solid, completely inorganic, bulk heterojunctions do not require expensive sealing but have been pursued with limited success so far. Here, we report a new approach towards what is referred to as the 3D-solar cell concept (Fig.1). In this type of nanocomposite, a wide bandgap n-type semiconducting oxide and a p-type visible light sensitive semiconductor are mixed on a nanometer scale. This solar cell concept may open a new horizon in the manufacture of future photovoltaic devices. We employed Atomic-Layer Chemical Vapour Deposition (AL-CVD) for infiltration of CuInS₂ inside the pores of nanostructured TiO₂. First, a dense film of TiO₂ (~100 nm) is applied onto TCO-glass (SnO₂:F). On top of this, a 2 μm thick nanoporous TiO₂ film is applied with primary particles between 10 and 50 nm in diameter. Next, these substrates are infiltrated with CuInS₂ using ALD. The process conditions are: 2 mbar reactor pressure, temperature between 350 and 500 °C, and CuCl, InCl₃, and H₂S as precursors.

The chemical composition of the cells is investigated with X-ray diffraction, Raman spectroscopy, and photoluminescence spectroscopy. CuInS₂ films have the chalcopyrite crystal structure with a minor fraction of the Cu-Au ordered structure. The fraction of the Cu-Au phase can be determined with Raman spectroscopy and is about 5% in volume. Cu-Au is associated with the presence of anti-site point defects. Photoluminescence has been used to determine the energy levels associated to these anti-site defects, being 0.07 eV for In_{Cu}^{\bullet} and 0.15 eV for $Cu_{In}^{\prime\prime}$. The efficiency of CuInS₂ (CIS) solar cells is determined by the presence of anti-site defects because they introduce energy levels in the band gap, which determine the conductivity type and the minority carrier lifetime.

The electrical properties of nanostructured TiO₂/CuInS₂ heterojunctions are investigated by studying Current-Voltage (IV), Time of Flight (TOF), Impedance Spectroscopy (IS), and Deep-Level Transient Spectroscopy (DLTS) as a function of temperature. Preliminary results give us strong indications that these cells behave different in dark than dye-sensitized solar cells (Grätzel-type). The IV recorded in the dark shows space-charge limited current behaviour with an activation energy for the hole mobility of 150 meV, which is associated to holes trapped on the anti-site defect $Cu_{In}^{\prime\prime}$. The energy levels of electron and hole traps derived with IV, IS and DLTS techniques agree with those found in our previous photoluminescence study. TOF results indicate that drift rather than diffusion governs the charge transport.

Since alignment of the conduction bands is required an In_2S_3 buffer layer must be applied. The solar cells show photovoltaic activity between 360 and 900 nm with a maximum monochromatic incident photon-to current conversion efficiency of 80%. If AM1.5 irradiation is applied, the open-circuit voltage is 0.49 V, the short circuit current 18 mA cm^{-2} , and the fill factor 0.44, which add up to an overall energy conversion efficiency of 4% (Fig.2). This is twice the performance of the best inorganic 3D solar cell reported so far.

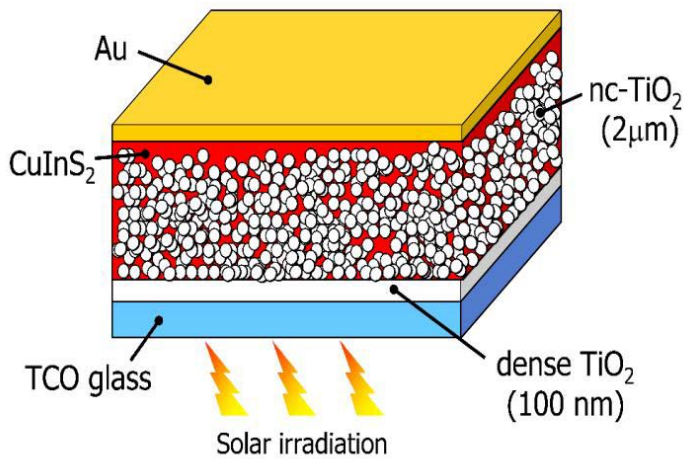


Fig. 1 Structure of an inorganic 3D solar cell.

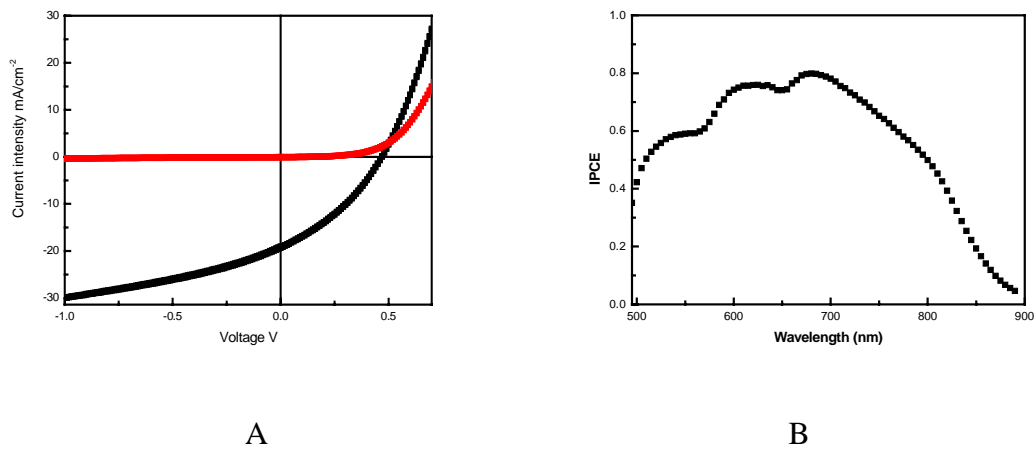


Fig 2. Current-voltage response of a 3D solar cell comprising a TiO_2 and CuInS_2 nanocomposite. Black line: current in darkness; red line: current with AM 1.5 irradiation (A). The incident-photon to current conversion efficiency of this cell (B).