Pulsed laser deposition of Cu_2ZnSnS_4 absorber layers assisted by a reactive sulfur beam for solar cells

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Thin film solar cells based on the p-type semiconductor Cu_2ZnSnS_4 (CZTS) have emerged as an alternative to CdTe and $CuGaInSe_2$ (CIGS), the two leading commercial thin film solar absorber materials, due to the low-cost, earth-abundance and non-toxicity together with the high absorption coefficient and an optimal bandgap of 1.45 eV.

By using pulsed laser deposition (PLD), a technique suitable for depositing high-quality films with complex stoichiometry, a metallic target of Cu_2ZnSn was ablated in vacuum onto a substrate. A reactive sulfur beam, which enhances the chemical reactivity of sulfur, is directed towards the substrate in order to reduce the loss of the volatile element S in the deposited film. By this approach, the subsequent annealing in toxic gases (e.g. H_2S) is avoided. All films are deposited onto Mo coated soda-lime-glasses in the temperature range from 150 ^{o}C to 550 ^{o}C. The excimer laser used is working at 248 nm with a pulse length of 20 ns. The pulse repetition rate is set at 15 Hz and the deposition process is lasting 80 min. For the characterization of the films, Raman spectroscopy, scanning electron microscopy (SEM) and X-ray spectroscopy (EDX) are used.

The Raman spectra show an increase of the intensity and a sharpness of main peak associated to kesterite CZTS up to a substrate temperature of 450 ^{o}C. Above the deposition temperature of 450 ^{o}C the film decomposes into multiple secondary phases. EDX analyses depict the optimal composition for solar cells in all the samples below 450 ^{o}C and SEM demonstrates the dependence of the grain size on temperature.