

Super-cycle atomic layer deposition of indium gallium zinc oxide

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Indium gallium zinc oxide (IGZO) is a promising candidate as a transparent conductive oxide (TCO). The amorphous phase of this compound shows a band gap of 3.0 eV and exhibits a high charge carrier mobility and concentration with reported values of 50-80 cm²/Vs and 10¹⁷-10²⁰ cm⁻³, respectively. ^[1,2] Therefore, it is well suited for photovoltaic applications, light emitting diodes, and thin film transistors. In contrast to single crystalline TCO, it is much easier and cheaper to realize IGZO thin films. To date, the established deposition methods for IGZO layers such as DC/magnetron sputtering and pulsed laser deposition fail when the deposition of uniform films over a large substrate area is required. Also, controlling the elemental composition of the quaternary system is pivotal for achieving the desired electrical properties. Atomic layer deposition (ALD) can meet both challenges by combining the ALD cycles of their respective binary compounds in a distinct sequence and ratio. This so-called super-cycle process allows controlling the composition of the target film by adjusting the individual cycle ratio.

In this work, we present a super-cycle approach to deposit IGZO films on a 4-inch wafer. The depositions have been carried out in a SENTECH plasma-enhanced ALD (PEALD) reactor, in which we applied a thermal process for zinc oxide (ZnO) as well as plasma-enhanced processes for gallium and indium oxide (Ga₂O₃, In₂O₃). The growth mechanism of each individual process within the super-cycle has been investigated and monitored by *in-situ* ellipsometry (i-SE, SENTECH ALD Real-Time-Monitor). An *ex-situ* chemical analysis of the

compound has been performed by X-ray photoelectron spectroscopy (XPS) to determine the film composition and correlate it to the expected values based on the super-cycle sequence.

The i-SE revealed a nucleation delay for the thermal ZnO process when the layer is grown on either a Ga₂O₃ or an In₂O₃ surface, making it challenging to properly adjust the cycle ratio. Thus, the thermal ZnO cycle has been replaced by a plasma-enhanced ZnO process, which shows no nucleation delay. In further *in-situ* investigations a higher growth rate for the PEALD-ZnO deposition on a Ga₂O₃ surface than on an In₂O₃ surface was observed. Based on this knowledge, we were able to set-up the ideal cycle sequence for the IGZO process. Accordingly, XPS revealed a direct correlation between the applied cycle ratio and the elemental composition of the film. Together, these findings confirm that a full PEALD super-cycle process is an effective approach to deposit well-defined IGZO films.

[1] P. Barquinha, L. Pereira, G. Gonçalves, R. Martins, E. Fortunato, J. Electrochem. Soc. 156 (2009) H161

[2] H. Hosono, J. Non-Cryst. Solids 352 (2006) 851.