

SPATIAL-ALD OF TRANSPARENT AND CONDUCTIVE OXIDES

A. Illiberi^{1*}, T. Grehl², A. Sharma¹, B. Cobb¹, G. Gelinck¹, P. Poodt¹, H. Brongersma^{3,4}, F. Roozeboom^{1,4}¹ Holst Centre/ TNO 5600 HE Eindhoven, The Netherlands,² ION-TOF GmbH Heisenbergstrasse 15, 48149, Muenster, Germany³ Department of Materials, Imperial College, SW7 2AZ London, United Kingdom⁴ Department of Applied Physics, Eindhoven University of Technology, 5600 MB Eindhoven, The Netherlands

*email: andrea.illiberi@tno.nl

ABSTRACT

The rapid growth of the electronics and solar industry has guided the investigation of Zn-based transparent and conductive materials. The industrial needs for low-cost deposition processes with high throughput has driven the development of atmospheric pressure Spatial-ALD, which combines the advantages of conventional ALD with high growth rates (up to nm/s). We have used the Spatial-ALD technique to grow polycrystalline i-ZnO, Al:ZnO, In:ZnO and amorphous InZnO, InGaZnO.

ZnO films have been deposited by sequentially exposing a substrate to water and the metal precursors (i.e. diethylzinc together with trimethylaluminum or trimethylindium and/or triethylgallium) vapor, spatially separated in the spatial ALD injector, so that a purge step is no longer needed. The electrical properties of transparent (85 % in Vis) i-ZnO films have been controlled, ranging from *n*-type conductive ($n = 7 \cdot 10^{19} \text{ cm}^{-3}$ and $\mu = 30 \text{ cm}^2/\text{V}\cdot\text{s}$) to insulating, by varying the DEZ partial pressure and the deposition temperature (150 - 250 °C). The carrier density increases with Al or In content, ranging from $7 \cdot 10^{18} \text{ cm}^{-3}$ (i-ZnO) to a maximum value of $5 \cdot 10^{20} \text{ cm}^{-3}$ (Al/Zn ≈ 0.09) and $6 \cdot 10^{20} \text{ cm}^{-3}$ (In/Zn ≈ 0.02), as shown in Fig. 1. A transition from polycrystalline In:ZnO to transparent (90% in the Vis) and conductive (4 mOhm·cm, 180 nm thickness) amorphous InZnO and InGaZnO occurs with increasing In and Ga content (up to In/Zn ≈ 0.3 and Ga/Zn < 0.06), as revealed by XRD analysis (inset in Fig. 1).

A nucleation phase of about 300 ALD-cycles is found for InGaZnO films by measuring the film thickness with Spectroscopic Ellipsometry, before the onset of the bulk growth at a rate of about 0.03 nm/cycle. The early stages of the nucleation phase (from 5 to 100 ALD-cycles) have been investigated by Low Energy Ion Scattering (LEIS), a non-destructive surface analysis technique. The surface coverage of the Si-substrate by the different metal elements (Zn, In and Ga) is resolved and the composition of the nucleating film is measured by the LEIS technique (Fig. 2). An initial In-rich phase (Zn/In ~ 0.06) is found after 5 cycles, followed by a film closure (no Si detected) at 100 cycles with Zn/In ~ 1.3 . The amorphous structure of the film bulk is confirmed by XRD-diffraction. The electrical properties of the bulk can be controlled by varying its metal composition: carrier density and mobility range from $7 \cdot 10^{18}$ to $6 \cdot 10^{20} \text{ cm}^{-3}$ and from 1 to $20 \text{ cm}^2/\text{Vs}$, respectively, when varying the Ga/Zn from 0 to 0.06 and In/Zn from 0 to 0.32, as measured by EDX analysis. The use of Spatial-ALD GIZO films as an active channel in Thin Film Transistors has been tested after a post-deposition annealing, resulting in a device mobility of $3.5 \text{ cm}^2/\text{Vs}$ and on-off ratio of 10^4 .

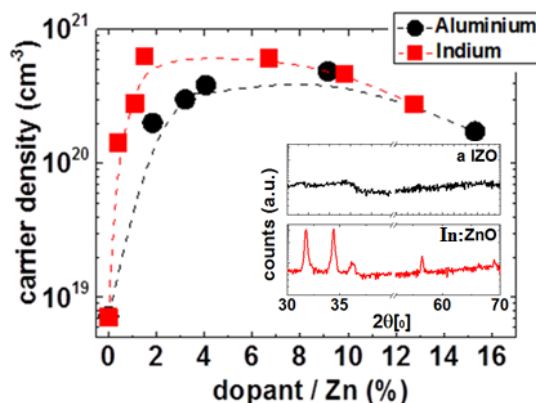


Fig.1 Carrier density of In:ZnO and Al:ZnO versus In/Zn and Al/Zn content. Inset: XRD spectra of polycrystalline In:ZnO and amorphous InZnO

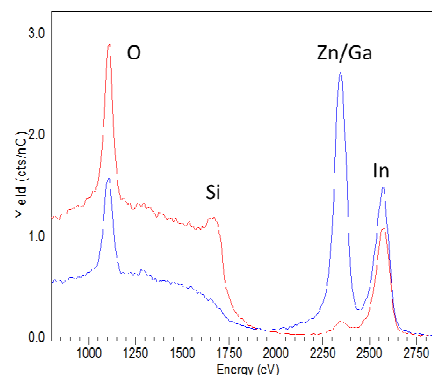


Fig. 2 Elemental composition of S-ALD GIZO films after 5 (red line) and 100 (blue line) cycles.

