



Contribution ID : 144

Type : Poster

Optoelectronic Properties of Al and Cr Co-Doped ZnO Bilayer Systems for Chemiresistive Gas Sensing of Volatile Organic Compounds

Metal-oxide semiconductors with controlled defect chemistry are essential for high-performance chemiresistive sensors targeting volatile biomarkers in exhaled breath for non-invasive diabetes monitoring. This work presents a bilayer chemiresistive sensor based on Cr/Al co-doped ZnO (AZO:Cr), featuring a porous, lightly doped top layer (0.1 at.% Cr) deposited by sol-gel spin-coating on a denser moderately doped bottom layer (1–4 at.% Cr) grown by RF magnetron sputtering. Dopant concentrations and post-annealing at 500 °C (Ar or air atmosphere) engineer oxygen vacancies, zinc interstitials, and Cr³⁺ d-state levels, while Cr passivation of Zn vacancies enhances lattice thermal stability and enables carrier density modulation governed by Cr/Al ratio.

Structural analysis (XRD, SEM-EDS) confirmed a highly c-oriented wurtzite phase in the bottom layer and a hierarchical porous network of 15–30 nm nanocrystallites in the top layer, promoting gas diffusion and active site formation. Spectroscopic techniques (Raman, cathodoluminescence, and variable-angle ellipsometry) revealed the evolution of defects, sub-bandgap transitions, the Burstein-Moss shift, and modulation of the dielectric function induced by Cr/Al incorporation. Gas sensing tests at 160–220°C showed excellent response to 0.1% V/V acetone (sensitivity >50%) via ionosorption and Eley-Rideal kinetics. These findings established structure-property correlations for stable and transparent (>80% transmittance) TCO-based sensor devices.

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Session Classification : Poster Fisica del Estado Solido