Electronic Supplementary Material (ESI) for Dalton Transactions.

Solution synthesis and dielectric properties of alumina thin films: Understanding the role of the organic additive in film formation.

Rudolf C. Hoffmann[1], Maciej O. Liedke[2], Maik Butterling[2], Andreas Wagner[2], Vanessa Trouillet[3], and Jörg J. Schneider[1]*

[1]Eduard-Zintl-Institut für Anorganische und Physikalische Chemie, Technische Universität Darmstadt, Alarich-Weiss-Straße 12, 64287 Darmstadt, Germany

[2]Institute of Radiation Physics, Helmholtz-Zentrum Dresden-Rossendorf, Bautzner Landstraße 400, 01328 Dresden, Germany

[3] Institute for Applied Materials (IAM-ESS) and Karlsruhe Nano Micro Facility (KNMF), Karlsruhe Institute of Technology (KIT), Hermann-von-Helmholtz-Platz 1, 76344 Eggenstein-Leopoldshafen, Germany

$$\Phi_{\mathrm{M}} = \frac{\Sigma\, \mathrm{n(MCZ)}}{\Sigma\, \mathrm{n(ALN)}} = \frac{\Sigma\, \mathrm{n(C_2H_6N_2O_2)}}{\Sigma\, \mathrm{n(AlN_3O_9)}} = \frac{10}{15} = 0.67$$

Scheme 1: Calculation of Φ_M using the valencies for the different elements according to *Jain et al.*, i.e. Al (-3), C (-4), H (-1), N (±0), O (+2).

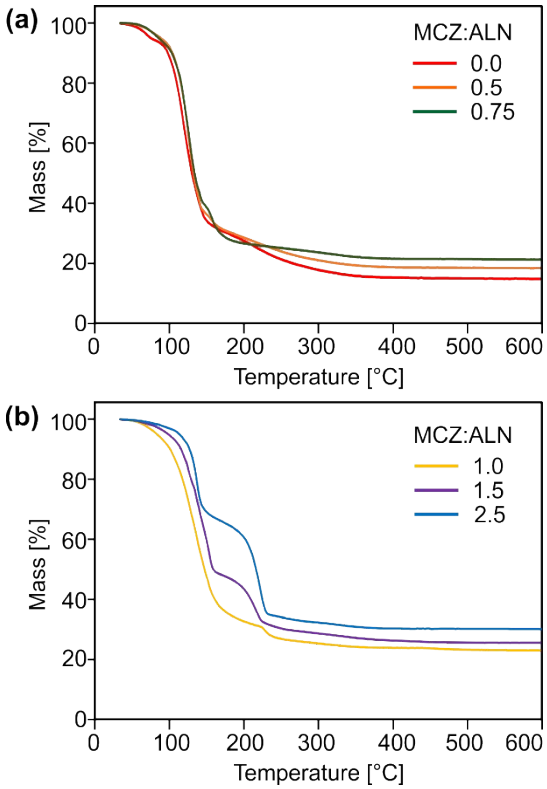

Figure 1: Thermogravimetric mass loss curves of precursor mixtures containing various MCZ:ALN ratios.

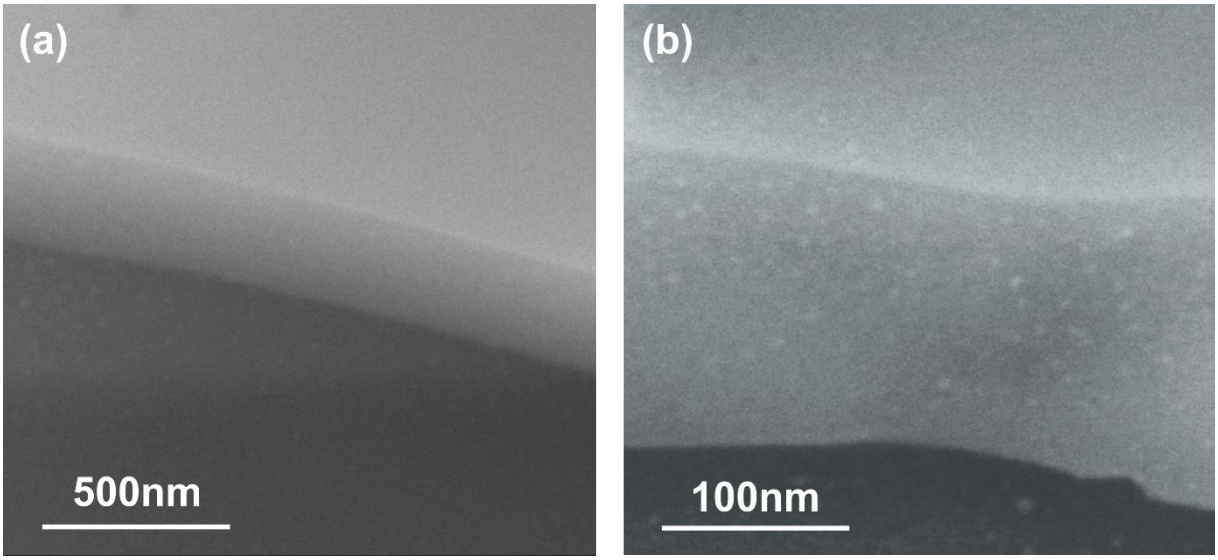

Figure 2: SEM micrographs depicting a cross section of an alumina film on a silicon substrate obtained from spincoating of the precursor mixture (ALN 0.4 M, MCZ:ALN = 2.5) and annealing at 450 °C. (a) and (b) present different magnifications.

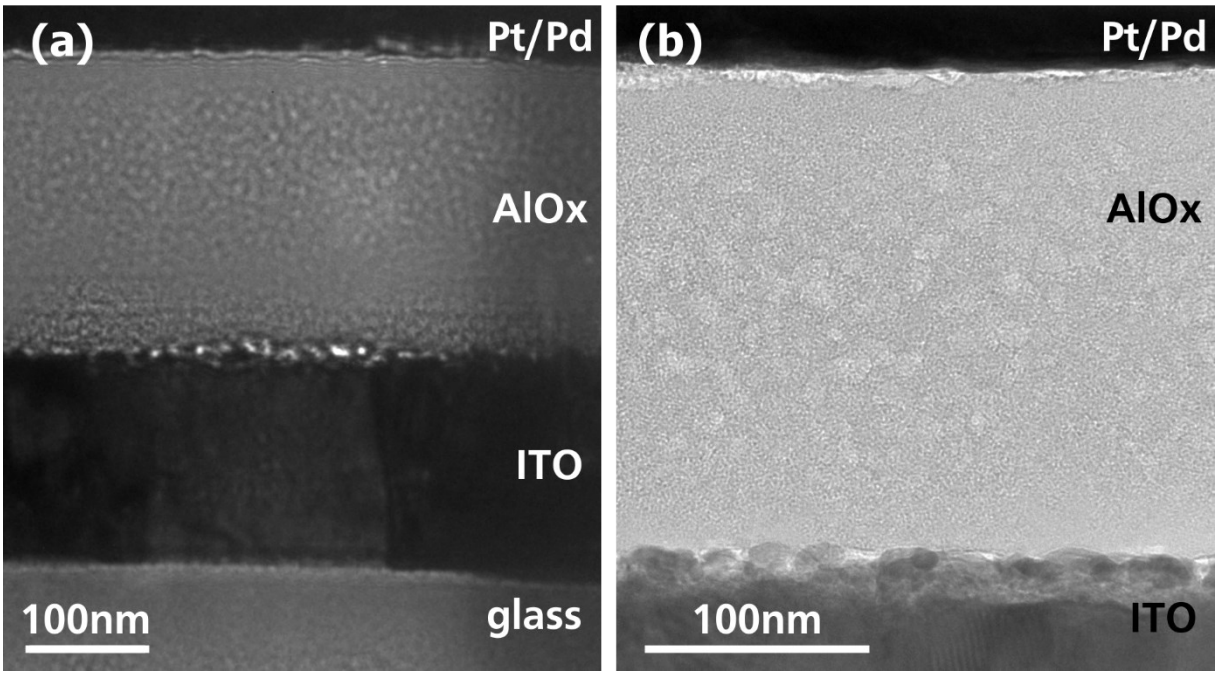

Figure 3: HRTEM images depicting a cross section of an alumina film on a ITO/glass substrate obtained from spincoating of the precursor mixture (ALN 0.4 M, MCZ:ALN = 2.5) and annealing at 450 °C. The roughness at the interfaces is probably an artifact from FIB preparation and due to delamination. (a) and (b) present different magnifications.

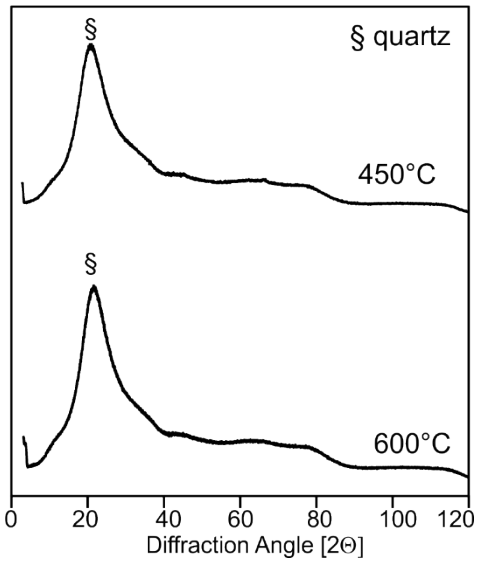

Figure 4: X-Ray diffractograms obtained from spincoating of the precursor mixture (ALN 0.4 M, MCZ:ALN = 2.5) on quartz and annealing at various temperatures.

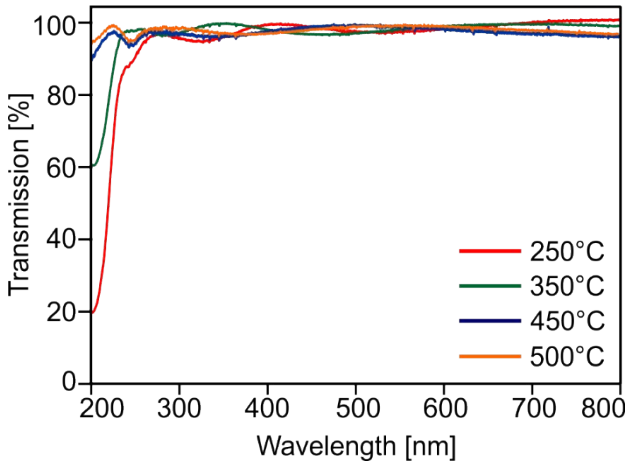

Figure 5: UV/VIS spectra obtained from spincoating of the precursor mixture (ALN 0.4 M, MCZ:ALN = 2.5) on quartz and annealing at various temperatures.

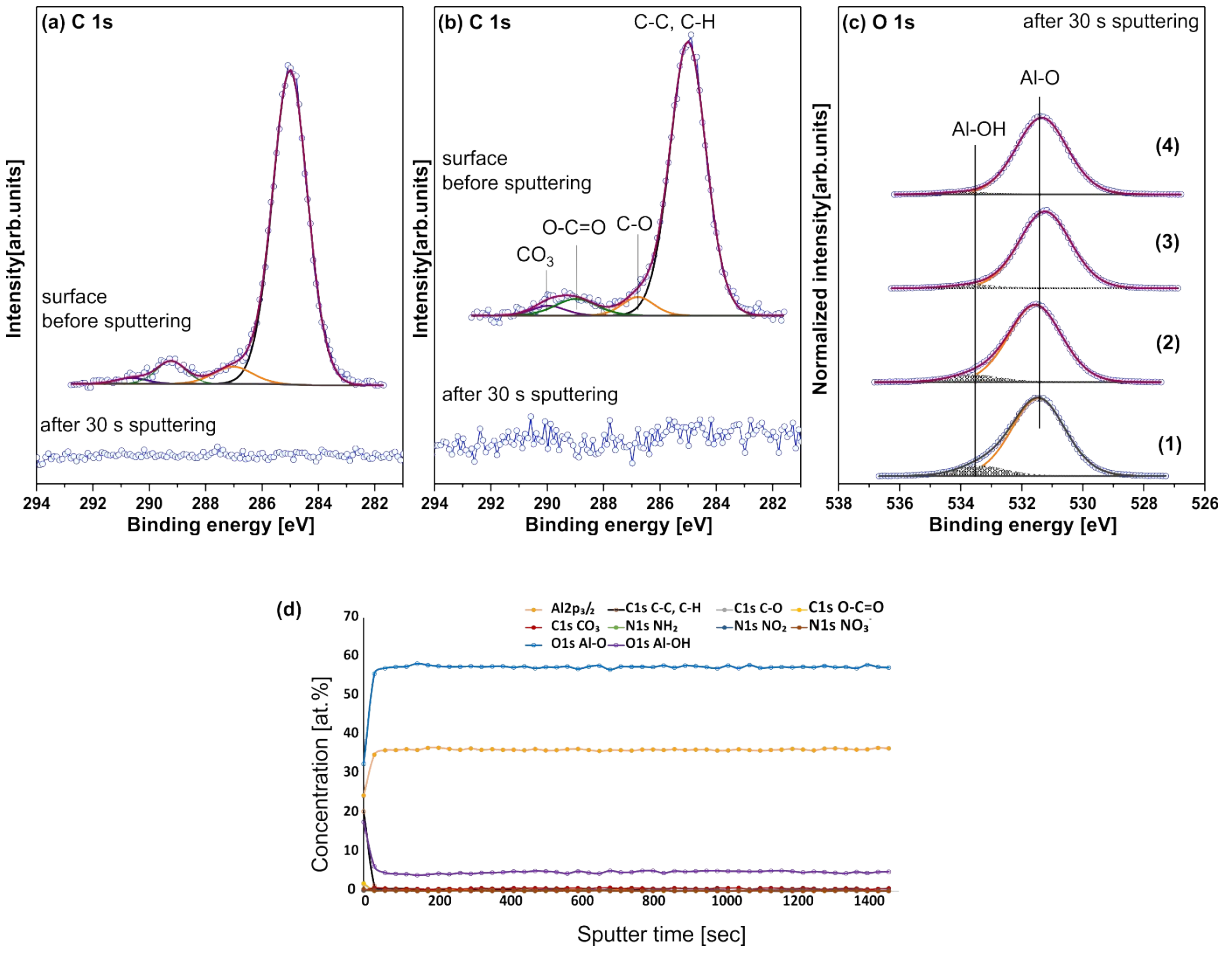

Figure 6: XPS spectra depicting (a) C1s region of (**3**), (b) C1s region of (**4**), (c) comparison of O1s signals of samples (**1**)-(**4**) as well as (d) depth profile of (**1**) showing the behaviour of relevant contributions of various elements (Al, O, C and N).

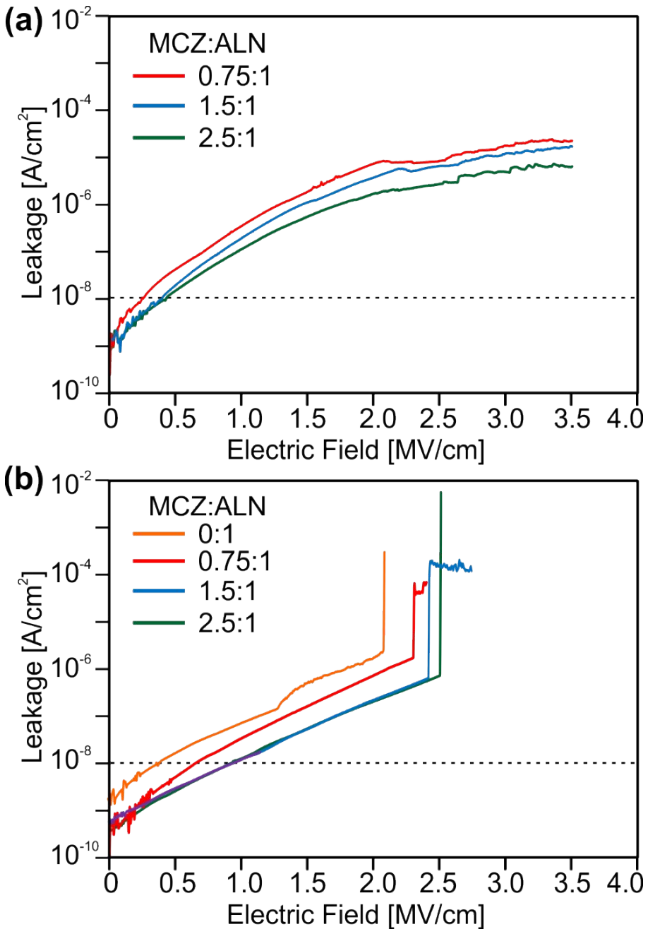

Figure 7: Typical breakthrough curves of films from precursor mixtures (ALN 0.4 M) with various MCZ:ALN ratios. Coatings obtained from spincoating of (a) 3 layers (~110 nm) and (b) 4 layer (~145 nm).

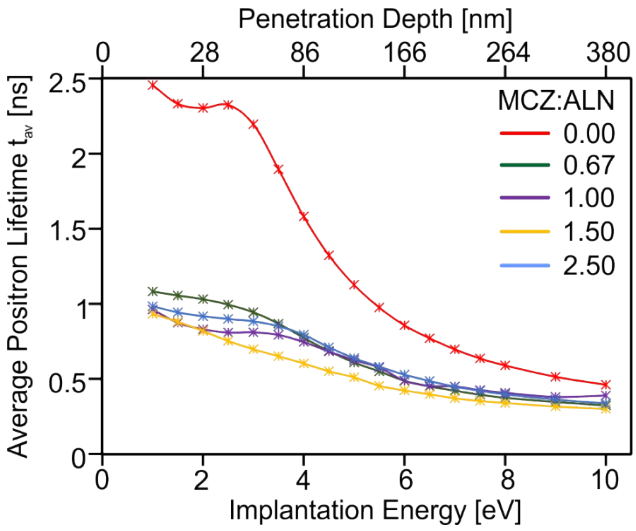

Figure 8: Positron lifetimes as a function of implantation energy of alumina films on silicon substrates obtained from precursor mixtures with various MCZ:ALN ratios.

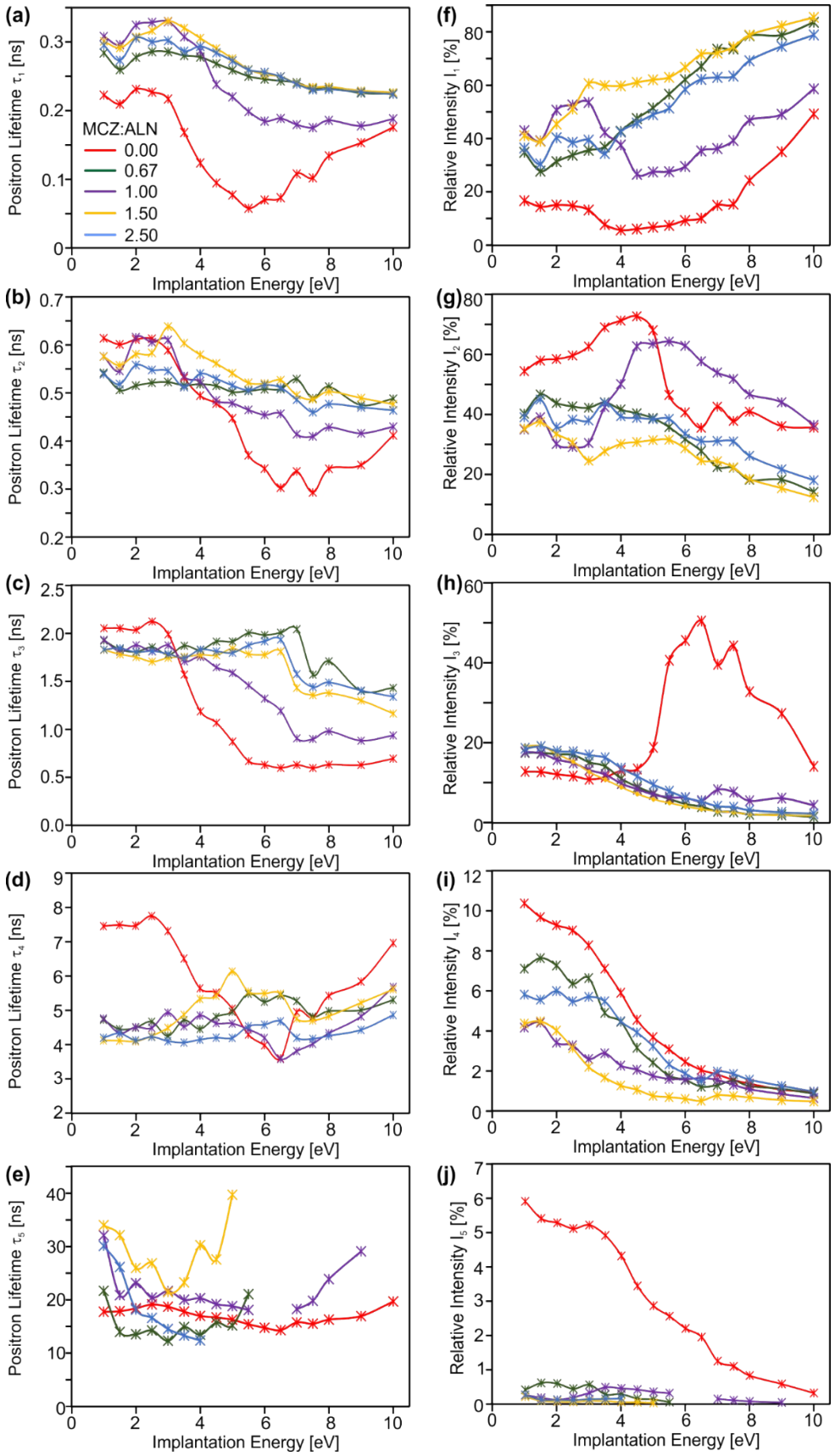

Figure 9: (a-e) Positron lifetime components as a function of implantation energy of alumina films on silicon substrates obtained from precursor mixtures with various MCZ:ALN ratios. (g-j) Corresponding relative intensities.