In-situ observation of irregular void growth in Al thin films during solid state dewetting

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Al thin films are applied as interconnect materials in microelectronics. Elevated temperatures during film processing or device operation can induce degradation of thin films as a result of dewetting due to the high surface to volume ratio.[1] Film break-up can occur via solid state dewetting at temperatures below the melting point and even with a capping layer which suppresses surface diffusion. Despite a passivating oxide Al thin films dewet via film retraction below the continuous surface oxide layer and voids form. The process is dominated by interface and grain boundary diffusion.[2] In our present work, a bicrystalline Al thin film was deposited by molecular beam epitaxy on a (0001) sapphire (alpha-Al_2O_3) substrates. The passivated film served as model system. Environmental scanning electron microscopy (ESEM) allowed to study the dynamics of the solid state dewetting process in-situ during annealing. Before and after annealing, the microstructure and orientation relationships were analysed by SEM and transmission electron microscopy (TEM) methods including electron backscatter diffraction (EBSD) and C_s-corrected atomic column resolved scanning TEM (STEM).

The mazed bicrystalline microstructure consists of two twin-related growth variants possessing the following orientation relationships: {111} Al || (0001) alpha-Al_2O_3 with ±<-110> Al || <10-10> alpha-Al_2O_3 (Fig.1a). The texture of the Al film did not change during annealing as observed by EBSD. Void formation and growth was observed in-situ by ESEM only during annealing in oxidizing (O_2, ~30 Pa) atmosphere at temperatures up to 550°C (Fig. 1b). Site-specific cross-sections of the film|void interface prepared by focused ion beam sectioning revealed the presence of drum-like voids which are continuously covered by a thin oxide layer (Fig. 1c). Beside growth of existing voids, new voids are forming after variable incubation times.

The voids have been found to grow discontinuously because of the pinning of the retracting Al film at obstacles. Their sizes increase according to a power law with exponents spanning from 0.1 to 0.55 for the different, individual voids. The voids adopt a hexagonal shape for undisturbed growth conditions. Pinning at e.g. steps at the substrate leads to irregular void shapes (Fig.1b). The in-situ ESEM experiment allowed to study faceting as well as fingering instabilities caused by pinning. A novel kink-flow mechanism was observed which had not been reported before. Al is transported along the void|Al interface, i.e. the Al free surface, resulting in faceting instabilities. In contrast, no voids were observed to form during experiments in reducing atmosphere (67% H_2/33% N_2, ~50 Pa) which probably impedes the surface oxide stability.

References

[1] C. V. Thompson, *Annu. Rev. Mater. Res.* **2012**, *42*, 399-434.
[2] S. W. Hieke, G. Dehm, C. Scheu, *Acta Mater.* **2017**, *140*, 355-365.

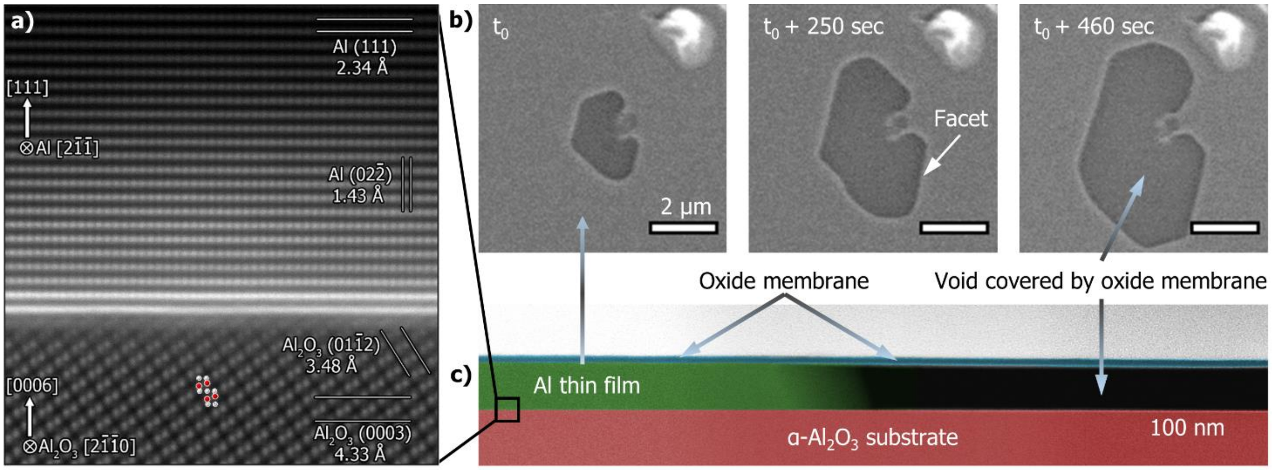

Figure 1: a) Atomic resolved STEM micrograph of a region adjacent to the Al|void interface confirming OR a/b (overlay: Al red, O white). b) Evolution of a faceted void while annealing in the ESEM at 500 °C in oxidizing atmosphere. c) The cross-sectional color-coded HAADF STEM image shows a continuous alumina membrane (bright blue) covering both, the void (black) and the film (green).