In-situ environmental scanning electron microscopy: an investigation on the retraction front during solid-state dewetting of Ag-Cu thin films on amorphous silicone nitride

Okle, P.[1], Wang, Z.[2], Willinger, M.[3] and Spolenak, R.[1]

[1] ETH Zurich, Switzerland, [2] Fritz Haber Institute of the Max Planck Society, Germany, [3] Scientific Center for Optical and Electron Microscopy-ScopeM, ETH Zürich, Switzerland

Thin metal films on amorphous Si_3N_4 are thermodynamically unstable due to the high interfacial energy of the metal compared to the ceramic surface. Enabling sufficient diffusion (annealing) causes the system to minimize the energy contributions of its surfaces and interfaces and to agglomerate into particles. DC magnetron sputtered thin films of immiscible copper and silver (either co-sputtered or as 20 nm layers) with a total thickness of 40 nm were thermally dewetted ($T < T_m$) in an environmental scanning electron microscope to investigate the process of solid-state dewetting in detail. The sputtered thin films are polycrystalline with an average grain size of 20-30 nm. In-situ observation was performed at temperatures between 440 to 550°C with pressures close to $2e^{-4}$ Pa. Additional experiments under gas atmosphere were carried out alternatively with H_2 with a pressure of 30 Pa and CO with partial pressure of roughly 30 Pa with additions of N_2 as imaging gas.

Up to today, several theories of solid-state dewetting are still being discussed, as many effects are influencing the dynamics of the process (i.e. interfacial energies, stresses) [1]. In-situ microscopy enables a dynamic investigation of the process at all stages, from hillock formation to hole formation and growth to retraction and particle agglomeration. The focus will be on the retraction front, were very different velocities of the retraction front could be observed. Local out-of-plane curvatures were analyzed with the in-situ dataset and compared to ex-situ TEM cross-sections of the particles (prepared via lift-out procedure). Other theories than local out-of-plane curvatures are discussed as other factors may play an important role during the process of solid-state dewetting, i.e. the size of grains at the retraction front.

In-situ measurements were performed on multiple samples (either co-sputtered or as 20 nm layers) at different temperatures as well as atmospheres. All stages of the dewetting process are compared on the samples. The different results are compared to each other using traditional surface coverage as well as Minkowski functionals, which allows to investigate the morphologies during the process with a focus on the stochastical behavior.

Different morphologies during dewetting in a either H_2 or CO atmosphere can be seen. While it is not assumed that different pressures have an impact, the influence of these atmospheres on the dewetting process is discussed and compared to similar experiments from P. Jaquet *et al.* [2]. An influence of the atmospheres on the hole formation and on the retraction front as well as on grain growth is observed.

[1] Carl V. Thompson; Annual Review of Materials Research; Vol. 42:399-434 (2012)

[2] P.Jacquet, R.Podor, J.Ravaux, J.Lautru, J.Teisseire, I.Gozhyk, J.Jupille, R.Lazzari; Acta Materialia Vol. 143:281-290 (2018)