Thin Films and Coatings

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TEM investigation of the effect of silicon alloying on the microstructure and stability of amorphous and crystalline Al_2O_3-coatings

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Alumina (Al_2O_3) is a very important polymorphic ceramic material with a wide range of applications due to excellent physical properties. Besides the thermodynamically stable α-Al_2O_3-phase it exhibits several metastable structures, the so called transition aluminas, such as γ-, η-, δ-, θ-, κ-, χ-Al_2O_3. Among these transition aluminas, the γ-Al_2O_3-phase exhibits comparable mechanical properties as the α-Al_2O_3-phase [1]. However, the phase transformation of the metastable γ-Al_2O_3-phase into the stable α-Al_2O_3-phase at high temperatures and the corresponding volume change may induce adhesive and/or cohesive failure and may hence limit coating lifetime. Therefore enhancing the thermal stability of γ-Al_2O_3 is of particular interest. One possibility to improve the thermal stability of γ-Al_2O_3 is to alloy it with a suitable alloying element. Different alloying elements, such as Y, Er, Ti, B, Mo, As, W, Sc, N, S or Si, and their effect on the thermal stability of transition aluminas have been discussed in literature [2, 3]. Based on the ab initio results of Jiang et al. [2], silicon has a stabilizing effect on γ-Al_2O_3. Hence silicon was chosen as an alloying element for Al_2O_3-layers in the present study.

To investigate the effect of Si-alloying on the thermal stability of the Al_2O_3-coatings crystalline and amorphous Al_2O_3-samples with and without silicon alloying have been deposited by using the Filtered Cathodic Arc (FCA) technique.

The unalloyed/Si-alloyed crystalline γ-Al_2O_3-coatings are a few micrometers thick ("Figure 1 a."). The samples were annealed ex situ in atmosphere to temperatures in the range of 1173.15 K to 1473.15 K. For TEM investigations thin cross sections of these samples were prepared by Focused Ion Beam (FIB) using a FEI Strata FIB 205 work station.

The TEM samples of the unalloyed/Si-alloyed amorphous Al_2O_3-coatings were prepared by conventional preparation techniques. The Si-wafer was removed by grinding, dimple grinding and etching with potassium hydroxide (KOH). A 50 nm thick Si_3N_4-layer was added as a etch stop between the Si-wafer and the Al_2O_3-layer to protect the Al_2O_3 against the etching of the KOH. The thickness of the Al_2O_3-layers is limited to a few hundred nanometres to ensure the electron transparency of the samples. The layers structure of the samples before and after the preparation is explained in "Figure 1 b." and "Figure 1 c.". The resulting TEM disc samples were annealed ex situ in an Ar-atmosphere at temperatures in the range of 873.15 K to 1373.15 K.

The TEM investigation of the crystalline and amorphous Al_2O_3-samples were performed by using a FEI Tecnai F20 microscope operated at 200 kV. For the TEM investigations conventional TEM methods, such as bright field imaging (TEM BF) and selected area diffraction (SAD), and Scanning TEM (STEM) techniques were applied to characterize the unalloyed and Si-alloyed nanocrystalline and amorphous samples. During the TEM study the investigation of the morphology, porosity and of the microstructure of the Al_2O_3-coating was of special interest. The TEM data suggest that the Si-alloying influences the morphology and the phase stability of both, the crystalline and the amorphous samples ("Figure 2." and "Figure 3.").

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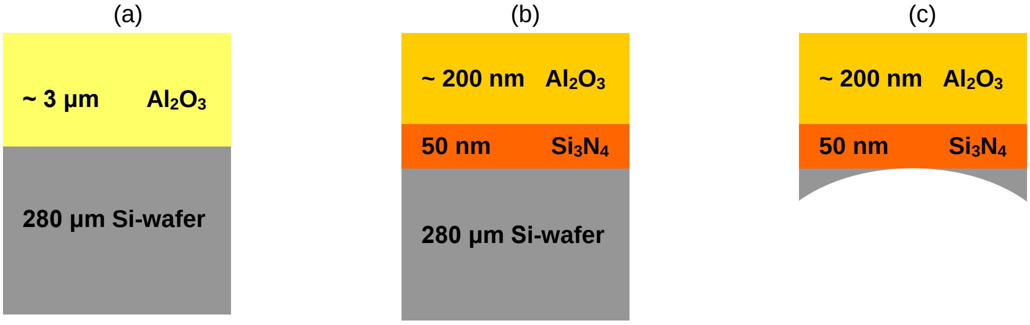

Figure 10. a) Coating structure of the unalloyed /Si-alloyed nanocrystalline Al_2O_3-layers (b) Layer structure of the unalloyed/Si-alloyed amorphous Al_2O_3-layers and (c) Layer structure of the unalloyed/Si-alloyed amorphous Al_2O_3-layers after dimple grinding and etching.

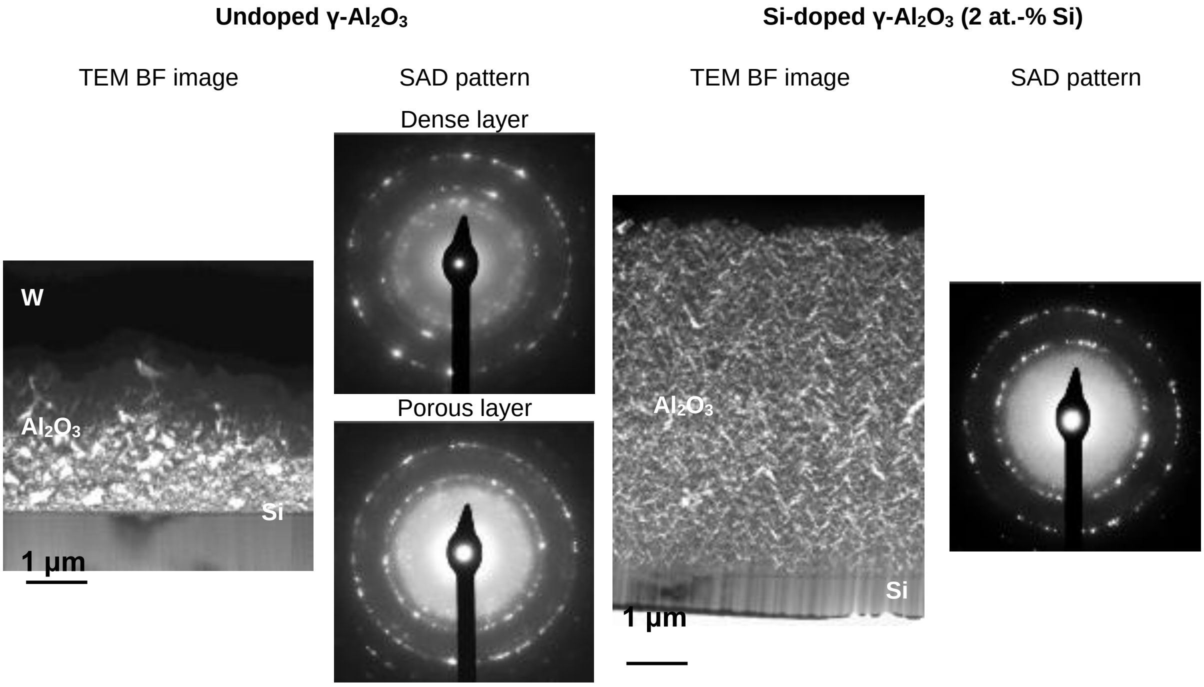

Figure 11. TEM BF overview images of the FIB lamellae of the undoped/Si-doped nanocrystalline Al_2O_3-samples after annealing to 1173.15 K and the corresponding selected area diffraction (SAD) pattern.

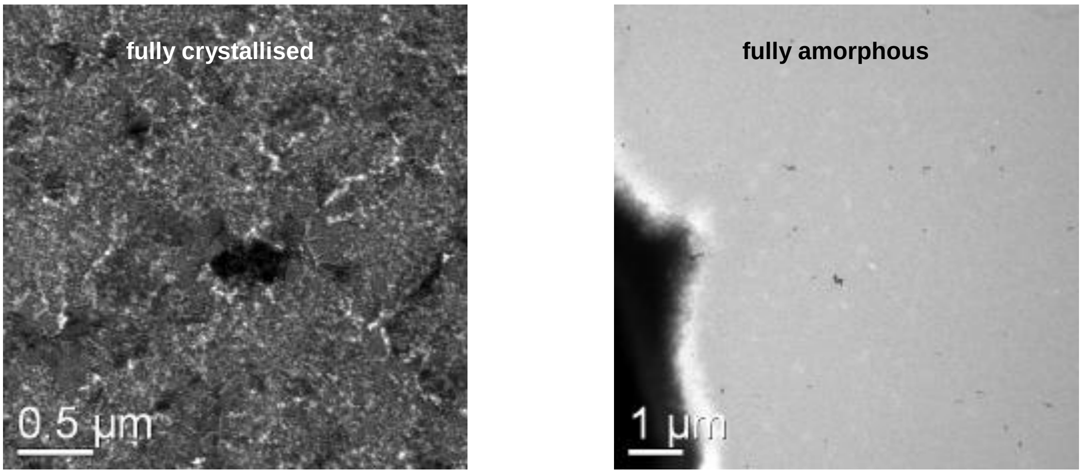

Figure 12. TEM BF images of the annealed undoped amorphous Al_2O_3-sample (923.15 K, on the left) and of the annealed Si-doped (2 at.-% Si) amorphous Al_2O_3-sample (1023.15 K, on the right).