

Supporting Information:

Locally reinforced polymer-based composites for elastic electronics

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1. Silica coating on Ultra-High Magnetic Responsive (UHMR) alumina platelets

The surface modification of UHMR alumina platelets was performed by a modified Stöber method described by Graf and co-workers¹. These platelets are first functionalized with a non-ionic, amphiphilic polymer (polyvinylpyrrolidone, PVP, MW 360 = kg.mol⁻¹) in water and subsequently transferred to an ammonia/ethanol solution. Next, the silica coating is formed by slowly adding tetraethyl orthosilicate (TEOS) with a syringe pump in order to reduce secondary precipitation of SiO₂ particles. In a typical synthesis, 10.0 g of UHMR platelets are dispersed into 900 mL solution of PVP in water (C = 8.75 g.L⁻¹) and stirred overnight. The platelets were then filtered and washed with 100 mL of ethanol. After drying at room temperature for approximately one hour, the platelets were transferred to a 4.2 vol% solution of ammonia in ethanol absolute and stirred for 30 min at 600 rpm. For the silica coating, 40 mL of TEOS solution in ethanol absolute were added by a syringe pump working at a flow rate of 800 μL.h⁻¹. Finally, the silica-coated platelets (SiO₂@UHMR-Al₂O₃) were filtered, washed with approximately 1L of ethanol and dried at 70°C for 4 hours.

2. Characterization of SiO₂@Al₂O₃

Figure S1a shows the Fourier Transform Infrared (FT-IR) spectra in Diffuse Reflectance mode (DRIFT) of UHMR-Al₂O₃ (black curve) and SiO₂@UHMR-Al₂O₃ (red curve). The coated platelets exhibit typical infrared absorptions of SiO₂, such as stretching of free (3660 cm⁻¹) and associated –O-H groups (2800-3500 cm⁻¹) and asymmetric (1186 cm⁻¹) and symmetric (1067 cm⁻¹) bending of Si-O-Si groups. The

absorption band at 1655 cm^{-1} was assigned to the stretching of carboxyl groups of the pyrrolidone moiety,² which suggests that PVP remains adsorbed on platelets' surface.

The silica coating was further characterized by Scanning Electron Microscopy (SEM) and Energy Dispersive X-ray Spectroscopy (EDX). Nanoparticles adsorbed on the platelet surface can be identified in the SEM image shown in Figure S1b. Qualitative analysis using EDX on different regions of the platelet surface indicated that the bare surface of alumina platelets were coated with silica (Figure S1c, red square in Figure S1b). Interestingly, EDX analysis of the green square in Figure S1b indicated the presence of alumina, silica and magnetite, suggesting that the magnetic nanoparticles were also coated by a layer of SiO_2 during the *in situ* chemical deposition process.

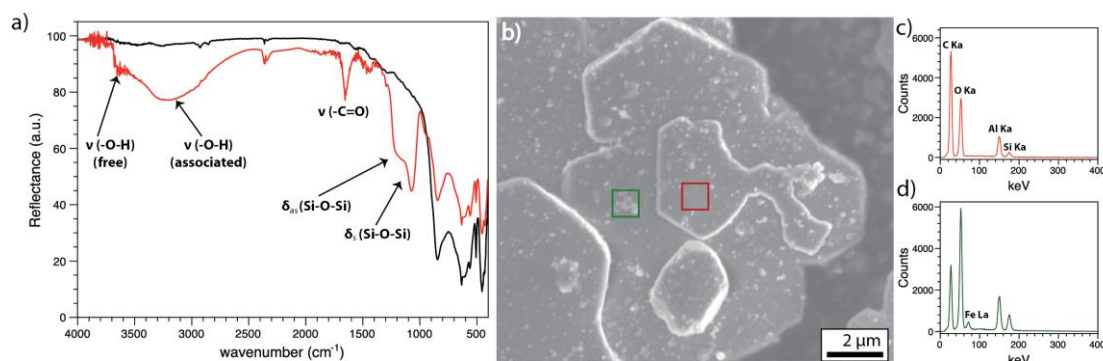


Figure S1 - Characterization of the SiO_2 @UHMR- Al_2O_3 platelets. (a) FT-IR spectra in DRIFT mode of UHMR- Al_2O_3 (black) and SiO_2 @UHMR- Al_2O_3 (red). (b) SEM image of SiO_2 @UHMR- Al_2O_3 showing the presence of nanoparticles adsorbed on the platelet surface. (c) and (d) EDX qualitative spectra of the regions indicated in (b).

3. Elastic modulus of polymer-based composites as a function of the concentration of alumina platelets

Figure S2 shows the effect of the concentration of alumina platelets on the elastic modulus of poly(vinyl alcohol) (PVA), polyurethane (PU) and polydimethylsiloxane (PDMS). The plots indicate a remarkable reinforcement of the polymer matrices upon addition of the alumina platelets. In particular, the hydrophobized platelets interact strongly with the PDMS matrix as was further observed with a good connection apparent in scanning electron micrographs of the freeze-fractured cross-section of such a sample (Figure S3).

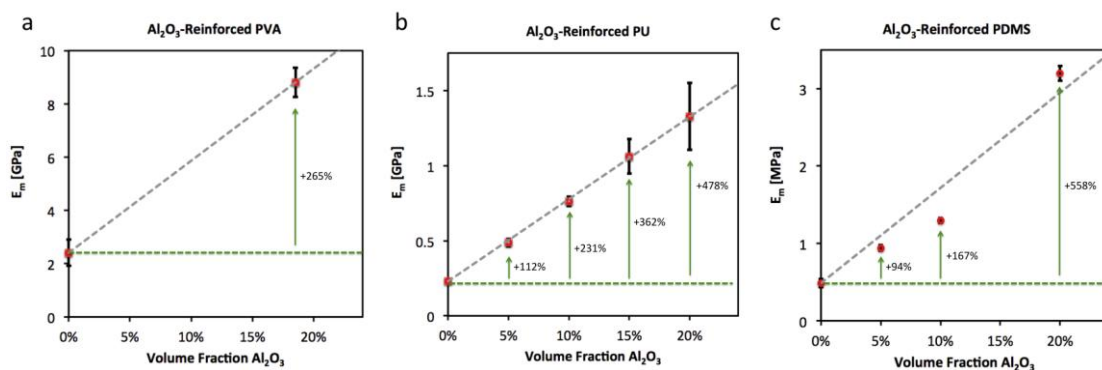


Figure S2 – Al_2O_3 reinforced (a) PVA, (b) PU, and (c) PDMS matrices at varying levels of alumina content.

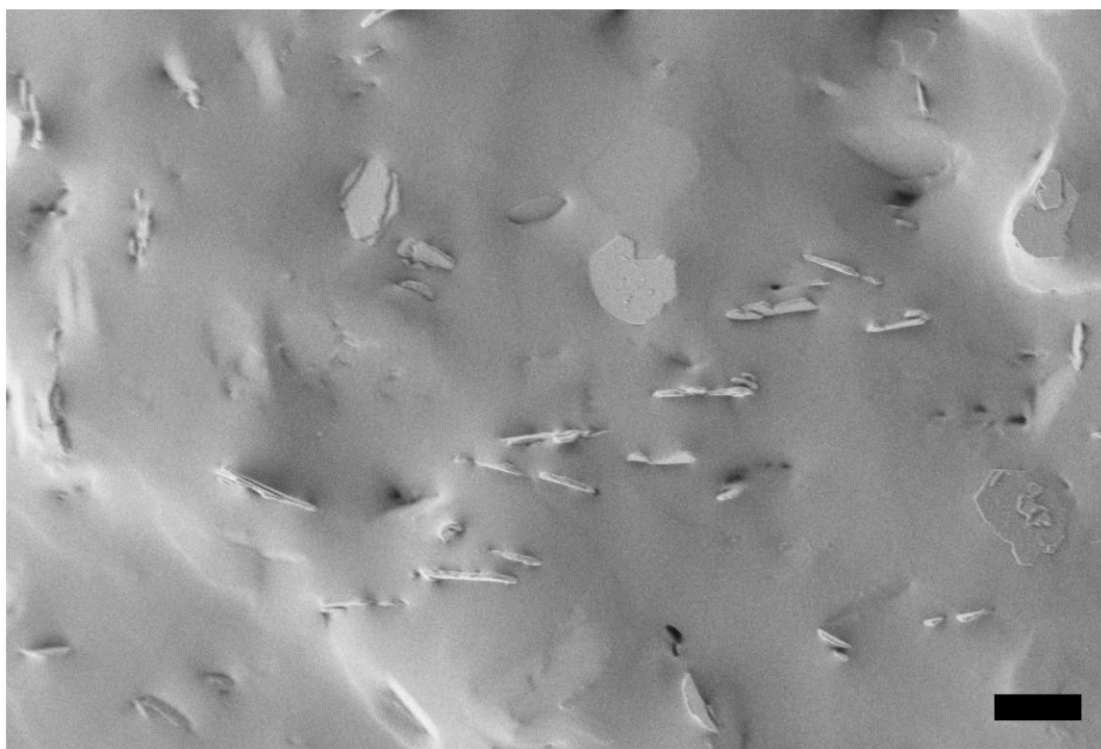


Figure S3 – Scanning electron micrograph of a freeze-fractured cross-section of Al_2O_3 reinforced PDMS showing a good interaction between the hydrophobized platelets and the matrix. The scale bar represents 10 μm .

References:

- [1] C. Graf, D. L. J. Vossen, A. Imhof, A. van Blaaderen, *Langmuir* 2003, 19, 6693.
- [2] J. J. Behen, R. F. Dwyer, B. A. Bierl, *Analytical Biochemistry* 1964, 9, 127.