## Supporting Information

## Ultrastrong Composites from Dopamine Modified Polymer-Infiltrated Colloidal Crystals

Faroha Liaqat<sup>1</sup>, Muhammad Nawaz Tahir<sup>1\*</sup>, Hannah Huesmann<sup>1</sup>, Phillip Daniel,<sup>1</sup> Michael Kappl<sup>2</sup>, Günter K. Auernhammer<sup>2</sup>, Dirk Schneider<sup>2</sup>, Ingo Lieberwirth<sup>2</sup>, Kookheon Char<sup>3</sup>, George Fytas<sup>2, 4</sup>, Hans-Jürgen Butt<sup>2</sup>, and Wolfgang Tremel<sup>1\*</sup>

<sup>&</sup>lt;sup>1</sup> Institut für Anorganische Chemie und Analytische Chemie, Johannes Gutenberg-Universität, Duesbergweg 10-14, 55099 Mainz, Germany

<sup>&</sup>lt;sup>2</sup> Max Planck-Institut für Polymerforschung, Ackermannweg 10, 55128 Mainz, Germany

<sup>&</sup>lt;sup>3</sup> School of Biological and Chemical Engineering, Center for Functional Polymer Thin Films, Seoul National University, Seoul 151-744, South Korea

<sup>&</sup>lt;sup>4</sup> Department of Materials Science, University of Crete and IESL/FORTH, 71110 Heraklion, Greece



Scheme S1. Synthesis of the poly (active ester) poly (pentafluoro-phenylacrylate) (PFA).



Scheme S2. Synthesis of the dopamine modified polymer



Scheme S3: Sketch of the connectivity of the  $Fe_3O_4$  nanoparticles through the dopamine modified polymer.



Figure S1. <sup>1</sup>H NMR spectrum of poly(pentafluorophenyl acrylate)



Figure S2. <sup>19</sup>F NMR spectrum of poly(pentafluorophenyl acrylate)



Figure S3. <sup>1</sup>H NMR spectrum of the dopamine modified polymer.



**Figure S4.** A comparison of the FTIR spectra of pure dopamine modified polymer (black) and the nanocomposite with  $Fe_3O_4$  NPs (green) is shown. The binding between the polymer and iron oxide NPs is evident from the absence of the phenolic –OH stretching in the hybrid.



**Figure S5.** Digital photograph A) as prepared dopamine modified polymer solution in DMA B) after heating the solution at 120°C for 20 minutes, C) dissolution in DMA using ultrasonication for several hours.



Figure S6. Nano-indentation of a multilayer  $Fe_3O_4$ / dopamine modified polymer composite. The strong crosslinking between the  $Fe_3O_4$  nanoparticles and the multidentate polymer ligand help the matrix resist deformation and make the composites harder and stronger. This figure shows a model illustrating the cohesive role of the catechol-polymer in the multilayer films. The multilayers contain a high crosslinking density. When the film is indented, the polymer strands have to be unfolded and a multitude of strong metal-catechol must be broken.



**Figure S7.** Structural colors shown by multilayers of dopamine modified polymer/Fe $_3O_4$  due to multilayer interference with increasing bilayers (BL)



Figure S8. EDX analysis of dopamine modified polymer/Fe<sub>3</sub>O<sub>4</sub> multilayers coupled with TEM. (A) TEM image of the multilayers showing long range ordering. The two points indicated on the TEM micrographs correspond to the spectra in (B) representing the elemental content in the multilayers.



**Figure S9.** A comparison of TEM images of different samples of dopamine modified  $/Fe_3O_4$  nanoparticles consisting of a varying number of multilayers. **(A)** A sample prepared by 32 spin-coating cycles shows the existence of large defected regions as the number of multilayers increase. **(B)** On the other hand, a sample prepared by 12 repeated dip-coating cycles reveal the existence of ordered regions (red circles) and defects (green circle) in addition to a regular cross-linked network, highlighting the presence of inhomogeneous regions in contrast to the superior spin coating method.