

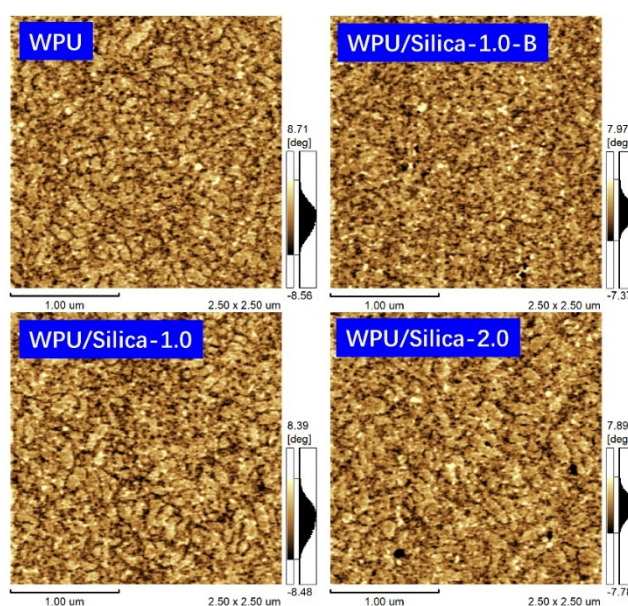
# In situ incorporation of alkyl-grafted silica into waterborne polyurethane with high solid content for enhanced physical properties of coatings

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**Figure s1.** Phase images of WPU and WPU/Silica coatings.

Microphase structure is essential for mechanical properties of PU coatings. It is known that hard-segment and soft-segment phases contribute to the strength and flexibility of polyurethane, respectively. The AFM phase images of WPU (Figure 1) showed a typical micro-phase separation structure in which the bright domains corresponded to the microphase of the hard-segments while the dark domains attributed to the soft-segments. With incorporation of alkyl-grafted silica, the bright areas increased because the silica particles were embedded in the polymer molecular chains, forming hybrid domains with higher modulus.

hydrophobic fumed silica had been successfully incorporated into organic matrix of polyurethane (NWPU) at the concentrations of 0.5, 1.0, 1.5, and 2.0 wt% (labeled as WPU/Silica-0.5, WPU/Silica-1.0, WPU/Silica-1.5, and WPU/Silica-2.0, respectively). (below 1.5 wt%), and a strong interfacial bonding existed between the organic-inorganic matrix and silica particles. As a result, the thermal stability and stiffness of NWPU/S hybrids was significantly enhanced. Moreover, the hybrids were found to be superior in the UVA irradiation performance over pure NWPU. Moreover, the hybrids increased the phase separation of polyurethane, resulting in a higher glass transition temperature. Further increasing content of hydrophobic fumed silica, silica particles were observed

**Figure s2.** Photos of WPU and WPU/Silica coatings

The photos of WPU and WPU/Silica coatings were taken by putting them on a piece of printed paper. As shown in Figure 2, there was no apparent specular reflection on all samples. Besides this, the incorporation of alkyl-grafted silica had little influence on transparency of coatings when the content of silica is less than 2%. This is consistent with research conducted by Eslami et al [1], where they found the addition of nano-silica below 2% would not result in a decrease in the degree of the transparency.

#### **Reference:**

1. Eslami, R.; Bagheri, R.; Hashemzadeh, Y.; Salehi, M. Optical and mechanical properties of transparent acrylic based polyurethane nano silica composite coatings. *Progress in Organic Coatings* **2014**, *77*, 1184-1190.