Project B6: Multifunctional hybrid polymers for adaptive nanocrystalline multilayer composites

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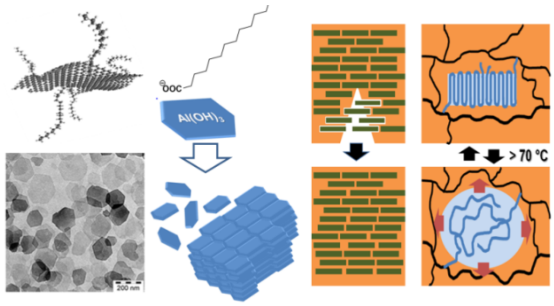
Current state of the research. The development of advanced lightweight materials is inspired by natural multilayer composites, such as nacre. A synthetic example is the layer-by-layer (LbL) assembly of polyelectrolytes and nanoparticles in thin films (Decher group). Multilayer coatings, produced from aqueous polymer solutions with clay nanosheets, afford high strength and stiffness. However, layered single crystal alumina and graphene composites are superior regarding mechanical reinforcement. Due to the high polymer melt viscosity, the dispersion and layer-assembly of inorganic nanosheets in polymer melts is very difficult. Tailoring the surface of the nanoparticles can facilitate processing. Hence, an important objective is to understand how polymer modification of two-dimensional nanosheets (e.g. graphene, inorganic single crystals) enables the control of layer formation in molding and coating applications (see Figure). Also, the role of polymer crystallization is unexplored and offers a promising potential for designing advanced nanocrystalline multilayer materials. In principle, polymer crystallization in the confined environment of the interlayers could lead to the formation of polymer nanocrystal, accounting for adaptive mechanical reinforcement and self-healing properties (see Figure).



Contributions of the proposers. The Lutz group has expertise in tailoring and characterizing block and graft copolymers. The Mülhaupt group has expertise in chemistry, characterization and processing of polymer nanocomposites. During the first funding period, their close collaboration, and also with the Reiter group, has led to semicrystalline composites with POSS nanoparticles and crystalline isotactic polystyrene. This successful collaboration will be expanded towards multilayer composites.

Research project and collaborations. First, by joining expertises (Lutz/Mülhaupt) new polymer-modified 2D hybrid materials will be derived from graphene and single crystal aluminum hydroxide (Gibbsite). By exploring “grafting from” processes (ROMP and ROMP/ hydrogenation tandem catalysis) amorphous polyoctenamer elastomer and semicrystalline polyethylene are attached to norbornene-functionalized nanosheets. Tailoring such polymer-modified 2D hybrids will reveal the influence of amorphous and crystalline polymer on nanostructure formation. This includes nanostructure formation via the interplay of 2D hybrid polymer modifiers with segments of nanophase separated polyethylene-block-poly(ethylene-co-butene) copolymers. Second, melt processing and LbL assembly by spraying (Decher) will be explored to produce novel multilayer coatings.

Work plan. The PhD project has three work packages. *WP1. Preparation of polymer-modified 2D hybrid materials:* Preparation of graphene, single-crystal Gibbsite and alumina dispersions (Mülhaupt). Grafting-from polymerization for polymer hybrids (Lutz). Polymer characterization (Lutz) and nanostructure formation (Mülhaupt). *WP2. Self-assembly and multi-layer formation:* Tailoring polymer modification (Lutz) and melt processing of 2D hybrids (Mülhaupt). LbL assembly of via spaying (Decher). *WP3. Nanocrystal formation and adaptive self-reinforcement:* Polymer crystallization in interlayer confinement (Mülhaupt). Study of temperature- and stress-induced self-healing of multilayer composites (Mülhaupt). Film characterization (Decher). Collaboration: crystallization (Reiter), tribology (Gauthier).