# GREEN ONE-POT SYNTHESIS AND PROCESSING OF POLYIMIDE-SILICA HYBRID MATERIALS

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Inorganic-organic hybrid materials allow for combining features of an inorganic component with those of an organic component material by intimately connecting both physically or chemically. Employing this concept, one can for instance couple the low densities and ease of processing found in organic polymers with the mechanical performance of inorganic materials, such as ceramics. Currently employed syntheses of those materials, especially of the organic component, unfortunately require harsh and toxic conditions. We have recently reported a technique that allows for the green synthesis of fully aromatic polyimides (PIs) in nothing but water – *Hydrothermal polymerization (HTP)* – and additionally generates outstanding crystallinity.[1-3] HTP mimics conditions under which many inorganic compounds, such as metal oxides, are formed in nature. The fact, that both reactions, the polymerization of the polyimide and the formation of the inorganic metal oxide are mechanistically speaking polycondensation reactions, motivated us to tackle simultaneous hydrothermal synthesis of both components. In terms of green chemistry, such concurrent one-pot approaches are clearly preferable over commonly employed sequential routes, as the reduced number of synthetic steps automatically diminishes e.g. solvent consumption and purification steps. With this contribution, we present the first example of a one-pot, fully green synthesis of an inorganic-organic hybrid materials.[4] Specifically, we selected a thermoplastic, semialiphatic PI as organic, and the metal oxide $SiO_2$ as inorganic component, using hexamethylenediamine (HMDA), pyromellitic acid (PMA), tetraethoxysilane (TEOS) and (3-aminopropyl)triethoxysilane (APTES) as precursors. Our study moreover reports a subsequent up-scaling and processing step, in which macroscopic cylindrical pellets (4 cm in diameter, 0.5 cm in thickness) were obtained *via* sintering.  All hybrid materials were fully characterized using ATR-FT-IR, SEM, EDAX, PXRD and solid-state MAS-NMR. The results of these analyses show that the PI and $SiO_2$ can be covalently connected in a green one-pot hydrothermal approach.

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[2] M. M. Unterlass, *Materials Today*, 2015, **5**, 242-243

[3] B. Baumgartner, M. Puchberger and M. M. Unterlass, *Polymer Chemistry*, 2015, **6,** 5773-5781

[4] L. Leimhofer, B. Baumgartner, M. Puchberger, T. Prochaska, T. Konegger and M. M. Unterlass *J. Mater. Chem. A.* 2017, doi: 10.1039/C7TA02498C