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Preparation of bioinspired, anisotropic monolithic and composite materials via magnetic self-assembly

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In this work, a magnetic template was prepared from superparamagnetic magnetite nanoparticles and used to control the structure of sol-gel processed materials. The sol-gel process, yielding in highly porous organic and inorganic materials is promising for structural control through magnetic forces as the liquid precursor state enables homogeneous mixing with the template. An externally applied magnetic field will self-assemble the magnetic template, which is dictating its structure to the organic or inorganic material. After the gelation, the structure is irreversibly fixed. Uniform magnetic fields were used to obtain fiber-like structure in the end material, while rotating magnetic fields resulted in stacked 2D platelet-like structures. First, the highly magnetically responsive and solid template was synthesized via mini-emulsion polymerization during which multiple superparamagnetic magnetite nanocrystals are embedded inside a polystyrene particle. Different stabilizing agents were used for the template to tune its interaction with the sol-gel reactive medium. In a second stage, the template was prepared by the formation of magnetite nanocrystal clusters through a solvothermal process.

We prepared inorganic silica and organic resorcinol-formaldehyde gels with anisotropic structures using the magnetic template. Structurally controlled silica monoliths were turned into composite material by the filling the porous structure with a polymer. The structural control, yielding in increased stiffness paired with the elastic properties of the polymer resulted in bioinspired, mechanically reinforced materials. The same procedure was tried in the case of titania monoliths. A machine-learning approach combined with statistical models was applied to study the influence of the reaction parameters on the mechanical properties of the anisotropic monoliths compared to their isotropic counterparts.

A pyrolysis step transformed the anisotropic organic gels in structurally controlled carbon pellets. It was revealed, that the magnetite template is not only controlling the morphology of the material, but also causes in pore alignment. Magnetite is reduced into metallic iron during the pyrolysis. Making use of dispersed iron catalyst in the carbon material and the aligned pores, the structurally controlled carbon pellets present themselves promising candidates for gas conversion experiments.

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