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Linking structural and mechanical properties within hydrogels based on ionene polyelectrolytes and clay nanoplatelets

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Hydrogels are at the forefront of scientific attention especially in the biomedical field thanks to their high-water content, their mechanical and swelling properties [1]. Association of hydrogel and clay nanoparticles as additives has a great potential in the design of materials that exhibit additional enhanced mechanical, swelling or barrier properties [2]. The functional properties of these materials are intimately linked to their structural organisation.

We investigate the organisation of clay nanoplatelets within a hydrogel based on modified ionenes [3], cationic polyelectrolytes forming physically crosslinked hydrogels induced by hydrogen bonding and π-π stacking. Combination of small angle X-ray and neutron scattering (SAXS, SANS) reveals the structure of the polyelectrolyte network as well as the organisation of the clay additives [4]. The clay-free hydrogel network features a characteristic mesh-size between 20 to 30 nm, depending on the polyelectrolyte concentration. Clay nanoplatelets inside the hydrogel organise in a regular face-to-face (stacking) manner, with a large repeat distance, following rather closely the hydrogel mesh-size. This suggests that the nanoplatelets "decorate" the underlying polyelectrolyte network. Further, the clay-compensating counterions (Na^{+}, Ca^{2+} or La^{3+}) and the clay type (montmorillonite, beidellite) both have a significant influence on nanoplatelet organisation. The degree of nanoplatelet ordering in the hydrogel is very sensitive to the negative charge location on the clay platelet (different for each clay type). Increased nanoplatelet ordering leads to an improvement of the elastic properties of the hydrogel. On the contrary, the presence of dense clay aggregates (tactoids), induced by multi-valent clay counterions, destroys the nanoplatelet "decoration" of the hydrogel network and a reduction of elastic modulus of the hydrogel is observed.

In the long-term, rheo-SAXS will be used to shed light on whether the anisotropy of clay nanoparticles could yield an anisotropic mechanical response of the hydrogel [5].

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[2] L. Z. Zhao, C. H. Zhou, J. Wang et al, *Soft Matter,* 11*,* 9229 (2015)
[3] L. Z Y. Misawa, N. Koumura et al, *Macromolecules*, 41, 8841-8846 (2008)
[4] C. Hotton et al. (in submission)
[5] A. M. Philippe, C. Baravian, V. Bezuglyy et al, *Langmuir,* 2917, 5315-5324 (2013)