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Superchaotropic polyoxometalates as smart crosslinkers for cellulose ethers

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α-Keggin poyloxometalates (POMs) with low charge density, such as SiW12O404- interact strongly with non-ionic hydrated interfaces driven by a water-mediated driving force, called the chaotropic effect. In aqueous solution of hydroxypropylcellulose (HPC), this binding results in more than 100-fold increase in viscosity at millimolar HSiW-concentrations, and even induces gelation at higher HSiW-concentration at c(HSiW) > 30 mM. By combination of SAXS, SANS, NMR, we show that this thickening effect appears because the POMs bind tightly to the polymer with a binding constant KA= 200 mM-1. The ion-binding leads to electric charging of the polymer at low Siw-concentration, which transitions into physical crosslinking of adjacent polymer chains by the bound ions under screened electrostatics. As Keggin POMs are photocatalysts, photoredox reactions can be employed to reduce the POM, increase its charge density and thus diminish its binding to HPC. We thus demonstrate that photoredox cycles can be used to generate switchable HPC-solutions and gels. The concept of smart chaotropicity for POMs enables a unique and simple strategy to make non-ionic polymers addressable through UV-light for soft material applications.

Abstract Title

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