Thermodynamics of the anisotropic equilibrium state in thermo responsive core-shell hydrogels

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This work is a theoretical investigation of spherical hydrogel particles exhibiting anisotropic equilibrium states. With polyelectrolytes such states can occur through sorption of oppositely charged macroions, leading to a complete collapse of some regions of the particle while leaving other regions fully swollen. With neutral polymers they can appear in gels with two regions having different critical solution temperatures (CST).

As an example of the latter, this work deals specifically with particles consisting of a poly(N-isopropylacrylamide) core and poly(N-isopropylmethacrylamide) shell. These polymers have lower CSTs at 34 °C and 44 °C, respectively. Experimental studies including intra-particle density distributions in such particles have been reported [1].

Using a parameterised Flory-Huggins model for the free energy of mixing [2], local osmotic equilibrium is coupled to mechanical equilibrium in the particle as a whole [3,4]. The equilibrium deformational state of the particle can be predicted by a nonlinear differential equation.

Although only semi-quantitative agreement with experiments is achieved, the model predicts the expected general characteristics of how the regions affect each other. It provides a better understanding of the elastic interplay between the core and shell regions and its impact on the equilibrium swelling. This will be relevant for investigating the possibilities of true phase separation within gel particles.

Deformational equilibrium state calculated at a temperature between the CSTs. The collapsed core causes anisotropic and inhomogeneous deformation of the swollen shell.