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Pressure-induced phase transition in polymer brushes: structural studies and thermodynamic predictions

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Polymers are key elements in modern material science, and, since the first polymer synthesis in 1907, tremendous progress in polymer chemistry were made. Today, an endless assortment of polymers –whose properties are adapted to specific needs –is available. In many cases, a thin polymer film, the coating, is sufficient to impart desired properties to the material.
One of the most employed stimuli to tune phase transition in polymer coatings is temperature. Since the first reports of the thermo-responsive behaviour of poly(N-isopropylacrylamide) (PNIPAM) in an aqueous environment in the late sixties, PNIPAM has become the most studied model for non-ionic polymer systems. However, temperature is not the only physical parameter to tune the phase behaviour of polymeric systems. Pressure can be similarly used to control the phase behaviour of polymer solutions and thin films.
Herein, we provide an extensive overview of the phase behaviour of end-grafted PNIPAM brushes as a function of pressure and temperature. The phase behaviour, extracted from the neutron reflectometry curves, is compared with the phase behaviour of semi-dilute solutions. Further, we show that the coexistence line as a function of pressure and temperature can be predicted assuming a two-state model –swollen and collapsed –knowing the different derivatives of the free energy of collapse as a function of pressure and temperature. These quantities can be precisely accessed using calorimetric and densitometric measurements.
The results evidenced that the pressure-temperature phase behaviour of polymer solutions and coatings can be predicted from simple, laboratory-scale experiments, paving the way for the rational design of smart coatings with pressure and thermo-responsive behaviour.

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