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Kinetics of mesoglobules formation and disintegration in solutions of thermoresponsive polymers after fast pressure jumps

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The formation and disintegration pathways of polymeric nanoparticles are of key importance for their use for the transport and release of substances. Poly(N-isopropylacrylamide) in aqueous solution forms nanoparticles ("mesoglobules") above its cloud point, which depends on pressure. In the temperature-pressure frame, the coexistence line is an ellipse with a maximum at ~60 MPa and 35 °C. Isothermal pressure jumps across this line allow for time-resolved small-angle neutron scattering investigations of the formation and disintegration of mesoglobules from the solution with a time-resolution of 50 ms and in a large range of length scales [1].

In the low-pressure regime, we find that the growth of the mesoglobules proceeds via diffusion-limited coalescence and, later, via hindered growth, which is due to the rigidification of the mesoglobules. In contrast, in the high-pressure regime, the chains stay hydrated and mobile, and the diffusion-limited coalescence proceeds without hindrance. The disintegration of the mesoglobules proceeds via the release of single polymers from the mesoglobules' surface or via swelling, depending on the target pressure and thus on the osmotic pressure of water. Thus, pressure jumps allow the identification of a number of kinetic processes that are relevant for the efficient formation or cargo release.

B.-J. Niebuur, L. Chiappisi, F. A. Jung, X. Zhang, A. Schulte, C. M. Papadakis, ACS Macro Lett. 7, 1155 (2018), Macromolecules 52, 6416 (2019) and RSC Nanoscale 13, 13421 (2021)

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