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Formation and growth of mesoglobules in aqueous poly(N-isopropylacrylamide) solutions revealed with fast pressure jumps

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Understanding of the kinetics of phase separation is of importance for numerous systems, for example to control demixing processes. The thermoresponsive polymer Poly(N-isopropylacrylamide) (PNIPAM) may serve as a model system to investigate the kinetics of phase separation. In aqueous solutions at temperatures above the cloud point, PNIPAM forms stable mesoglobules with a size and degree of hydration that depend strongly on pressure [1]. Previous time-resolved small-angle neutron scattering (TR-SANS) investigations of mesoglobule growth following a temperature change were hampered by the slow temperature equilibration [2]. To elucidate the formation and early stage growth of the mesoglobules in an aqueous PNIPAM solution, we applied TR-SANS after pressure jumps inducing the phase separation, enabling a time resolution of 50 ms. Three processes of mesoglobule growth are identified. During the first ~1 s, clusters of chains are formed which grow only slowly due to interconnecting chains. After disruption of these networks, mesoglobule growth is limited by diffusion and inner restructuring: Gradually, a dense polymer shell is formed that hinders the coalescence at later stages, resulting in a very slow growth.
[1] Niebuur, B.-J.; Papadakis, C.M. et al., ACS Macro Lett. 2017, 6, 1180
[2] Meier-Koll, A.; Papadakis, C.M.; Müller-Buschbaum, P. et al., Langmuir 2012, 28, 8791

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