

Supporting Information

Effect of Filtration Forces on the Structure of Casein Micelles

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GISAXS models and fits. Generally, it is difficult to distinguish prolate and oblate shapes from small angle scattering data in solution [1]. In contrast to the case in solution, all particles in the deposits are oriented in one direction due to the filtration force. Our model assumes ellipsoids as shown in Fig. S1.

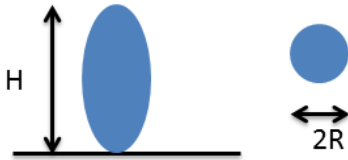


Fig. S1: IsGISAXS model for an ellipsoidal particle shape [2].

We fitted the scattering functions with the IsGISAXS program. As starting conditions of the fit we assumed a spherical shape ($H = 150$ nm; $R = 75$ nm). In all cases the fit converged in the right direction (towards prolate at 400 mbar and towards oblate at 600 mbar). Fig. S2 shows exemplary both the data and the fit for 600 mbar.

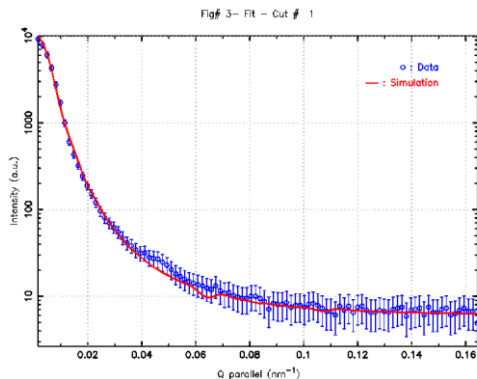


Fig. S2: Data ($\Delta p = 600$ mbar) and simulation resulting from a fit with the IsGISAXS program [2].

Atomic force microscopy. We used a WITec– alpha500 atomic force microscope for intermittent-15 contact measurements (AC mode) in air under room conditions ($T = 23\text{ }^{\circ}\text{C}$, relative humidity: 30 %). The cantilever had a resonance frequency of 285 kHz and a spring constant of 42N/m. The scan rate was 0.66 Hz and images were recorded with 300 Point/Line. AFM supports the GISAXS data concerning the deformation of casein micelles. Fig.S3 shows an AFM image of casein micelles (diameter measured by solution DLS: 150 nm) deposited on a micro-sieve using a trans-membrane pressure of $\Delta p = 600\text{ mbar}$. The mean lateral size of the casein micelles is $D \sim 250\text{ nm}$ and corresponds to the width resulting from the analysis of the GISAXS data. Furthermore, signs of a flattening are visible (inner circle marked by arrow).

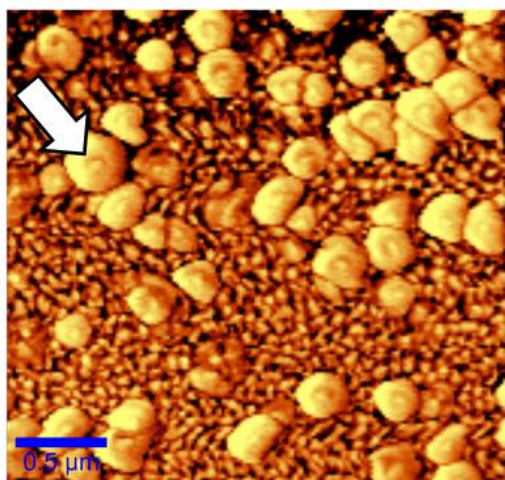


Fig. S3: AFM image of casein micelles deposited on the non-porous regions of a micro-sieve during frontal filtration ($\Delta p = 600\text{ mbar}$).

Background correction for the SAXS data. For background correction we used the scattering signal from an empty pore on the micro-sieve, which was scanned under the same conditions as used for the filled pore. Fig. S4 shows the scattering functions of both empty pore (A) and pore filled with casein (B) obtained after radial averaging and transformation on a q -scale. No further data treatment or mathematical operation was performed. As the comparison in Fig.S4 C,D shows, both scattering functions show similar scattering intensities at $q > 2\text{ nm}^{-1}$ while there are substantial differences in the q -range between $0.3\text{--}2\text{ nm}^{-1}$. Hence, we directly subtracted the background signal from the signal of the sample. The background signal does not change in the considered q -range as measurements on different empty pores showed.

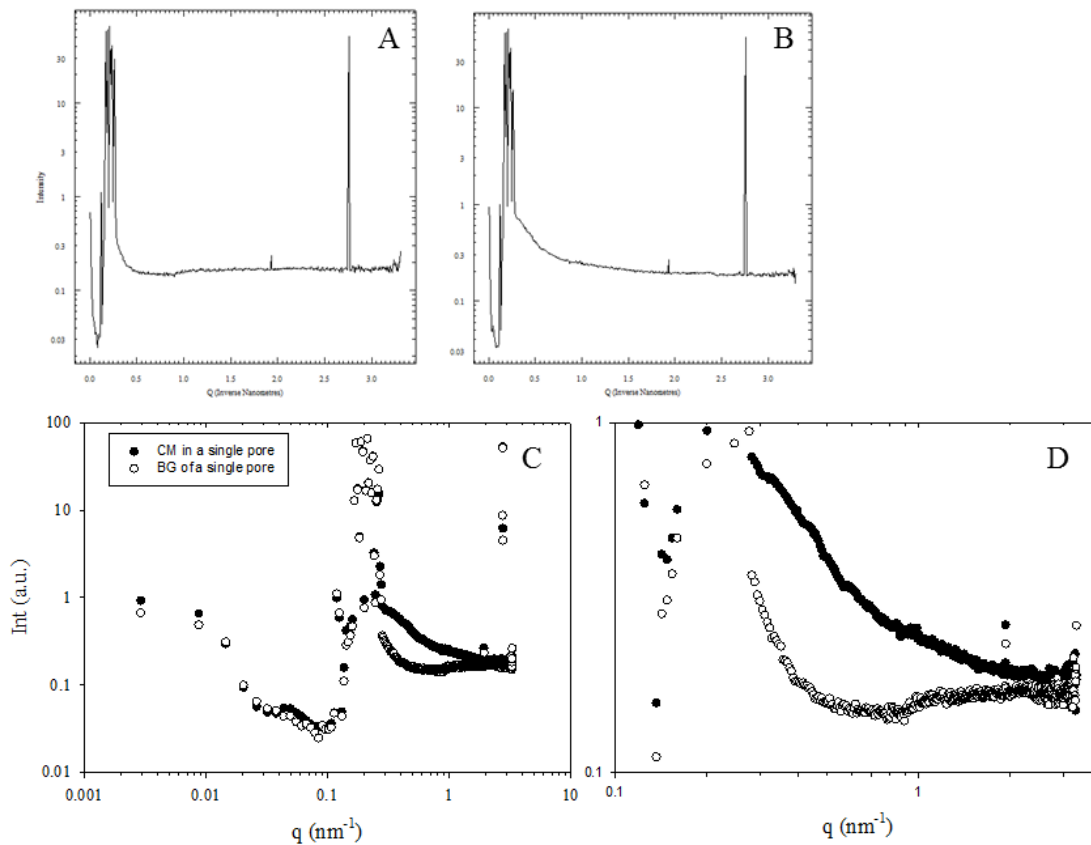


Fig. S4: (A) Scattering function of an empty pore (A) and a pore filled with casein (B) of the micro-sieve obtained after radial averaging of the scattering patterns; (C) Comparison of both scattering functions on a double logarithmic scale; (D) Enlarged view of the relevant q -range in (C).

Literature:

[1] Vass S., Pedersen J.S., et al., (2008). *Langmuir*. **24**, 408

[2] R. Lazzari (2002). *J. Appl. Cryst.* **35**, 406-421