

Changes in flow-induced aggregated structures of shear-thickening silica suspensions

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In this report, we performed small-angle neutron scattering (SANS) under shear for the 4.93 vol% fumed hydrophobic silica particles, whose surface is chemically coated by dimethyl n-hexadecane silicone, suspended in dioxane and n-hexadecane. The wavelength was selected to be 0.7 nm. The suspensions pre-conditioned by a debubbling mixer were transferred to a quartz co-axial rheometer cell with the gap length of 1 mm. The sample to detector distances were 4 m and 12 m. The resulting scattering intensity was normalized to the isotropic scattering intensities of the respective dispersants after subtraction of the background due to the corresponding dispersants. The SANS experiments were performed under applied shear rates ranged from 0 to 600 1/s at 27 °C.

The rheological measurements of the corresponding silica suspensions were performed using a commercial rheometer of MCR-300: rheochaos response, namely irregular time variation in the stress/shear rate at a constant shear rate/stress arising from nonlinearities in the viscoelastic constitutive equations, was observed for the silica suspension in dioxane at limited shear rates, whereas such a response was not detectable for the silica suspensions in n-hexadecane during the shear rates examined. Similar rheochaos responses were observed in several soft matters, such as worm-like micelle solutions, lamellar, onion, and sponge phases of surfactants, and dense colloidal suspensions.

Figure 1 shows a double logarithmic plots of the scattering intensity ($I(q)$) as a function of the wave vector (q) for the silica suspension in dioxane. The values of $I(q)$ apparently superimpose on the same curve, irrespective of the shear rate. The respective log-log plots of $I(q)$ versus q scale lin-

early a slope of -2.0 below $q = 0.2$ 1/nm and -4.0 above this q value, where the $I(q)$ tends toward Porod's law. The former slope corresponds to the mass fractal dimension (D_m) for the aggregated objects, while the latter slope gives the surface fractal dimension (D_s) according to the relationship, $I(q) \sim q^{-(6-D_s)}$, leading to the D_s value of 2.0. The resulting D_s value is smaller than that for the hydrophobic R202 silica particles, whose surface is chemically coated by silicone oil with the degree of polymerization of 6 in trans-decalin. Moreover, the q value intersected between the slopes of -2.0 and -4.0 gives the size of the primary silica particle and leads to 11.5 nm, which is almost equivalent to the diameter of the corresponding fumed silica. However, we can notice that the magnitude of the $I(q)$ values at the shear rate ranges from 20 to 300 1/s where rheochaos response is observed, is clearly larger than that without rheochaos response by the plots of $I(q)$ as a function of $\log q$.

On the other hand, the scattering curves for the silica suspensions in n-hexadecane can be much better superimposed than those in dioxane, irrespective of the applied shear rate. This means that changes in the scattering intensities are not clearly observed when shear rate is varied. However, the difference between the scattering curves of the silica suspensions in dioxane and n-hexadecane was observed at the q values higher than 0.1 1/nm, where the former scattering intensity was clearly larger than the latter one if both scattering curves in dioxane and n-hexadecane at lower q regions were superimposed.

In conclusion, this SANS experiment shows that the rheochaos response is well correlated with the changes in the scattering curves and the fractal structures of the

silica suspensions can not be easily broken out by the applied shear rates.

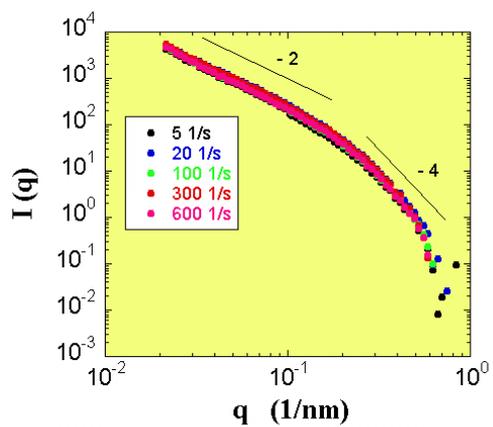


Fig. 1. SANS scattering curves of the silica suspensions in dioxane under the applied shear rates.