CROSS TOKAI **Experimental Report** J-PARC	承認日 Date of Approval 2017/12/9 承認者 Approver Jun-ichi Suzuki 提出日 Date of report 2017/11/16
実験課題番号 Project No. 2016C0004 実験課題名 Title of experiment Development of Rheo-SANS measurement on the TAIKAN instrument 実験責任者名 Name of principal investigator Hiroki Iwase 所属 Affiliation CROSS	装置責任者 Name of Instrument scientist Jun-ichi Suzuki 装置名 Name of Instrument/(BL No.) TAIKAN (BL15) 利用期間 Dates of experiments 2016/6/24 – 6/28 2017/2/2 – 2/5

1．研究成果概要（試料の名称、組成、物理的・化学的性状を明記するとともに、実験方法、利用の結果得られた主なデータ、考察、結論、図表等を記述してください。

Outline of experimental results (experimental method and results should be reported including sample information such as composition, physical and/or chemical characteristics.

We developed an apparatus for simultaneous small-angle neutron scattering (SANS) and rheological measurement, so-called "Rheo-SANS" on TAIKAN. Figure 1 shows a photograph of the Rheo-SANS experimental setup. A stress-controlled rheometer MCR302 (Anton Paar) with an inner aluminum chamber was placed on a sample stage of TAIKAN. A double-cylindrical flow-cell with an inner diameter of 48 mm and the gap of 1 mm was employed. The temperature of the samples was kept constant at 30 °C.

As a part of a demonstration measurement, we investigated wormlike micellar structures formed by gemini-type cationic surfactants composed of quaternary ammonium bromide in an aqueous solution. The gemini-type cationic surfactant (12-2-12) consisted of two hydrocarbon chains (hydrocarbon chain length of 12) and two hydrophilic groups connected by spacer chains. Sample volume fraction (ϕ) was 0.0134. For the first scan, with a logarithmically increasing shear-rate ($\dot{\gamma}$) from 30 to 10^3 s^{-1} in 90 min, both SANS and shear stress were measured. The neutron beam was irradiated to the radial position with a beam size (S_s) of ϕ10 mm. For the second scan, the neutron beam was irradiated to a tangential position with S_s = W0.5 mm × H10 mm. The shear-rate was step-wisely increased. These 2d-SANS profiles for both radial and tangential direction were obtained through the integration of a time-width of 2 min and 25 min, respectively.

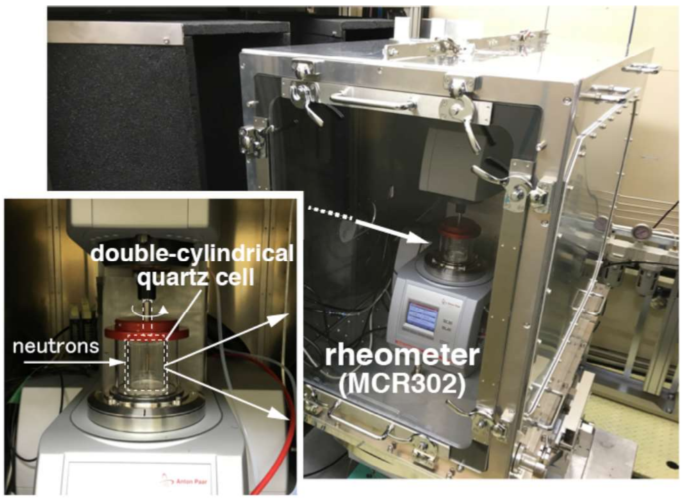

Figure 1. Photographs of rheometer (Anton Paar MCR302) installed on sample table at TAIKAN (Rheo-SANS setup).

1．研究成果概要（つづき） Outline of experimental results (continued).

Figure 2 shows the highlight of the Rheo-SANS results. Figures 2A is a two dimensional (2D) SANS profile $I(Q_x, Q_y)$ at a shear-rate of (1) $\dot{\gamma}$ = 45 s^{-1} and (2) 705 s^{-1}, respectively. The horizontal direction corresponds to the flow direction. For the lower shear-rate ($\dot{\gamma}$ = 45 s^{-1}), the 2D-SANS profiles for both radial and tangential directions were isotropic. On the other hand, for the higher shear-rate ($\dot{\gamma}$ = 705 s^{-1}), the 2D-SANS profiles were anisotropic for the radial direction, whereas isotropic for tangential one, which is due to the orientation of the wormlike micelles along the flow direction. Sector-averaged SANS profiles for horizontal (parallel) and vertical (perpendicular) directions are shown in the inset of Figure 2A. All SANS profiles for the 12-2-12 solution showed broad peak-profiles in the Q-range from 0.015 to 0.03 Å^{-1}. These peaks were attributed to the electrostatic repulsion between surface charges of the micelles. Here, the peak-intensity [$I(Q_m)$] was plotted as a function of $\dot{\gamma}$, and directly compared with the behavior of the viscosity (Figure 2B).

In the low shear-rate region ($\dot{\gamma}$ < 80 s^{-1}), the $I(Q_m)$ for both vertical and horizontal directions agreed with each other, and were no change to increasing $\dot{\gamma}$. The wormlike micelles were randomly distributed in an aqueous solution. In the $\dot{\gamma}$-range from 80 s^{-1} to 90 s^{-1}, on the other hand, the viscosity drastically grew by increasing $\dot{\gamma}$, which is a well-known behavior called "shear thickening". This time the difference of the $I(Q_m)$ for both the horizontal and vertical directions was apparent. Wormlike micelles formed by 12-2-12 were rapidly oriented in parallel to the flow direction. This is due to shear-induced elongation of the wormlike micelles in 80 s^{-1} < $\dot{\gamma}$ < 90 s^{-1}. Furthermore, when the shear-rate was further increased from 90 s^{-1} to 300 s^{-1}, the viscosity was gently increased in comparison with the behavior at the $\dot{\gamma}$ region of $\dot{\gamma}$ ~ 80–90 s^{-1}. The $I(Q_m)$ for the vertical direction increased by increasing $\dot{\gamma}$, whereas in the horizontal direction it decreased. The behavior of $I(Q_m)$ indicates that the wormlike micelles were gradually and continuously arranged parallel to the flow direction. We concluded that the micellar length had gradually become longer. In $\dot{\gamma}$ > 300 s^{-1}, the decrease in viscosity was observed (so-called "shear thinning"), and the $I(Q_m)$ for both directions were constant with an increase in $\dot{\gamma}$, suggesting that a highly oriented state of the wormlike micelles was maintained. To determine the structural parameters of wormlike micelles, a model-fitting analysis is now in progress.

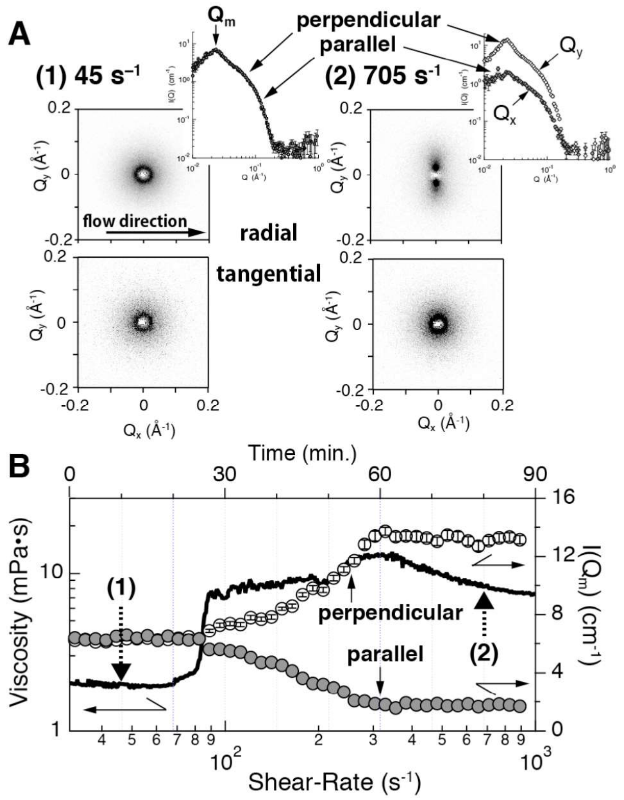

Figure 2. Rheo-SANS results of gemini-type cationic surfactant (12-2-12) solutions; A) SANS 2D counter map for shear-rate of (1) 45 s^{-1} and (2) 705 s^{-1}, respectively. Insets are sector-averaged SANS profiles. B) Shear-rate dependence of viscosity (solid lines) and peak-intensity I(Q_m) observed in the sector-averaged SANS intensities in the perpendicular (open circles) and parallel (filled circles) directions, respectively.

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