Supporting information file for

Neutron-Reflectometry-Based *In Situ* Structural Analysis of an Aligning Agent Additive for the Alignment of Nematic Liquid Crystals on Solid Substrates

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S1. Effect of UV/ozone cleaning

A pair of a glass slide and a silica substrate were irradiated with ozone for 20 min using an ultraviolet (UV)/ozone cleaner (Ebara Jitsugyo Co. Ltd., EKBIO-1100). The contact angle of water on the substrate was measured using a contact angle meter PCA-11 (Kyowa Interface Science Co., Ltd). The used sample was 5CB mixed with h-CTAB ($\varphi = 1 \times 10^{-3}$).

Fig. S1 is the time dependence of contact angle for a glass slide and a silica substrate in the air after UV/ozone irradiation. After about 200 min, the hydrophilicity was lost. Hence, all samples in the main texts were introduced in slide glass or NR cells within 120 min after UV irradiation.

S2. Langmuir model and surface tension

Langmuir adsorption model is expressed as

$$\frac{\Gamma}{\Gamma_{max}} = \frac{K\varphi}{1 + K\varphi},$$

where Γ is the areal density of surfactant, Γ_{max} the maximum adsorbed density, $K = k_{adsorb} / k_{desorp}$ the adsorption-equilibrium constant, φ the fraction of surfactant in solution[55,56]. It is equivalent to

$$\frac{\Gamma_{max}}{\Gamma} = \frac{1}{K\varphi} + 1$$

Fig. S2 shows the Langmuir plot of obtained data by the NR measurement. The vertical axis is the inverse of areal density Γ and the horizontal axis is the inverse of d-CTAB fraction φ. We assumed that $\Gamma_{max} = 2.6$ nm^{-1} = constant. The apparent $K = 3.2 \times 10^5 \pm 2 \times 10^4$. If the Langmuir-isotherm process dominates the adsorption process, the data should obey linear relations. It indicates the Langmuir adsorption model did not reproduce the adsorption process in our experiment.
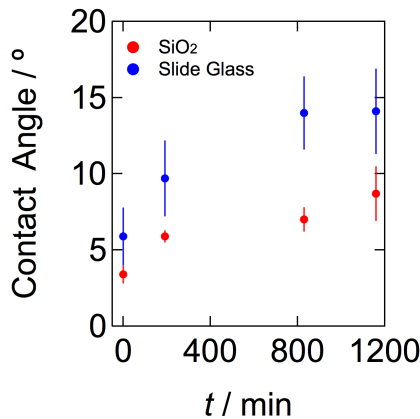
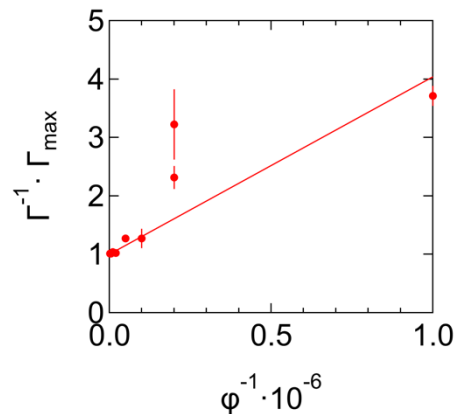Fig. S1. Time dependence of static contact angle of water on substrates in air after UV/ozone cleaning.

Fig. S2. Langmuir plot for the obtained data.