Mechanism Underlying Bioinertness of Self-assembled Monolayers of Oligo(ethyleneglycol)-terminated Alkanethiols on Gold: Protein Adsorption, Platelet Adhesion, and Surface Forces

(Supporting Information)

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Evaluation of packing densities of thiol molecules on an Au(111) surface

The packing densities of thiol molecules relative to that of *n*-alkanethiols were evaluated based on the method by Harder et al.[1] We used an angular-resolved XPS system (ThetaProbe, Thermo), and the emission angle from 17 to 27 degree from the surface normal was used for the analysis. In this method, the attenuation length of Au4f photoelectrons by methylene moieties was estimated evaluated from the results of the C4 (1-butanethiol), C8 (1-octanethiol), C10 (1-decanethiol), and C12 (1-dodecane) SAMs. Then, the 'effective' thicknesses of the other SAMs were calculated and then converted to their densities.

SEM images of Self-assembled monolayers after the platelet adhesion experiments [Figure 1s (a), (b), and (c)]

SEM images of three SAMs (OH, C8, and EG3-OH) adhered by platelets are presented. The degree of platelet activation on the C8 SAM is significant compared with that on the OH-SAM. The EG3-OH SAMs deterred most of platelets from the adhesion.

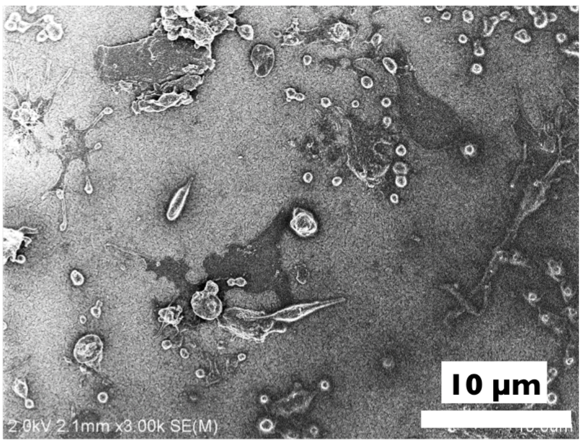

Figure 1s(a) OH-SAM

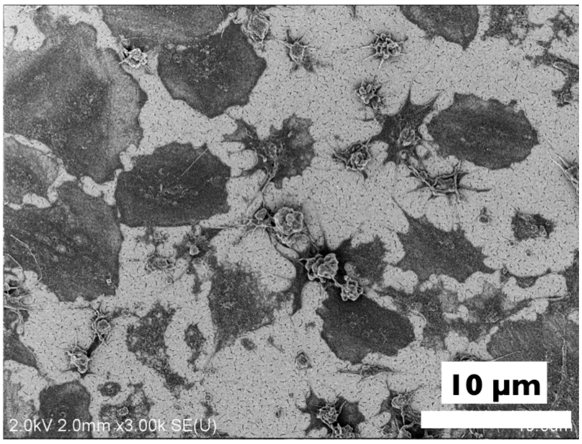

Figure 1s(b) C8-SAM

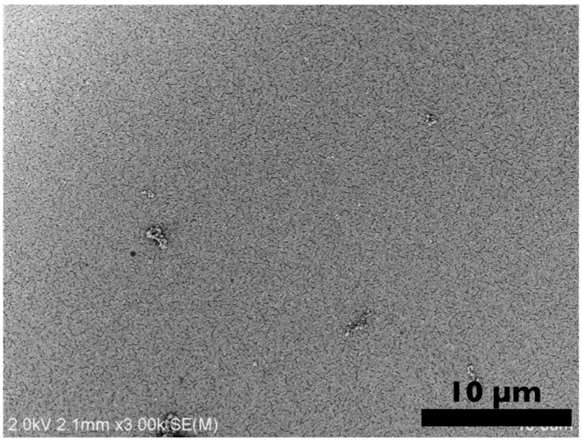

Figure 1s(c) EG3-OH SAM

Interaction of the EG3-OH SAM (substrate) with NH_2-SAM (probe) in pure water [Figure 2s]

The interaction between EG3-OH and positively-charged NH_2 SAMs in pure water was measured to elucidate the origin of the interfacial charges. This result clearly shows an attraction between them, indicating that negative ions (most probably hydroxide ions) are concentrated in the vicinity of the EG3-OH SAM.

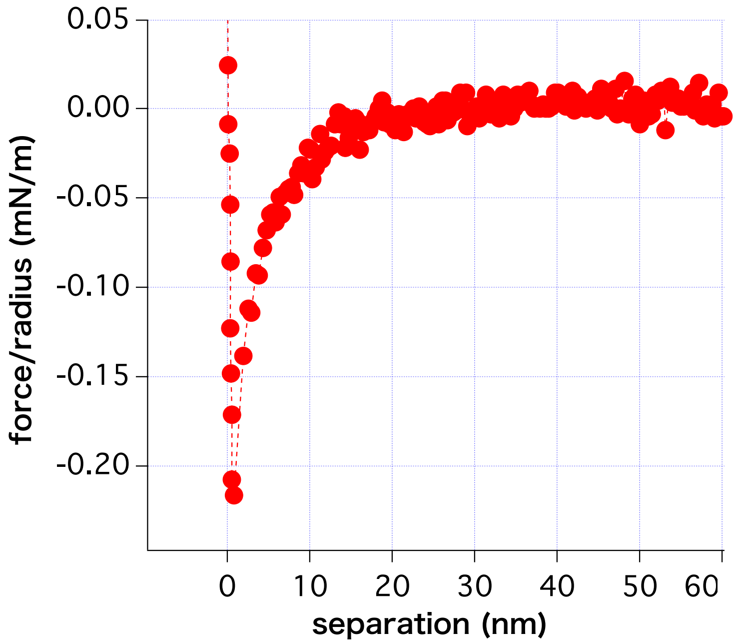

Figure 2s Force-separation curves recorded on the approach of the NH_2 SAM (probe) to the EG3-OH SAMs (substrate) in pure water

Interaction between the EG3-OH SAMs at different concentrations of NaCl

Interaction between the EG3-OH SAMs was measured in PBS buffer, in which the total concentrations of NaCl in solution was increased up to 0.4 and 0.6 M. We did not observe noticeable change in the interactions, indicating that the repulsion has no electrostatic character. As shown in figure, in addition, the range of the repulsion is much longer than theoretically-predicted electrostatic repulsion.

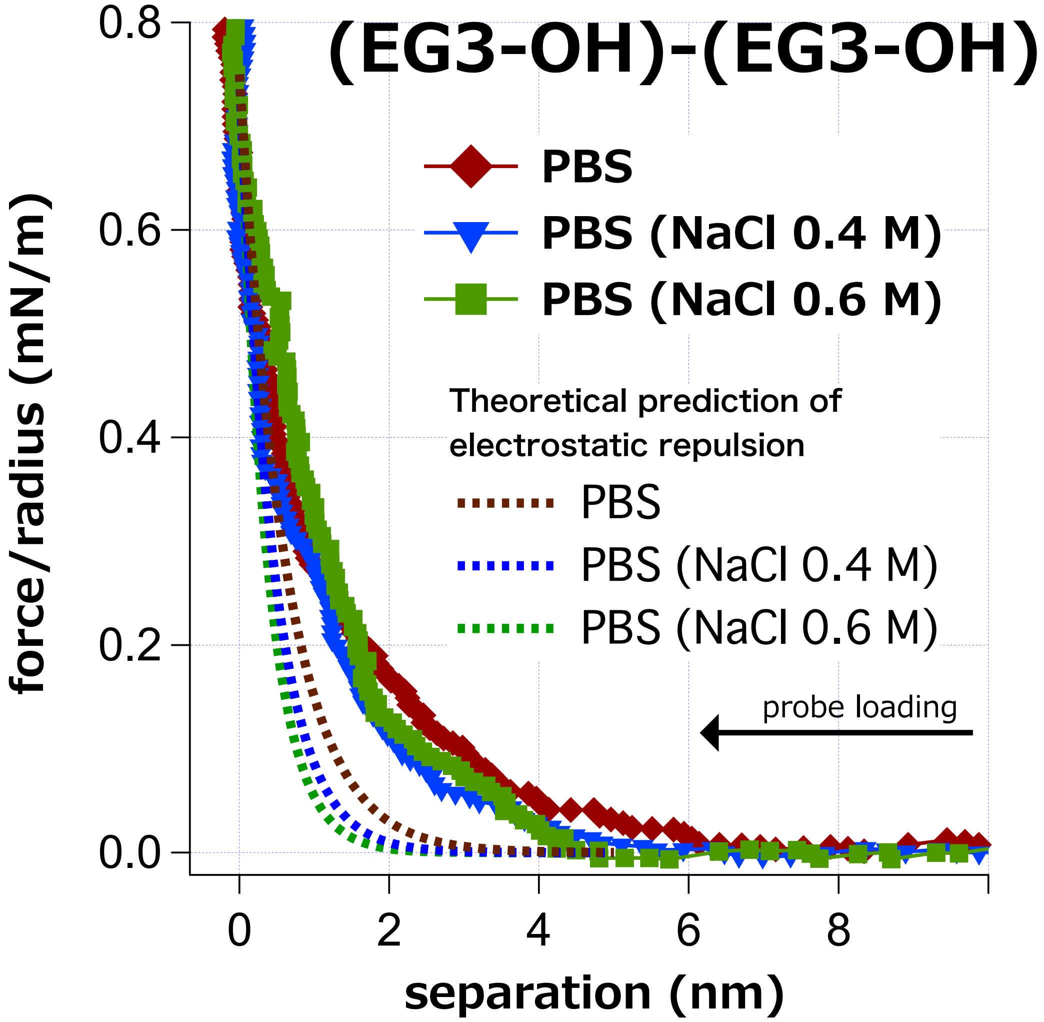

Figure 3s Force separation curves between the EG3-OH SAMs measured at different concentrations of NaCl and electrostatic repulsion calculated based on the DLVO theory

Interaction of the EG3-OH SAM (substrate) with NH_2-SAM (probe) in solution in various pH [Figure 4s]

The interaction of the EG3-OH SAM (substrate) with NH_2-SAM (probe) was measured in solution in various pH values (3.6 to 10.8) to find the pH values, at which the electrostatic interaction is attractive. The pH values were adjusted by mixing HCl (1 mM) and NaOH (1 mM). We found that the electrostatic interaction is attractive in pH of 4.4 and 5.8.

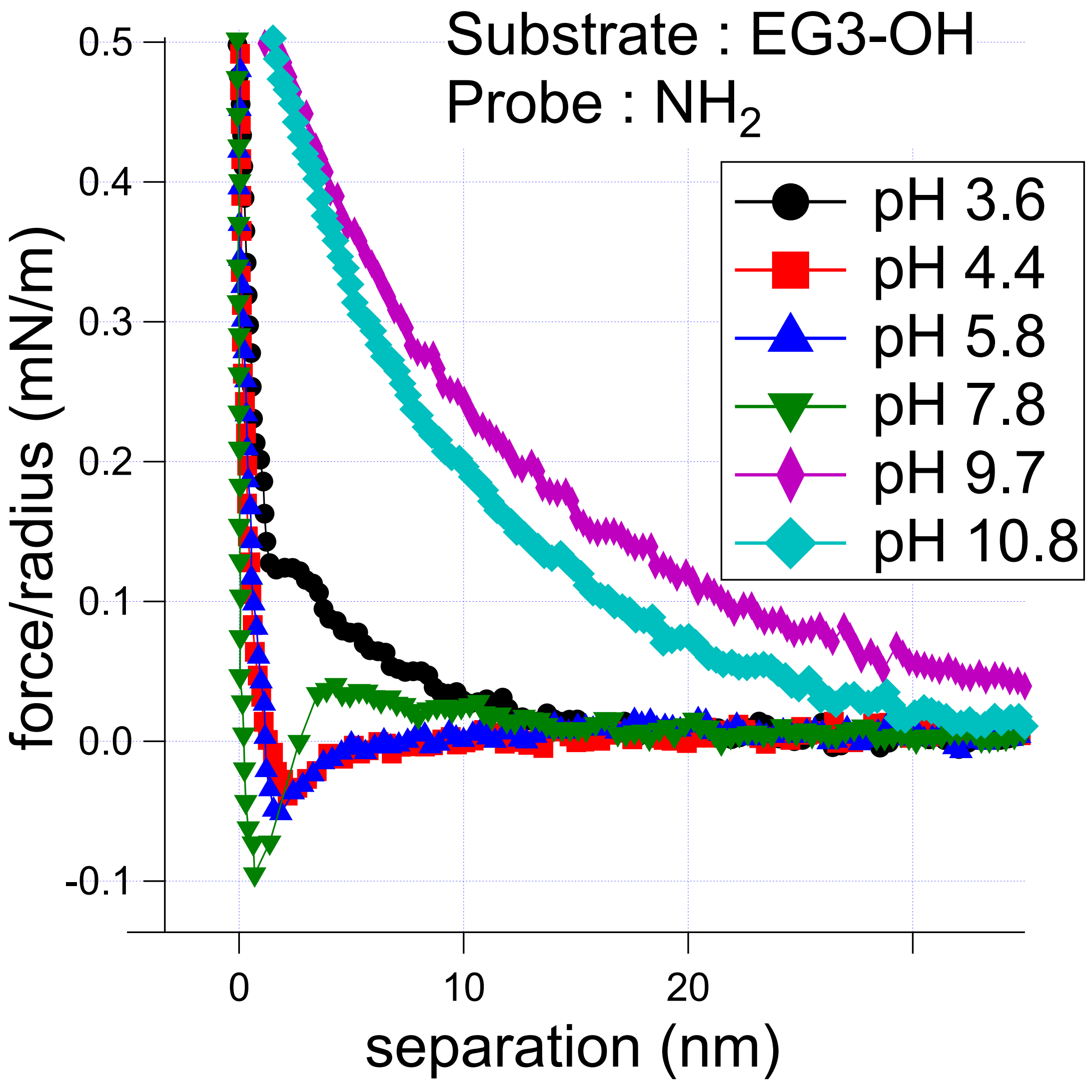

Figure 4s Force-separation curves recorded on the approach of the NH_2 SAM (probe) to the EG3-OH SAMs (substrate) in pure water

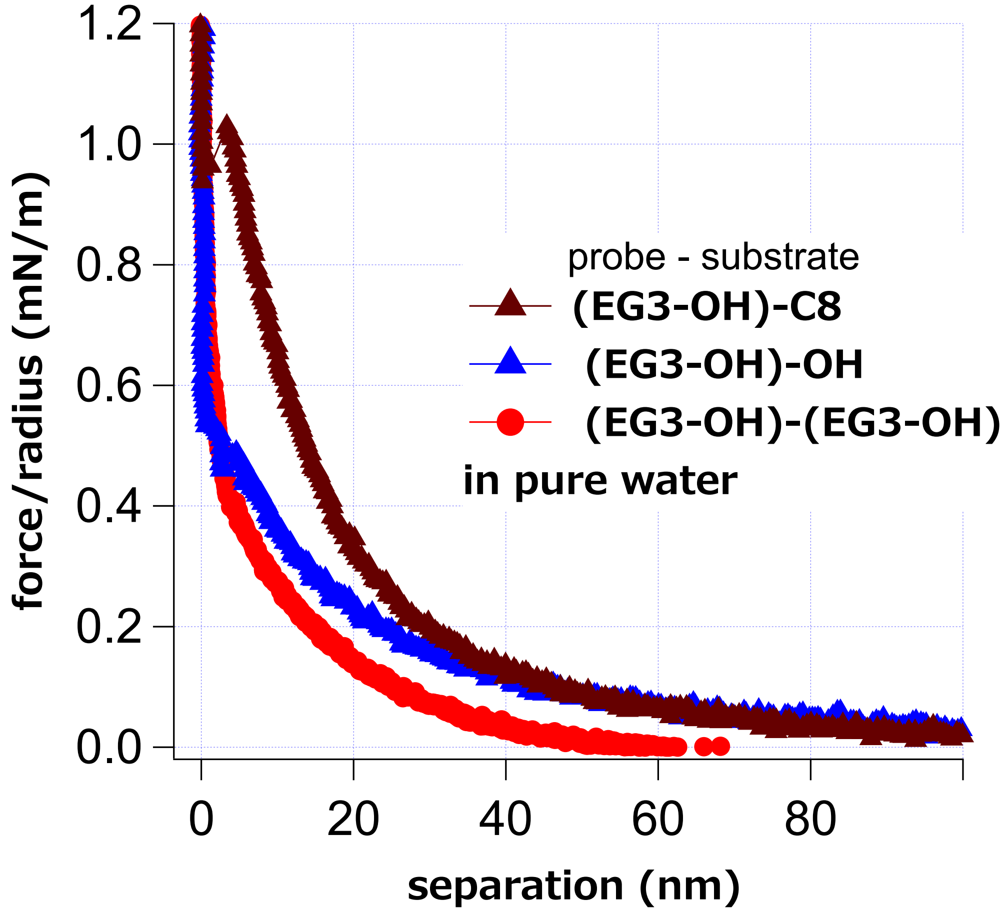

Figure 5s Force vs distance curves recorded on the approach of the EG3-OH SAM (probe) to various neutral SAMs (substrate) with different water wettabilities in pure water. Through this experiment, the same probe was used and only the substrates were changed.

References

1 P. Harder, M. Grunze, R. Dahint, G. M. Whitesides and P. E. Laibinis, *J. Phys. Chem. B*, 1998, **102**, 426-436.