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Structure of muscovite/aqueous NaCl solution interface studied by x-ray CTR scattering measurements

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Introduction

Mineral/liquid interfaces are ubiquitous in the earth's crust and control the many important properties in geochemistry and geophysics such as the partitioning of elements, the swelling and dispersion of clay minerals, and the frictional strength of rocks at fault zones. Structures of mineral/liquid interfaces have a strong correlation with these properties and help the fundamental understanding of these properties. Here we investigated the electron density profiles at the muscovite/NaCl solution interface using the surface x-ray scattering method.

Experiments

Surface of a mineral gives rises to a rod-shaped x-ray scattering perpendicular to the surface, which is called crystal truncation rod (CTR) scattering. The shape of the intensity around the Bragg reflection points of the mineral is proportional to the Fourier transform of the electron density distribution around the surface. Therefore the interfacial electron density distributions can be studied by the analysis of the CTR profile.

The surface x-ray scattering experiments was made at the BL-4C of the Photon Factory, KEK, Japan with monochromated x ray of 11.0 keV. The CTR refers to scattering with the momentum transfer vector Q perpendicular to the (00l) surface of muscovite. The CTR profile was obtained in the range Q = 0.59–4.38 Å^{-1} with individual rocking scans at given Q values.

We measured the muscovite/pure water interface to confirm the validity of our experiments by comparing the results with those of Cheng et al. (2001) [1]. After that we measured the muscovite/NaCl solution interface.

The electron density profile of muscovite/NaCl solution interface can be obtained by comparing the experimental and calculated CTR profiles which was the Fourier transform of an atomistic model structure. The model structure includes a fixed semi-infinitesimal bulk muscovite structure, a relaxed surface structure, adsorbed layers of ions and water molecules, and bulk water above the interface. The electron density of adsorbed layers and bulk water were represented by the superposition of Gaussian distributions. The fitting was performed by a least-squared method using modified ANA-ROD program [2].

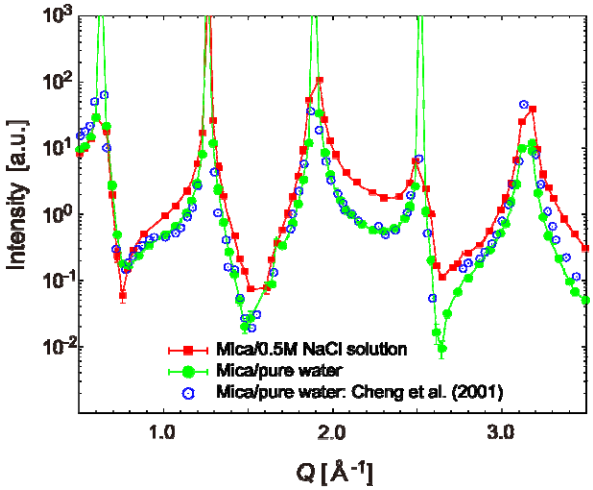

Fig. 1 The x-ray CTR profiles of mica/pure water and mica/0.5M NaCl solution interfaces. The profile of mica/pure water interface measured by Cheng et al. (2001) was plotted as open circles for a comparison.

Results

We confirmed that the CTR scattering profile of the muscovite/pure water interface corresponded with that by Cheng et al. (2001) [1] as shown in Fig. 1. This means that our experimental setup can be applied to measure the CTR scattering profile of the muscovite/aqueous NaCl solution interface.

The CTR profile of muscovite/0.5 M aqueous NaCl solution interface was also shown in Fig.1. The shape was different from that of muscovite/pure water interface. The high intensities of CTR profile at $Q \approx$ 0.6, 1.3, 1.9, 2.5, 3.1, and 3.8 Å^{-1} correspond to the Bragg reflection points of muscovite depicted by the (00l) (l = 2, 4, 6, 8, 10, and 12, respectively) plane. The shape of the CTR profile between the Bragg points gives the information about the structure of the muscovite/NaCl solution interface.

The best-fit model for the electron density profile of muscovite/NaCl solution interface could reveal the real structure of the electric double layer on a muscovite surface.

References

[1] L. Cheng et al., Phys. Rev. Lett. 87, 156103 (2001).
[2] Original ANA-ROD program is available from the web site of ESRF SciSoft (http://www.esrf.eu/computing /scientific/joint_projects/ANA-ROD/).

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