Phase Behavior of Synthetic Clay Gels

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Introduction

Layered clay minerals suspended in salt solutions take the form of charged platelets, which interact electrostatically to produce rich concentration dependent phase behavior. Structures ranging from suspended solid aggregates to isotropic gels to liquid crystalline ordering have been observed. Many interesting open questions remain in systems like these: the local, orientational and long range ordering, the effect of surface charge and water content on the structure, and the dynamics of the gels' formation are largely unknown.

Methods and Materials

We have studied Na fluorohectorite, a synthetic layered clay that exhibits striking gravity induced phase separation. The present samples (2.9 wt% Na-fh, 6×10^{-4} M NaCl) exhibit what appears to the eye as two distinct phases, with a layer of dense "sediment" lying below a translucent gel phase. The samples were studied by transmission of 19 keV x-rays through glass tubes, with the momentum transfer normal to the vertical layering axis (Fig. a: curves are shifted for clarity).

Results

The top curves show the background scattering from the empty ($\cdots$) and water-filled (- - -) glass tube. Scattered intensity from the gel phase (– –) reveals a peak at 0.42 Å^{-1}, corresponding to the (001) reflection from crystallites having two molecular water layers intercalated within the microscopic unit cell. Scattering through the thicker sediment at the base is characterized by more intense and numerous Bragg peaks. The background scattering at $q \approx 2$ Å^{-1} shows that bulk water is mostly excluded from this region, unlike the case of the gel above. It is not clear whether the pronounced difference between the gel and sediment is due entirely to the difference in the overall density, or whether siginifcant changes in orientation of the platelets occur.

Discussion

These measurements have the potential to reveal considerable information about the structure and dynamics of the gel phase. Scattered intensity from the (001) peak (Fig. b) as a function of height in the tube shows the boundary between the solid and gel phases evolving with time as the particles settle. The oscillations on millimeter length scales in the gel phase are due to density inhomogenieties that are static over periods as long as several hours. The data also suggest the existence of a boundary at a height of about 40 mm between a more uniform area in the upper part of the gel, and a more inhomogeneous one below, that persists as the particles settle.

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