

## **Pulsed field gradient proton NMR study of the self-diffusion of H<sub>2</sub>O in montmorillonite gel: Effects of temperature and water fraction**

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### **ABSTRACT**

Self-diffusion coefficients of water molecules (H<sub>2</sub>O) in Na-montmorillonite gel were measured as a function of water fraction (54.8 to 100 wt%) and temperature (30.4 to 60.0 °C) using proton nuclear magnetic resonance (NMR). Spin-echo pulse sequences with magnetic field gradient pulses for the diffusion measurement were applied to the montmorillonite gel at the Larmor frequency of 20 MHz. The self-diffusion coefficient,  $D$ , of H<sub>2</sub>O in the clay gel is expressed phenomenologically by  $\ln(D/D_0) = 1.77\{\exp[0.0798(w - 100)] - 1\}$ , where  $D_0$  is the water diffusivity in bulk water and  $w$  is the water fraction of the gel (wt%). The data for  $w > 84.7$  wt% can be explained by a theoretical diffusion model for the randomly distributed clay grains. The activation energy of the water diffusivity in the montmorillonite gel was nearly equal to that in bulk water, so the normalized diffusivity,  $D/D_0$ , obeys the temperature-independent master curve. The transition from the free diffusion to the restricted diffusion was not observed for gradient pulse intervals ranging from 5 to 120 ms. This indicates that the average pore size of the gel is much smaller than a few tens of micrometers, so the random walk trajectory of water molecules in the gel is geometrically restricted by the packing of clay mineral grains. Water diffusivity higher than that of the present NMR study was found by computer simulations and neutron scattering experiments in which effects of bound water were considered but those of the tortuosity of the grain packing were neglected. Thus, the predominant factor controlling the diffusivity in the NMR experiments is not the bound water near the clay surface but the geometrical tortuosity of the packing of clay mineral grains.