

RAMAN SPECTROSCOPY STUDY OF VAPOR-LIQUID  
EQUILIBRIA IN H<sub>2</sub>-H<sub>2</sub>O-NaCl SYSTEM

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Hydrogen is important natural volatile controlling redox conditions in the crust. Moreover, it is one of the new perspective sources of energy. However, our knowledge of water-hydrogen solubilities and phase relations at high T-P-xH<sub>2</sub> remains poor. The aim of this study is to provide in situ Raman spectroscopy data for H<sub>2</sub>-H<sub>2</sub>O-NaCl system in a wide T-P-mNaCl range (25-400°C, 1-1500 bar, 0-3.0 mol/kg). The experiments have been performed in fused pure silica capillary (Chou et al., 2005; Lu et al., 2006) coupled with a stainless steel pressure line and Linkam heating stage. The observation of vapor-liquid immiscibility and collection of the Raman spectra of each phase have been performed with optical microscope of the Raman microprobe (Labram HR, ® Horiba). These new measurements were complemented by in situ H<sub>2</sub> fugacity measurements in Ti-alloy autoclave equipped with H<sub>2</sub>-permeable membrane at 300-350°C and 100-170 bar.

Composition of vapor and liquid phases obtained from our Raman measurements are in good agreement with data of Seward and Franck (1981). At high pressures, we detected additional O-H (in vapor) and H-H (in liquid) vibrations, not present in the spectra of pure water or pure hydrogen at the same T-P. These new peaks are shifted to lower frequencies. Thus, we suppose the assignments of these peaks to H<sub>2</sub>O-H<sub>2</sub> cluster formation at high P. According to our new experimental data the temperature effect on the hydrogen salting-out coefficients is much weaker than previously considered (Kertes, 1981). In order to describe H<sub>2</sub>-H<sub>2</sub>O-NaCl system, the new equation of state was developed.

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Chou et al. (2005) *Advances in High-Pressure Technology for Geophysical Applications*, 475, Elsevier, Amsterdam; Kertes (1981) *IUPAC Solubility Series*, Pergamon Press, Oxford; Lu et al., (2006) *Appl. Spectroscopy* 60, 122; Seward & Franck (1981) *Ber. Bunsenges. Phys. Chem.* 85, 2.