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High Pressure Studies on Methane-Ethane System and Prediction of their States in the Titan's Interior

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Saturn's satellite Titan was explored by Huygens Probe in January 2005. According to the reports, Titan's surface is covered by water ice and solid hydrocarbons. No oceans or lakes were found on the surface. Some evidences, however, for once flowing liquid hydrocarbons are shown by dry river beds or offshore structures. Titan's atmosphere consists of nitrogen, methane, hydrogen, ethane, argon, and so on. According to the theoretical calculations, methane and ethane on the surface were exhausted by photolysis within 10ˆ7 years. So a certain large 'Reservoir', which supplies methane and ethane is expected to exist in the Titan's interior. However, knowledge of their states of the methane-ethane system under high pressure has been limited so far.

In this study, high pressure experiments of methane-ethane system were performed in the pressure range of 0.1 MPa to 20.4 GPa at room temperature by using diamond anvil cell. Initial methane/ethane ratio of two samples is 90% / 10% and 85% / 15%. Single component of ethane was also examined in the pressure range from 0.1 MPa to 15.7 GPa at room temperature.

Fluid ethane crystallized at 2.9 GPa. Pressure-induced solid ethane was observed for the first time in this study. At crystallization, additional two C-H vibration modes and one C-C vibration mode appeared. In-situ X-ray diffractometry revealed phase changes as follows. The diffraction patterns obtained were almost same, but a few new peaks appeared at approximately 8.0 GPa. The X-ray diffraction patterns under 8.0 GPa were indexed as an orthorhombic system, and the lattice parameter of the phase I at 7.0 GPa is 5.043 A, b= 4.998 A, c= 3.661 A. The patterns above 8.0 GPa were indexed as a monoclinic system. In addition, the slopes of d-value with pressure discontinuously changed at 12 GPa and 14 GPa. Raman spectroscopy also revealed clear changes in slopes of Raman shift with pressure at 8 GPa, 12 GPa and 14 GPa. These phases of solid ethane were called 'phase I', 'phase II', 'phase III' and 'phase IV' in this study. The ethane molecures in these phases were inferred to show only different orientations in the common fundamental structure.

In the methane-ethane mixtured system, the mixed fluid kept up to 5.3 GPa and finally crystallized at that pressure. And an unknown phase appeared at the crystallization, and it was kept up to 10.8 GPa. This phase was called 'phase X' in this study. This phase consists of methane and ethane, but it is completely different from neither single solid methane nor single solid ethane according to the X-ray diffraction patterns and the Raman spectra. The XRD patterns of the phase X were indexed as an orthorhombic system, and the lattice parameters are a=8.142 A, b=7.970 A, c=2.990 A at 6.4 GPa. The intramolecular vibration modes of ethane molecular in the phase X show lower Raman shift by 20 to 30 cmˆ-1 than those in the single solid ethane, which indicates that larger interaction occurs among the molecules. Therefore, the phase was thought to be a van der Waals compound formed under high pressure in methane-ethane system.

The present results suggest that some fluid of methane-ethane mixtures could have existed or could exist at Titan's shallow or middle mantle. Though P-T conditions of Titan's interior has not been clear at present, the van der Waals compound of methane-ethane is expected to exist at the deeper mantle. In any case, ethane content may have a great deal of effect to the properties of the Titan's interior.