Phase changes of Methane-Ethane System Under High Pressure and existence of a van der Waals Compound

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High pressure experiments of methane-ethane system were performed in the pressure range from 0.1 GPa to 20.1 GPa at room temperature by using diamond anvil cell (DAC). Five compositions with ethane mole % of 3 mol%, 10 mol%, 15 mol%, 25 mol% and 50mol % were examined. Characterizations were made by in situ optical microscopy, powder X-ray diffractometry (XRD) and Raman spectroscopy. After filling the sample into the DAC, the samples of four compositions (3 mol%, 10 mol%, 15 mol% and 25 mol%) exhibited homogeneous fluid. With increasing pressure, the homogeneous fluid separated into two fluids. Above 5.3 GPa, two solid phases appeared, one is fine acicular-shaped crystallites and the other is large platy crystal. X-ray diffraction patterns showed existence of solid methane and an unknown phase. Raman spectroscopy showed that the platy crystal was composed only of methane and that the acicular crystallites were composed of methane molecules and ethane molecules. These results suggested that the acicular crystal was a van der Waals Compound in methane-ethane system. The XRD patterns of the samples, 25 mol% and 50mol%, showed solid ethane and the same van der Waals Compound. The results suggested that there is only one vdWC in methane-etnae system, of which composition is approximately 15 to 25ethane mol%.

The XRD pattern of a pressure-induced solid ethane was indexed as a tetragonal system, with the lattice parameters of a=5.028(2) A, c=3.654(3)A at 7.0 GPa. This structure can be related to the temperature-induced solid ethane previously reported.