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Vapour-Liquid Equilibrium Study of Silicon Tetrachloride-CO_2 System

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Supercritical CO_2 (critical P and T are 73.8 bar and 31.1 ºC respectively) occurs as fluid inclusions over a wide range of conditions from greenschist facies to granulite facies. Whether it can transport inorganic components at elevated P-T conditions has not been considered in the geological literature. Numerous studies have been conducted on vapour-liquid equilibrium of various organic compounds and carbon dioxide; however, there is very little published work on phase equilibria of binary systems involving inorganic compounds and CO_2, particularly metal and non-metal chlorides like silicon tetrachloride ($SiCl_4$). Experimental results are presented for phase equilibria in the binary system carbon dioxide-silicon tetrachloride at 323, 348 and 373 K and pressures up to 140 bar. The aim of these experiments is to gain insight into the possible role of supercritical CO_2 as a transport and reactive medium under certain conditions in the earth's crust.

One of the most important aspects of this study is the experimental design, which allows sampling of both liquid and vapour phases with minimal disturbance to the equilibrium of the system. It involves use of a high temperature, 8-port switching valve equipped with sampling loop which is connected to a highly sensitive thermal conductivity detector (TCD). The carrier gas (CO_2), following purification, continues to flow through the detector at a steady, low flow rate (monitored by a digital flowmeter), and a small amount (~20 microlitres) of the sample (mixture of $SiCl_4$ and CO_2; either from vapour or from liquid phase) from the pressure vessel is injected into the CO_2 stream. The difference in thermal conductivity of the sample compared to CO_2, detected by the TCD, is proportional to the amount of dissolved $SiCl_4$.

The binary system exhibits a two-phase equilibrium region extending up to approximately 160 bar. A modified Peng-Robinson equation of state was successfully applied to model the experimental data and also to calculate the 423 K isotherm for this system.