Neutron Diffraction Measurements on CO_2 in Both Undercritical and Supercritical States

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Neutron diffraction measurements on fluid CO_2 in a wide thermodynamic range covering both undercritical and supercritical states are presented. From these measurements, performed using 7C2 spectrometer at the Orpheé reactor, we have derived, for each investigated thermodynamic state, the total radial distribution functions thus gaining information on the microscopic structure of the fluid. The undercritical states are taken along the liquid side of the entire coexistence curve while supercritical states have been studied along three isotherms (313 K, 373 K and 473 K) with density ranging from 8.7 to 14.0 molecules/nm^3. In particular we have been able to follow two isochores (11.4 and 12.9 molecules/nm^3) starting from the coexistence line up to a supercritical temperature. We completed our program performing Molecular Dynamics simulations, at the same thermodynamic states, using several intermolecular potentials. The main results, valid for all the investigated densities and temperatures, are that:

1. Fluid CO_2 shows very relevant orientational correlations among first neighbor molecules; such correlations become more relaxed as the temperature increases although they are still quite evident up to 1.6 times the critical temperature.
2. T shaped orientations are found to be largely favored.
3. The explicit inclusion of electric quadrupole interactions in the intermolecular pair potential is essential in order to obtain agreement between Molecular Dynamics simulations and experimental results at least as far as the microscopic structure is concerned.