

Structure determination of high-pressure C_{70} phases through a joint XRD/DFT study

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Extended polymerization in ABC-stacked C_{70} fullerite at high pressure and high temperature (HP-HT) had been considered impossible to achieve, in opposite to C_{60} fullerite. Reasons invoked to explain such behavior ranged from the low molecular symmetry of C_{70} to its low chemical reactivity, when compared to C_{60} . Recently, however, we have shown that, indeed, C_{70} can form extended polymerized structures.

Two novel C_{70} phases have been discovered at 10GPa-270°C and 7GPa-600°C.^{1,2,3} Their crystal structures were determined through a combined experimental-theoretical effort. Samples recovered from HP-HT treatments display X-ray powder patterns with poor resolution, indicating a considerable degree of crystalline disorder. Thus, the detailed crystal structures could not be determined from X-ray crystallography analysis of the powder diffraction data, although important structural information, such as, lattice constants and molecular orientations compatible with the overall observed symmetry, were retrieved. Density functional theory (DFT) modeling, in particular crystal structure optimization with constrained lattice constants and molecular orientations, was performed. These simulations have showed that the new phases involve extended polymerization of C_{70} molecules. The first structure consists of one-dimensional (1D) zig-zag polymer, while the second structure involves buckled-hexagon two-dimensional (2D) polymerized planes (see fig.1). A structural relationship between these two structures can be established, the second structure being obtained from the first one through additional bonding.³

Higher-level DFT modeling, employing GGA-type functionals and PAW pseudo-potentials, was performed just recently. Full structure optimizations, with non-constrained parameters, give a good agreement with previous experimentally-constrained calculations.⁴

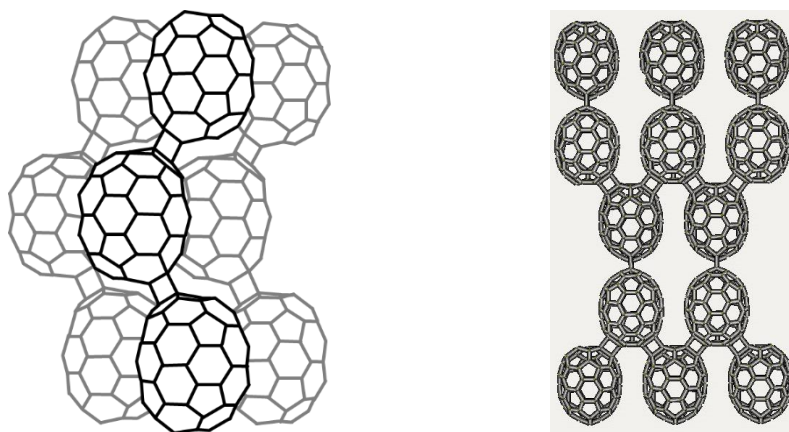


Figure 1: High-pressure C_{70} structures

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