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Synthesis of α-cristobalite-type CO_2-SiO_2 under extreme conditions

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Extreme conditions change the behavior and reactivity of elements and compounds and permit the synthesis of novel materials. In the case of group IV oxides, molecular CO_2 and a network solid silica, which were considered to be incompatible, are found to react under HP-HT conditions. A crystalline CO_2-SiO_2 solid solution was synthesized from molecular CO_2 and microporous silicalite SiO_2 at 16-22 GPa and temperatures above 4000 K in a laser heated diamond anvil cell [1]. Synchrotron X-ray diffraction data show that the crystal adopts a densely packed α-cristobalite structure (space group $P4_12_12$) with carbon and silicon in 4-fold coordination. This occurs at pressures at which SiO_2 normally adopts a 6-fold coordinated rutile-type stishovite structure. The P-T conditions used in this study represent a compromise between the respective stabilities of 3- and 4-fold coordination in CO_2 and 4- and 6-fold coordination in SiO_2. This solid solution can be recovered at ambient pressure at which the unit cell volume is 26% lower than that of α-cristobalite SiO_2. This is due to the incorporation of much smaller carbon atoms, resulting in the collapse of the oxygen sublattice. The unit cell volume and the different C and Si sites identified in Raman spectroscopy are consistent with a C:Si ratio of 6(1):4(1). The tetragonal c/a ratio increases from 1.283 at 16 GPa to 1.303 at ambient pressure and is lower than that of SiO_2 due to the more compact structure of the new material and essentially corresponds to that of the dense rutile-type oxygen sublattice. This can explain the small variation in volume observed for this phase corresponding to a bulk modulus of about 240 GPa. Due to the incorporation of silicon atoms, this hard solid based on CO_4 tetrahedra can be retained as a metastable phase. This strongly modifies standard oxide chemistry and shows that carbon can enter silica giving rise to a new class of hard, light, carbon-rich oxide materials with novel physical properties.

[1] *M. Santoro, F. A. Gorelli, R. Bini et al., Nat. Commun., 2014, in press.*

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