

Abstract No. Solo0083

Equation of State and Phase Stability of Turbostratic Carbon Nitride

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Beamline(s): X17B

Introduction: Very recently, Solozhenko *et al.* [1] reported the synthesis of turbostratic carbon nitride (t-CN) with a stoichiometry close to CN by high-pressure thermal decomposition of C-N-H precursors. The present work was undertaken to characterize this new phase under high pressures and temperatures.

Methods and Materials: Experiments up to 20 GPa were performed using the T-cup, a two-stage 6–8 style high-pressure system with a large anvil press SAM85 and energy-dispersive powder x-ray diffraction. Turbostratic carbon nitride was synthesized from the $C_3N_4H_{2.5}$ precursor at 4.8 GPa and 950 K in the multianvil apparatus MAX80 according to the method described in [1].

Results: Synchrotron radiation diffraction patterns of turbostratic carbon nitride show broad diffraction lines (001), (10), (11) and (002), which are typical for the two-dimensionally ordered (turbostratic) layered structures. With increasing pressure, the diffraction lines of t-CN broaden and weaken considerably and above 9 GPa only the lines associated with the 001 , 10 and 002 reflections could be resolved. Similar to all graphite-like structures, t-CN exhibits much higher compressibility along the c -axis than along the a -axis as demonstrated by the $k_c/k_a = 85(6)$ ratio between linear compressibilities in the above directions. The shapes of the c/c_0 curves of t-CN and graphite are very similar with t-CN considerably more compressible at all pressures. Within experimental uncertainty, the a -axis compression a/a_0 is linear within the pressure range of this study. An average linear compressibility along a -axis was therefore calculated, yielding an average a -axis modulus of t-CN $\beta_0 = 1640(80)$ GPa. This value is significantly higher than that of graphite (1250 GPa [2]) and the linear incompressibility of diamond (1326 GPa [2]). This fact can be attributed to a shorter C-N interatomic distance (1.403 Å) in the layer of turbostratic carbon nitride as compared to the C-C distances in graphite (1.426 Å) and diamond (1.544 Å).

A least-squares fit to the experimental p - V data (Fig. 1) using the Birch-Murnaghan (third-order Eulerian finite-strain) equation of state gives values of the bulk modulus $B_0 = 20.1(5)$ GPa and its pressure derivative $B_0' = 8.3(2)$, with the zero-pressure unit cell volume $V_0 = 16.82(4)$ Å³. These values are comparable to $B_0 = 29.5(8)$ GPa, $B_0' = 9.9(5)$ for turbostratic graphite [3] and to $B_0 = 17.5(8)$ GPa, $B_0' = 11.4(5)$ for turbostratic boron nitride [4].

According to the results of *in-situ* studies, at 17.8 GPa t-CN started to decompose at 1850(50) K to form diamond and nitrogen. The total disappearance of carbon nitride reflections was observed at 1940 K (Fig. 2). The formation of diamond proceeds through solid-phase decomposition of carbon nitride and not via formation of the intermediate graphite phase.

Acknowledgments: This study was supported by the National Science Foundation Grant Nos. EAR0125094, DMR-009533 and an International Travel Grant to V.L.S. (NSF EAR-9909145 Supplement).

References:

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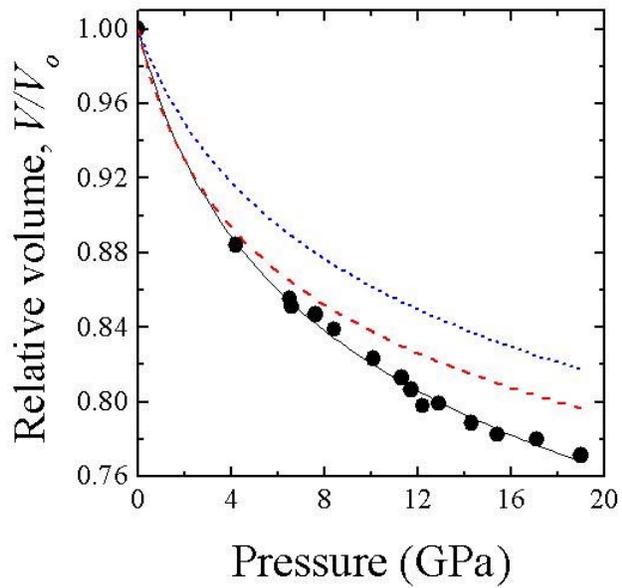


Fig. 1 The p - V data of turbostratic carbon nitride at 300 K. The solid line is the Birch-Murnaghan EOS fit to the experimental data. The dotted and dashed lines are EOS of turbostratic graphite [3] and turbostratic boron nitride [4], respectively.

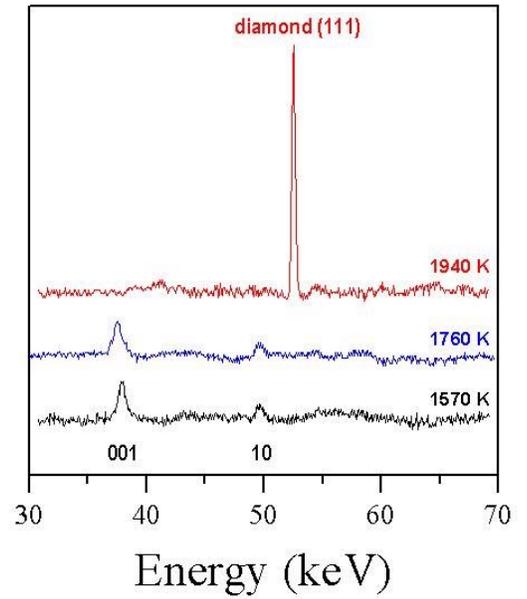


Fig. 2 *In-situ* diffraction patterns taken at heating turbostratic carbon nitride in steps at 17.8 GPa (after background subtractions). Bottom and top patterns correspond to t-CN and diamond, respectively.