Supplementary Information (SI) for Journal of Materials Chemistry A.

Supplementary Information for "Pitfalls in Parameters: Practical Process Development in Chemical Vapor Processing of SiC"

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Figure S1 is referenced in Section 3.1. Figure S1A (α=8) corresponds to the first integer reactant ratio α=H_2/MTS phase diagram that includes pure SiC as a product. The phase boundary for carbon is identified at low temperature and high pressure, running from 700 °C / 662 torr to 725 °C / 760 torr. Beyond this phase boundary, SiC is the only solid product thermodynamically calculated to form. Figure S1B (α=36) is the first phase diagram where the experimental conditions used for both CVD and CVI are expected to exclusively form SiC. The carbon phase boundary is found to run from 1100 °C / 20 torr to 1325 °C / 130 torr – above this boundary, C-SiC codeposition is expected. The initial reactant composition α=61 (Figure S1C) is the highest composition where C-SiC is expected to form at the temperatures and pressures examined in this report. The phase boundary for carbon is constrained to low pressures and high temperatures – running from 1277 °C / 20 torr to 1325 °C / 29 torr. The phase diagram in Figure S1D is the highest α (120) calculated in this study and shows only SiC as a solid product over the temperature and pressure ranges examined. No formation of solid Si was predicted from α=0.5 to 120.

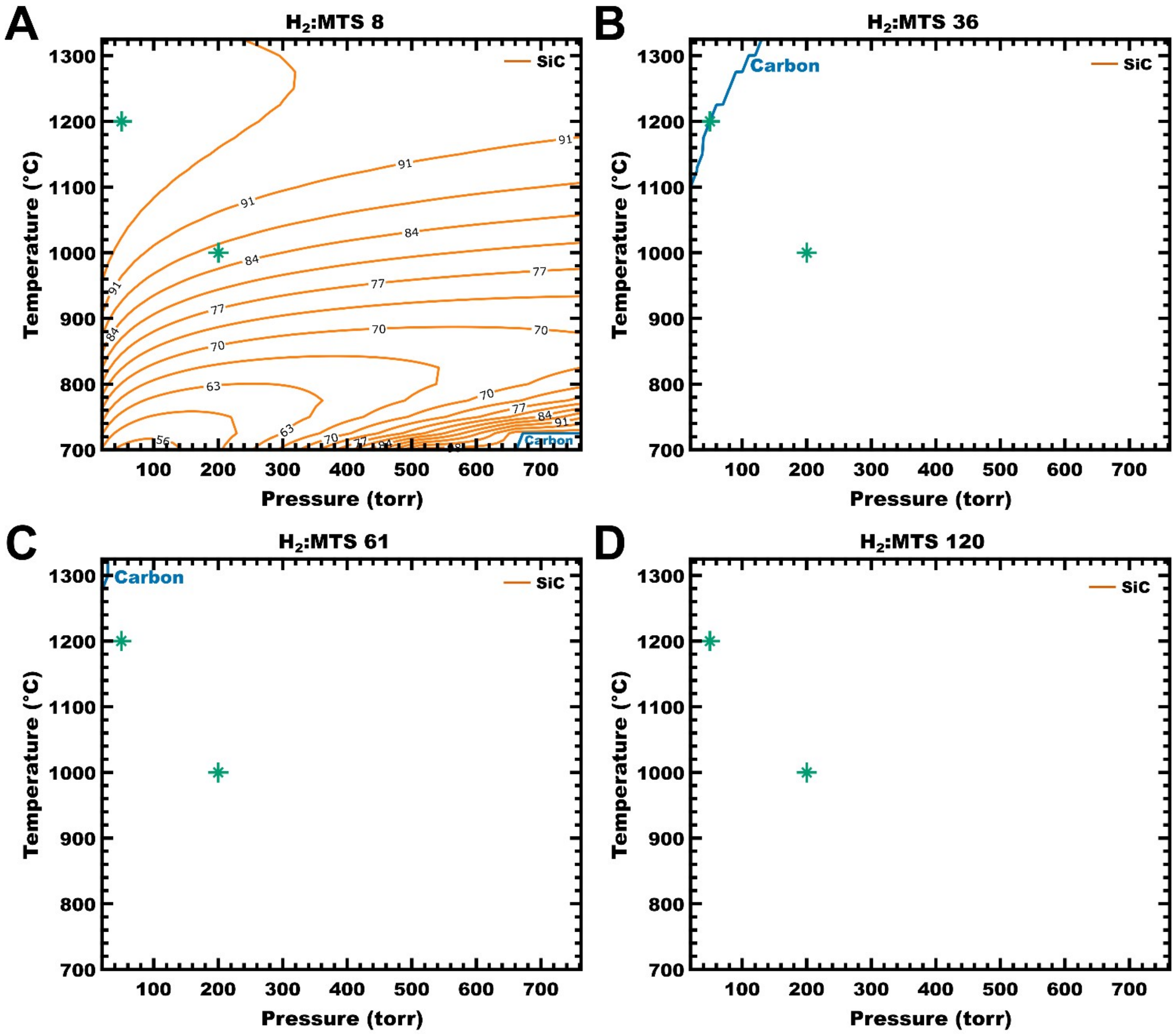

Figure S1 Calculated phase diagrams of solid products produced from $\alpha = H_2/MTS$ values of (A) 8, (B) 36, (C) 61, and (D) 120 over a range of temperatures and pressures. Orange contour lines correspond to amounts of solid products in mole-percent (mol%) of SiC (balance carbon). Green asterisks (*) mark the experimental conditions used for CVD and CVI SiC.

Figure S2 is referenced in Section 3.2. The signature peak of silicon can be seen in the B-doped Si (<100> face, Ted Pella, USA) sample at 520 cm^{-1}, as well as a small feature at ~960 cm^{-1}. The broad, low-intensity ~960 cm^{-1} stretch is a two-phonon band of the TO mode.[1, 2] When magnified, single- and multi-phonon bands are also identified centered at 302 cm^{-1}.[1, 2] Figure S2 also includes the Raman spectrum of polycrystalline CVD SiC (high-resistivity grade, >99.99995% purity, Rohm & Haas – now PremaTech Advanced Ceramics, USA). The CVD SiC sample is consistent with the 3C polytype, with two sharp peaks at 796 and 972 cm^{-1}, identical to the positions reported by Nakashima, *et al.*,[3] for the TO and LO stretches of 3C-SiC, respectively. Lower-intensity, broad higher-frequency stretches centered around 172 and 560 cm^{-1} have previously been associated with Si-Si stretching in d-SiC (disordered SiC).[4, 5] At lower frequencies, broad bands centered approximately at 1527 and 1716 cm^{-1} have previously been associated with multi-phonon bands in 3C-SiC,[6, 7] instead of C-C stretches as seen in graphite (~1330 and 1580 cm^{-1}) or diamond (1332 cm^{-1}).[8, 9]

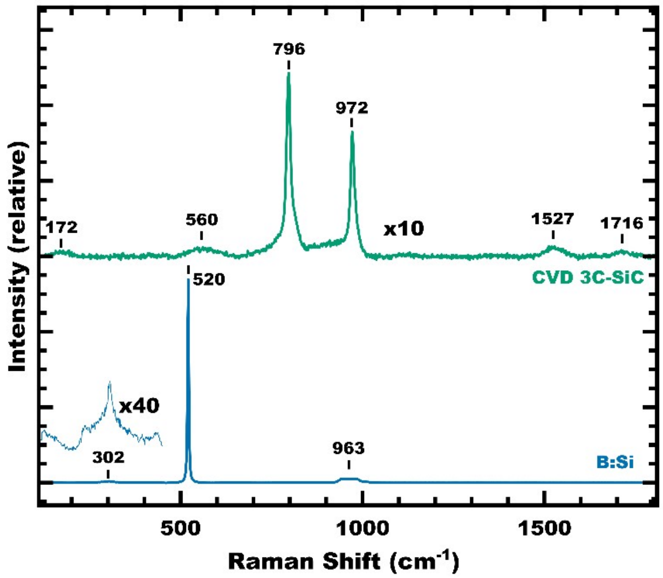

Figure S2 Raman spectra of reference samples, commercial CVD 3C-SiC and B-doped Si single crystal.

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