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Design principles and high-pressure syntheses of novel superhard materials\(^1\)
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The development of novel high-performance superhard materials, guided by reliable design theories, is highly anticipated for continuous progresses in processing techniques. In the past decade, we have established the hardness model for polar covalent single crystals, and revealed an extra hardening mechanism for polycrystalline materials, which shows a hardness–microstructural size correlation and provides further hardening at deep nanoscale due to quantum confinement effect. Therefore, nanostructuring diamond and cBN is still an effective way to enhance hardness. Based on our model, we estimate the hardness upper limits for diamond and cBN with nanograined and nanotwinned microstructures, respectively. Transformed from graphite-like carbon and BN precursors at high pressure and high temperature (HPHT), nanograin diamond and cBN with the smallest grain size of \(~15\) nm can be synthesized, showing enhanced hardness but reduced thermal stability. Starting from onion-like BN and carbon, we successfully synthesized nanotwinned cBN and diamond with average twin thickness of \(5\) nm or below at HPHT. The simultaneous enhancements in hardness, fracture toughness, and thermal stability were confirmed in our nanotwinned cBN and diamond. Our approach offers a general pathway to nano-structure superhard materials for excellent stability and ultrahardness, as well as exceptional tradeoff between hardness and toughness, through the creation of nanotwinned microstructure.

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