Mechanical properties of computationally designed novel carbon enriched $Si_{1-x}C_x$ ceramics: A molecular dynamics simulation study

Ashfaq Adnan *, Sheikh Fahad Ferdous

Mechanical & Aerospace Engineering, The University of Texas at Arlington, Arlington, TX 76019, United States

ABSTRACT

Silicon Carbide (SiC) exhibits excellent mechanical, thermal and electrical properties. Low fracture toughness is one of the limiting properties of SiC that hinders its widespread applications. Recent studies suggest that controlled alteration of local micro-structure may lead to dislocation nucleation in certain SiC poly-types. Here, we report classical molecular dynamics simulations results to demonstrate mechanical behavior of a new type SiC-based "C enriched" ceramics where certain Si atoms are substituted by C atoms. We studied four different systems with different fraction of "C" enrichment, namely 10%, 20%, 30% and 40%. Thermodynamic viability of such novel micro-structures have been studied recently (Adnan and Ferdous, 2015). Significant effects of "C" enrichment on the tensile and shear properties of the new ceramic materials have been observed. Compared to pure SiC, the tensile strengths of enriched systems always increase but to a different extent depending on the amount of enrichment. Shear strengths, however, tend to decrease with increase in carbon enrichment until enrichment is 40%. The elastic constants C_{11} and C_{44} were measured and both increase significantly with carbon enrichment. The area under the stress–strain tensile and shear curves have been estimated to assess the tensile and shear toughness properties and it has been found that both types of toughness increase significantly with carbon enrichment.

1. Introduction

Silicon Carbide (SiC) is an important structural material with excellent wear and corrosion resistance [1–3], high hardness and strengths [4], high temperature and electrical stability [5,6] and many other useful properties. However, poor fracture toughness properties [7,8] of SiC often limits the scope of its applications [1]. Knowing that enhancing fracture toughness would require additional strain energy dissipation during deformation, several research groups attempted to incorporate such mechanisms in SiCs via microstructure tailoring [9–14]. For example, Park et al. [9] and others [10,11] developed microstructure-tailored Si_3N_4–SiC nanocomposites by hot pressing a blend of Si_3N_4 and 3C–SiC at high temperatures (∼1800 °C) for 1–2 h. The process produces distinct Si_3N_4–SiC interfaces which lead to enhanced flexural strength and fracture toughness, both at room and elevated temperatures (∼1500 °C). Using Spark Plasma Sintering (SPS), SiC-based nanocomposites are processed by some groups [12–14]. For example, the YAG (Yttrium Aluminum Garnet) – SiC [13] and the SiC–TiC [14] nanocomposites developed by SPS method (between 1550 °C

and 1800 °C) exhibit both high strength and high fracture toughness compared to their constituents.

Forming altered microstructure to induce dislocation is also observed in monolithic SiCs. Most of these structures are formed during manufacturing stage [15–17] but often remain metastable. Nevertheless, such "process-induced" microstructures often yield enhanced properties. A recent work by Demenet et al. [15] suggests that toughness enhancement via dislocation is possible in certain SiC polytypes along the basal [1 1 1] planes where each basal layer contains only one atom type (Si or C). Such dislocations are realized in both cubic (3C type) and hexagonal type (4H and 6H) SiC crystals. Studies by Hamada [16] and Ma [17] reveal that a metastable super-screw type dislocation in hexagonal SiC is possible during bulk growth of this material. Some other studies suggest that microstructure of SiC can also be tuned by infusing extra "C" atoms in SiC systems [18,19], either as "Si" substitution or as interstitials. For example, Battistig et al. [19] used low energy ion bombardment to produce carbon-enriched 6H–SiC system. Their study suggests that the amount of carbon-enrichment is ion polarity dependent. Gogotsi et al. [20] and Wittorf et al. [21] independently observed that a distinct "excess carbon SiC" interface layer is formed between SiC and Diamond during diamond formation from SiC using CVD method.

* Corresponding author. Tel.: +1 817 272 2006.
  E-mail addresses: aadnan@uta.edu, ashfaqadnan@gmail.com (A. Adnan).
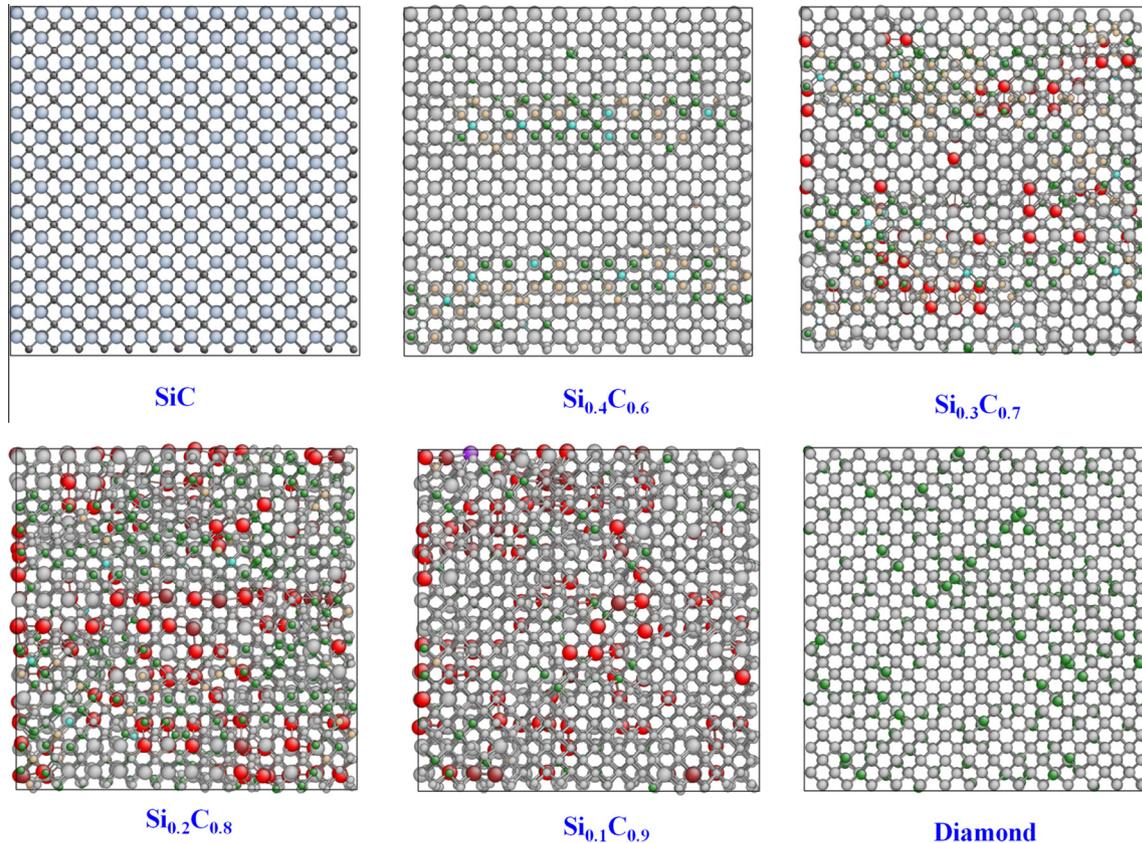
Fig. 1. Equilibrated crystal structure of different "C" enriched models projected on 2D plane.

While these studies did not focus on the mechanical behaviors, we believe that the enriched systems may exhibit improved toughness properties because of favorable microstructural change. We argue that if "Si" atoms from SiC are randomly substituted by "C" atoms, then the bond lengths in the vicinities of the newly formed "C–C" bonds (in place of "Si–C" bonds) will be reduced but the overall crystal structure will not be changed significantly because both "C–C" and "Si–C" bonds are sp^3 type (bonding type responsible for forming diamond/Zinc-blend lattice) [7,8]. As such, there will be a significant number of distorted crystals within the bulk system, which, during deformation, will assist dissipating additional strain energy.

Thermodynamically, developing carbon enriched SiC ceramics depends on "C" solubility in the SiC systems, and it is obvious from SiC phase diagram [22] that C and Si are not soluble in room temperature. Yet, works of Scace and Slack [23], Kobaywashi et al. [24] and Mykhaylyk and Gadriza [25] independently suggested that under certain conditions "C" can be diffused and dissolved in Si or SiC. Independent works by Wernera [26], Newman [27], Ivanovskii [28], Chiang [29] later prove that "C" can be dissolved in Si [26,27] or SiC [28,29]. Recently, using classical molecular dynamics simulation, we [30] investigated the thermodynamic feasibility of a five different "C-enriched" SiC or Si$_{1-x}$C$_x$ ($0.5 < x < 1$) systems where some Si atoms of SiC are substituted by C atoms. Here, we report the mechanical behavior of such systems using molecular dynamics simulation.

2. Molecular models and simulation procedure

Details on the development of the molecular models are described in Ref. [30]. Here we provide a brief description. To develop various "C" enriched models, we started with a base

3C–SiC single crystal [7,31] containing 2744 atoms in total. Four different "C" enriched models are then built by randomly substituting selected fraction of "Si" atoms with "C" atoms. In particular, 10%, 20%, 30% and 40% Si atom substitutions have been considered. To study the effect of model size, we developed larger models with identical "C" fractions (i.e. 10%, 20%, 30% and 40% excess carbon). Each model contains a total of 57,600 atoms. With same level of "C" fraction (e.g. 10%), we developed three independent models to find the effect of random distributions on the mechanical response. The interactions between Si and C have been described by the classical Tersoff Potential [32]. Periodic boundary conditions have been employed along all directions, in all of our simulations except for the cases when we studied the effect of boundary conditions on the mechanical properties. Unless otherwise specified, all simulations have been carried out at 300 K. To develop tensile and shear stress–strain curves, atomistic models were equilibrated first to attain stress-free state; then models were subjected to incremental tensile and shear loading, respectively. During equilibration, simulation was run up to 20,000 time-steps in NPT ensemble [33]. During each incremental mechanical loading, a uniform strain was prescribed on the entire MD model. This is done by uniformly scaling (expanding) the dimensions of the MD cell in the direction of the deformation and re-scaling the new coordinates of the atoms to fit within the new dimensions. After this initial deformation, the MD simulation was continued and the atoms were allowed to equilibrate within the new MD cell dimensions. This process is repeated for the subsequent increments of deformation. The applied-strain increment for the axial tension (y-direction) and shear (xz plane) was 0.25%. After that, all systems were relaxed for 0.25 ps (equilibration run), and then the developed components of "engineering virial" stresses [34–37] were averaged over an interval of 0.25 ps (production run). Stress–strain curves were then