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Elastic behavior of amorphous-crystalline silicon nanocomposite: An atomistic view

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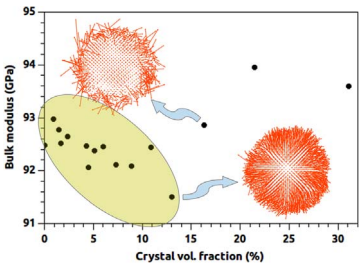
HIGHLIGHTS

- Elastic moduli of amorphous-crystalline silicon nanocomposite are computed.
- Simulations results are compared to an analytical micromechanical model.
- A pronounced effect of the particle-matrix interphase is observed.
- The bulk modulus exhibits anomalous variation with crystalline volume-fraction.

GRAPHICAL ABSTRACT



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ABSTRACT

In the context of mechanical properties, nanocomposites with homogeneous chemical composition throughout the matrix and the dispersed phase are of particular interest. In this study, the elastic moduli of amorphous-crystalline silicon nanocomposite have been estimated using atomistic simulations. A comparison with the theoretical model reveals that the elastic behavior is significantly influenced by the crystal-amorphous interphase. On observing the effect of volume-fraction of the crystalline phase, an anomalous trend for the bulk modulus is obtained. This phenomenon is attributed to the relaxation displacements of the amorphous atoms.

1. Introduction

Crystalline and amorphous solids exhibit widely different properties owing to fundamental structural differences. Thus, the composite materials containing both crystalline and amorphous phases present the opportunity to study a range of interesting phenomena from fundamental as well as engineering standpoints. Out of the many available amorphous-crystalline composites, an interesting class involves those having identical chemical composition in amorphous and crystalline phases. One such example is a nanocomposite containing silicon nanocrystallites embedded in amorphous-silicon matrix. This material is often preferred for fabricating high-performance photo-voltaic devices [1–4]. Although much work has been done to study the transport properties of these composites [5,6], their mechanical behavior is not well conceived so far. In particular, understanding the elastic response of a nanocomposite is known to be non-trivial. Experimental results [7] indicate that amorphization of crystalline silicon under irradiation causes a substantial change in the elastic property of the material. Therefore, it is justified to expect that the elastic properties of the amorphous-crystalline Si–nanocomposite would be different from those of its individual amorphous and crystalline phases. Changes in elastic moduli have already been observed in polymeric composites [8–10]. Analytical studies [11–14] have predicted the elastic moduli of nanocomposites to be

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dictated by the interphase between the constituent phases. Experimental measurements have also validated this idea for nanocomposites with heterogeneous chemical composition [12,14–18]. Nevertheless, systems with homogeneous compositions have not been investigated rigorously. In this backdrop, the amorphous-crystalline Si–nanocomposite serves as an interesting system to understand the role played by an interphase, which connects two phases that are distinct structurally, but not chemically. The present study explores the elastic properties of amorphous-crystalline silicon nanocomposite computed by means of atomistic simulations and the results are compared to theoretical model. Thus, the computations not only probe the elastic behavior of a chemically homogeneous nanocomposite, but also furnish a test case where the suitability of the micromechanical model is tested for a computer-generated virtual sample. Our findings indicate that although the analytical interphase-model remains valid for this system, elastic property of the interphase itself can appear to be anomalous in this regard. As a consequence, some interesting effect is obtained for the nanocomposite with dilute concentration of the crystalline phase.

2. Scheme of simulation

The interatomic interaction in the simulated composite system is described by a modified form of the Tersoff potential [19], which includes the bond-angle dependence. It has been shown to satisfactorily reproduce the elastic, structural and melting properties of both the amorphous and crystalline silicon phases [19,20]. A typical simulation cell consists of a crystalline nanoparticle of 5 nm diameter embedded in amorphous silicon matrix (see inset of Fig. 1). This size of the crystalline phase is consistent with those found in the experimental studies [2,4,5,21]. Although the results presented in this paper are for the 5 nm embedded crystallite, we have also performed these calculations for another system with 2.5 nm crystalline particle. The trends observed for the smaller nanocrystallite are qualitatively similar to those obtained for the larger embedded phase and the corresponding results are given in the supplementary material. The simulation cell is obtained through a selective melt-and-quench process. A spherical region is selected inside a block of crystalline silicon. All the atoms outside this region are heated to a temperature of 2500 K, and then quenched to 10 K at a cooling rate of 49.8 K/ps. During this process, atoms outside the selected region are kept frozen and zero hydrostatic pressure is maintained [22] at the boundaries of the simulation box. The system undergoes several melting-quenching cycles, followed by structural optimization to improve the quality of amorphous phase [23]. Keeping the size of the crystalline region fixed, dimensions of the simulation cell are varied to obtain the virtual samples with different volume-fractions of the crystalline phase. Periodic boundary condition is imposed in all the three directions. To measure the effective elastic constants of the simulated nanocomposite, longitudinal, shear and hydrostatic strains are applied on the simulation cell. Virial stresses [24] are computed and a linear fit to the stress-strain data yields the required elastic moduli, $c_{ij} = \partial\sigma_i/\partial\epsilon_j$ (see Fig. 1 for a typical example). Due to the presence of a single crystalline particle in the simulation cell with periodic boundaries, the moduli are essentially anisotropic. Hence, the Voigt-Reuss-Hill (VRH) [25–30] method is employed to average out the elastic constants over all possible orientations of the embedded nanocrystallite. The cubic symmetry of the diamond-cubic structure of Si yields the Voigt-average Young's (E_{voigt}), shear (G_{voigt}) and Poisson's (ν_{voigt}) moduli as,

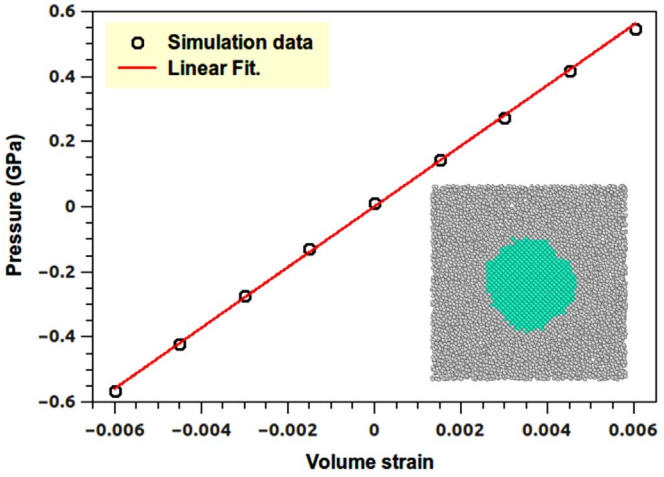

Fig. 1. A representative pressure $\left(\frac{\sigma_1+\sigma_2+\sigma_3}{3}\right)$ *vs.* volume strain $\delta V/V$ plot used for computing the bulk modulus. Open circles represent the discrete data points obtained from quasistatic simulations, while the straight line shows a linear fit. (inset) Cross-sectional view of a typical simulation cell shows the nanocrystallite (cyan) embedded in amorphous matrix (gray). (For interpretation of the references to color in this figure legend, the reader is referred to the web version of this article.)

$$E_{voigt} = \frac{(c_{11} - c_{12}+3c_{44})(c_{11}+2c_{12})}{2c_{11}+3c_{12} + c_{44}}, \tag{1}$$

$$G_{voigt} = \frac{c_{11} - c_{12}+3c_{44}}{5} \tag{2}$$

and

$$\nu_{voigt} = \frac{c_{11}+4c_{12}-2c_{44}}{4c_{11}+6c_{12}+2c_{44}}, \tag{3}$$

which specify the upper bounds on the averaged moduli. Similarly, the Reuss-averaging furnishes the corresponding lower bounds as,

$$E_{reuss} = \frac{5}{3s_{11}+2s_{12} + s_{44}}, \tag{4}$$

$$G_{reuss} = \frac{5}{4s_{11}-4s_{12}+3s_{44}} \tag{5}$$

and

$$\nu_{reuss} = \frac{2s_{11}+8s_{12} - s_{44}}{6s_{11}+4s_{12}+2s_{44}}, \tag{6}$$

where the expressions are given in terms of the compliance coefficients. Following the VRH approach, we obtain the macroscopic, statistically averaged values of the Young's and shear moduli as, $E = (E_{voigt} + E_{reuss})/2$ and $G = (G_{voigt} + G_{reuss})/2$, whereas the Poisson's ratio is $\nu = (E - 2G)/G$.

This yields the isotropic elastic constants as expected to be obtained at the macroscopic scale. All the simulations reported here have been performed using the open source MD code LAMMPS [31], while the OVITO [32] visualization tool is used for the purpose of graphical visualization and atomistic analyses.

3. Results and discussion

The scattered data in Fig. 2a and b display the effective shear and Young's moduli, respectively, as obtained from the atomistic computations (Fig. S1 in the Supplementary information shows these data for the 2.5 nm crystalline phase). We find that both of them exhibit an overall rising trend with increase in volume-fraction of