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The effects of vacancies in the mechanical properties of tungsten: A first-principles study

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ABSTRACT

Both mechanical and structural properties of bcc crystal tungsten in presence of mono and divacancy defects has been investigated by using accurate first-principles total energy methods based on density functional theory. A model for tungsten containing a concentration of vacancies of about 2% and 4% has been developed and used to compute the maximum tensile stress required to reach elastic instability under increasing load. Moreover stress effects on the crystalline structure have been characterized in terms of structural displacements.

1. Introduction

There is a widespread consensus in considering tungsten (W) and W alloys as good candidates for structural and shield material in the future nuclear fusion reactors [1,2]. Indeed W alloys are characterized by thermal properties, such as good thermal conductivity, high melting temperature and low sputtering erosion, of great interest in fusion applications. The main requirement on tungsten materials for structural applications is to be ductile within the operation temperature range. An armor material needs high crack resistance under extreme thermal operation conditions [3,4] and compatibility with plasma-wall interaction phenomena [5].

Thus a deep understanding of the mechanical properties of W and of their atomic-scale modifications in presence of impurities is unavoidable step for the use of W under extreme conditions. In this framework, the determination of the ideal tensile strength, that is an inherent property of materials, can help to identify those aspects of mechanical behavior that are tightly linked to crystal structure and bonding. However, the ideal tensile strength represents the stress at the elastic instability, thus only an upper limit to the yield strength in real materials mainly affected by the complex interplay of defects, such as dislocations and cracks.

The ideal tensile strength along the main directions of pure tungsten have been investigated by employing different first-principles (FP) computational approaches [6,7]. Recently, the calculation of the ideal tensile strengths along three different directions in W shows that the [0 0 1] direction is weaker than both [1 1 0] and [1 1 1] ones, due to the occurrence of structure transitions at low strain values [8]. Thus the [0 0 1] direction has been further analyzed to provide accurate data on the effect on the mechanical properties of alloying (W-Re, W-Ta and W-V alloys) on the ideal tensile strength [9,10]. W and W alloys, in view of their application as plasma-facing materials, need to be also characterized when radiation defects, such as vacancies or interstitials, are formed. Vacancy and vacancy clusters are addressed under both hydrogen [11] and He [12] rich conditions. Moreover self-interstitials and W-V and W-Ta alloys are considered to estimate formation and interaction energies of vacancies [13–16]. In this framework it is lacking an accurate characterization of the effect of vacancies on structural and mechanical properties of crystalline W.

In this paper we report on the determination of lattice parameters, bulk modulus, elastic constants, enthalpies of atomization, formation energy of vacancies and interaction energy of divacancy clusters to provide a clear assessment on the influence of the point defects on the crystalline structure. Moreover computational tensile tests to determine the ideal tensile strength for both W and W with different concentration of vacancies have been performed.

2. Computational details

The first-principles PWscf (Plane-Wave Self-Consistent Field) code of the Quantum ESPRESSO suite has been used [17,18] to accurately characterize W properties. PWscf performs many

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different kinds of self-consistent calculations of electronic-structure properties within Density-Functional Theory (DFT), using a Plane-Wave (PW) basis set and pseudopotentials (PP) [19,20].

Firstly a reliable physical model for W has been developed. Then total energy calculations are performed to check if the model can reproduce experimental structural properties. The same procedure has been employed for W in presence of vacancies. We have considered monovacancy, first-nearest-neighbor (1NN) divacancy and second-nearest-neighbor (2NN) divacancy cases. Then FP calculations have been extensively performed to accurately compute lattice parameters, bulk modulus, elastic constants, enthalpies of atomization, formation energy of vacancies, interaction energy of divacancy cluster and ideal tensile strengths of W and W with different concentration of vacancies.

We used the W Vanderbilt ultrasoft pseudopotential [21–23] with Perdew–Burke–Ernzerhof (PBE) [24] approximant GGA exchange–correlation potential, available in the Quantum ESPRESSO library [18]. 14 electrons (6s, 5d, 5p and 5s) of tungsten atom are explicitly taken into account in the wave functions, via nonlinear core correction and scalar relativistic approximations. All the calculations are performed in the supercell approximation with periodic boundary conditions meant to mimic an infinitely extended system. We considered a cubic supercell composed by a $3 \times 3 \times 3$ array of bcc conventional cells and 54 W atoms.

As shown in Fig. 1, the role of vacancies in tungsten bulk is investigated removing a W atom for the monovacancy case (concentration of defects about 2%), and removing a couple of W atoms for divacancy cases (concentration of defects about 4%).

Thus we considered four cases:

(a) tungsten without defects: a supercell of 54 W atoms (Fig. 1a);
(b) monovacancy case: a supercell of 53 W atoms in which a W atom is removed (Fig. 1b);
(c) divacancy 1NN case: a supercell of 52 W atoms in which a couple of first-nearest-neighbor W atoms are removed (Fig. 1c). The removed atoms are in the [1 1 1] direction at distance $\frac{\sqrt{3}}{2} a_0$;
(d) divacancy 2NN case: a supercell of 52 W atoms in which a couple of second-nearest-neighbor W atoms are removed (Fig. 1d). The removed atoms are in the [1 0 0] direction at distance a_0.

a_0 is the lattice constant of the W bcc crystal structure at zero temperature and zero loading.

The electronic wave functions were expanded in a plane-wave basis set with a kinetic energy cut-off equal to 340 eV (the charge density cut-off was ten times greater) and a $4 \times 4 \times 4$ Monkhorst–Pack mesh of k points is employed [25]. We used Marzari–Vanderbilt smearing [26] with a width of 0.02 Ry. The kinetic energy cut-off, the k sampling and the smearing width were optimized by preliminary calculations on tungsten pseudopotential.

The energy minimization is performed by using conjugate gradient (CG) minimization energy method, with the convergence threshold for self consistency equal to 10^{-7} Ry. Broyden–Fletcher–Goldfarb–Shanno (BFGS) quasi-newton algorithm is used to perform geometry optimization. Ionic relaxation is stopped when both the following conditions are satisfied: energy changes less than 10^{-4} Ry between two consecutive self consistent field (SCF) steps and all components of all forces are smaller than $5 \cdot 10^{-4}$ Ry/Bohr. We used BFGS quasi-newton algorithm for the variable cell relaxation by adding the further condition: all the stress tensor components smaller than 0.05 GPa.

By using first-principles zero-temperature total energy calculations we computed lattice constant a, bulk modulus B, enthalpy of atomization H_a as described in Ref. [9]. To check the mechanically stability of the four systems, the elastic constants C_{11}, C_{12} and C_{44} were calculated following Ref. [27]. If the elastic constants obey the conditions $C_{11} - C_{12} > 0, C_{11} + 2C_{12} > 0, C_{11} > 0$ and $C_{44} > 0$ then the material is found to be mechanically stable. The elastic isotropy has been also examined by the evaluation of the ratio $A = 2C_{44}/(C_{11} - C_{12})$. In an isotropic solid $A = 1$.

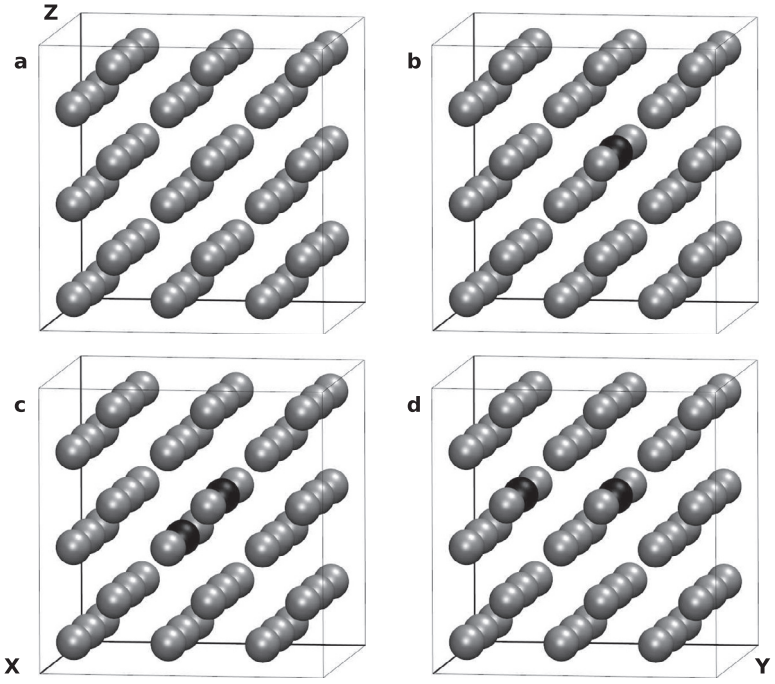

Fig. 1. Simulation cell of the crystalline structure, tungsten atoms are the gray spheres, vacancies are the black spheres: (a) crystalline W; (b) monovacancy; (c) divacancy 1NN along direction [1 1 1]; (d) divacancy 2NN along direction [1 0 0].