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Molecular dynamics simulation of vanadium using an interatomic potential fitted to finite temperature properties

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Abstract

Elastic constants of vanadium are obtained by means of molecular dynamics simulation of stress–strain response. Results at finite temperature are compared with experiments to provide validation of the interatomic potential. Previously, experimental elastic constants and lattice parameter, obtained at finite temperature, were used for parameter fitting at 0 K, leading to poor agreement in the temperature variation. In this work four out of the 11 parameters in modified embedded atom method potential of Baskes have been re-fitted. The calculated elastic properties and the lattice parameter now agree well with experiment.

1. Introduction

Vanadium alloys are considered to be one of the candidate materials for nuclear fusion applications, because of their low radioactivity properties, high-heat-loading capability and promising resistance against neutron irradiation [1,2]. Alloy development including material processing, thermomechanical treatment and fabrication method has been studied [3–5]. Mechanistic understanding of material response, including irradiation behavior such as hardening due to small defects formation under irradiation, is important both scientifically and technologically for further alloy development. Molecular dynamics (MD) simulation is a powerful tool for such microscopic study. It has been successfully applied to study many materials and properties; however, studies of transition metals like vanadium are not yet fully successful due to the difficulty of treating partially filled d-state electrons. Several interatomic potentials for vanadium have been published in the literature. Finnis and Sinclair [6] have derived an empirical parameterized potential that was modified by Ackland and Thetford [7] as a many-body model. Johnson and Oh [8] have derived a potential within the framework of the embedded atom method (EAM) [9,10]. Baskes [11] proposed a modified EAM (MEAM) potential that has the capability to treat angular dependent terms due to d-state electrons. The EAM potentials were applied in an irradiation damage simulation study of vanadium including point defect properties [12–14]. It has been shown that the interatomic potential strongly affects the results of MD simulation; for example, Becquart et al. [15,16] and Osetsky et al. [17] demonstrated that primary damage and subsequent relaxation phase were potential sensitive. In this paper, as the first step toward validation of the potential for vanadium, simple mechanical properties are calculated by MD simulation.

2. Interatomic potential and calculation model

2.1. Interatomic potential

Within the EAM framework, the potential energy E of a system containing N atoms can be written as

$$E = \sum [F_i((\bar{\rho}_i)) + \tfrac{1}{2}\sum \phi_{ij}(R_{ij})]. \quad (1)$$

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In this form, the embedding function F_i is the energy to embed an atom of type i into the background electron density at site i, $\bar{\rho}_i$, and ϕ_{ij} is a pair interaction between atoms i and j whose separation is given by R_{ij} as described in Ref. [11]. The formulation of the functions F_i and ϕ_{ij} depends on the model. The number of parameters of the potential is typically 7–11. They were fitted to the cohesive energy, the lattice parameter, the elastic constants and the vacancy formation energy. In this work the Finnis and Sinclair potential modified by Ackland and Thetford [7] (denotes FS), the Johnson and Oh [8] potential (denotes JO) and the Baskes' MEAM potential [11] (denotes MEAM) are validated.

2.2. Numerical tension test

Although a subset of elastic constants was one of the sets of experimental data to be fitted, other elastic constants were not used for the fitting. The values of the three independent elastic constants can be calculated for validation of the potential. The values of C_{11}, C_{12} and C_{44} were obtained from a numerical stress–strain experiment. The systems were comprised of 432 or 1024 particles surrounded by (0 0 1) plane under three-dimensional periodic boundary conditions. For the case of uniaxial strain to measure C_{11} and C_{12}, the strain was imposed by extending one component of the three basis vectors that define the shape and size of the simulation cell and keeping all other components unchanged from the reference state. The reference state was predetermined from a zero-pressure simulation at a given temperature using the Parinnello–Rahman boundary condition [18]. While systematically extending the cell in one direction, the stress components of σ_{11} and σ_{22} were measured to obtain the stress–strain curve. In the case of measuring the shear elastic constant C_{44}, pure shear strain was systematically applied to the simulation cell and σ_{13} was calculated. Temperature control of the simulation cell was maintained by the Hoover thermostat [19].

3. Results and discussion

3.1. Lattice parameter

The lattice parameter is a simple but essential structural measure of the material. The lattice parameter is well established in available databases such as the ASTM powder diffraction file (PDF). The parameter can be measured by X-ray powder diffraction methods with accuracy to the order of 0.0001 nm. It should be noted that several published potentials for vanadium were fitted to an incorrect value of 0.304 nm for the lattice parameter. The correct value for vanadium at room temperature is 0.30274 nm according to PDF number 22-1058. The previous data of 0.304 nm in file number 1-1224 has been replaced. The calculated lattice parameters at 0 K using the published potentials are summarized in Table 1. All of them show precise agreement with the fitted value. Although the lattice parameter depends on temperature due to thermal expansion, the value obtained at ambient temperature was used as the parameter for 0 K. The lattice parameter is overestimated at 300 K as shown in Table 2. Therefore,

Table 1
Calculated lattice parameter and elastic constants for vanadium at 0 K by molecular dynamic simulation using selected interatomic potentials[a]

	Finnis and Sinclair [7]	Johnson and Oh [8]	Baskes [11]	Exp. [6]
Lattice parameter (nm)	0.30399	0.30399	0.304	0.30399
C_{11} (100 GPa)	2.247	5.993	2.286	2.279
C_{12} (100 GPa)	1.177	4.893	1.171	1.187
C_{44} (100 GPa)	0.445	0.428	0.442	0.426
C' (100 GPa)	0.535	0.550	0.558	0.546

[a] For comparison, experimental data which to be fitted in Finnis and Sinclair type potential are shown.

Table 2
Calculated lattice parameter and elastic constants for vanadium at 300 K by molecular dynamic simulation using selected inter atomic potentials[a]

	Finnis and Sinclair [7]	Johnson and Oh [8]	Baskes [11]	Exp. [6]
Lattice parameter (nm)	0.30502	0.30467	0.30546	0.30399
C_{11} (100 GPa)	2.446	6.355	2.073	2.279
C_{12} (100 GPa)	1.431	5.038	1.011	1.187
C_{44} (100 GPa)	0.715	0.464	0.373	0.426
C' (100 GPa)	0.508	0.659	0.531	0.546

[a] For comparison, experimental data which to be fitted in Finnis and Sinclair type potential are shown.

it must be replaced by a better value as shown later in this paper.

3.2. Elastic constants

Typical stress–strain relations are shown in Fig. 1. The elastic constants were calculated from the set of stress strain points by assuming that they lie on a straight line. A least square minimization was adopted for the calculation. The elastic constants at 0 K calculated from the published potentials are shown in Table 1. The elastic constants were one of the fitted experimental values to determine the potential parameters. Therefore modest agreement with experimental data is obtained. As for the elastic constants, the FS potential was fitted to C_{44} and the bulk modulus B $(= (C_{11} + 2C_{12})/3)$, the JO potential was fitted to Voigt average shear modulus G $(= (3C_{44} + 2C')/5)$ and the MEAM potential was fitted to C_{44}, B and C' $(= (C_{11} - C_{12})/2)$. In Table 2, the elastic constants at 300 K are shown. The results are in poor agreement with experimental data at the corresponding temperature. The elastic constants at 300 K obtained using the MEAM potential are lower in value than those at 0 K. The others at 300 K show larger values than those at 0 K. In the same way, the experimental values used for the potential parameter fitting were obtained at ambient temperature but fitted to the values at 0 K. It must be fitted to the elastic constants at finite temperature.

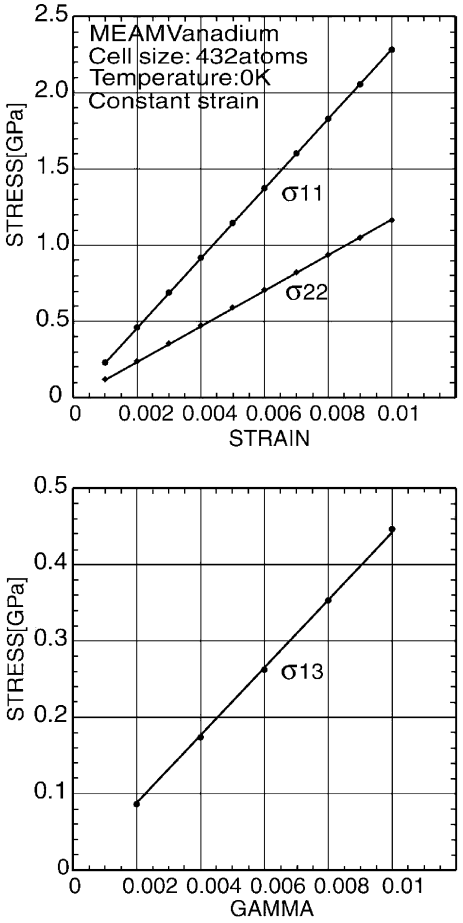

Fig. 1. Typical stress–strain curves as a result of numerical tension tests to obtain elastic constants.

3.3. Re-fitting potential parameters

As described in the previous section, the potential parameters should be re-fitted to the values at the corresponding temperature. The MEAM potential derived by Baskes was selected for re-fitting because it gives a reasonable temperature dependence of the elastic properties. The elastic constants of pure vanadium at room temperature were published in the literature [20–26] and summarized in Table 3. Variation of the values is due to differences in the preparation history of the samples, as well as in the methods of measurement. Most of the results were obtained by ultrasonic velocity measurements. The elastic constants to be fitted were carefully inspected and the data set obtained by Ko et al. [21] was selected in this report. They prepared vanadium specimens by floating zone electron beam refining and applied by a well-established method of measurement [21]. The MEAM potential has 11 parameters. Four of them are re-fitted to structural and elastic properties, since they are highly correlated with the lattice parameter and the elastic constants. The re-fitting was done by systematically repeating the numerical stress strain experiment at 0 and 300 K. The detailed procedure of the re-fitting will be reported elsewhere [27], only the resulting parameters are shown in Table 4 along with the Baskes' original values in Ref. [11].

3.4. Lattice parameter and elastic constants obtained by the re-fitted potential

The values obtained by the re-fitted potential are shown in Table 5. Since the re-fitting was successfully

Table 3
Experimental values of elastic constants for pure vanadium at room temperature in units of 100 GPa

C_{11}	C_{12}	C_{44}	C'	Reference
2.3050	1.2098	0.4308	0.5476	[20]
2.2432	1.1538	0.4125	0.5447	[21]
2.2342	1.1462	0.4253	0.5440	[21]
2.3089	1.1999	0.4336	0.5545	[22]
2.3097	1.2039	0.4290	0.5529	[23]
2.313	1.199	0.437	0.557	[24]
2.3099	1.2017	0.4377	0.5541	[25]
2.2795	1.1875	0.4255	0.5460	[26]

Table 4
Parameters in Baskes' potential for vanadium

$R^{(0)}$	α	A	$\beta^{(0)}$	$\beta^{(1)}$	$\beta^{(2)}$	$\beta^{(3)}$	$t^{(0)}$	$t^{(1)}$	$t^{(2)}$	$t^{(3)}$
2.61	5.03	1.00	4.25	1.0	1.0	1.0	1	4.20	3.83	−1.00
2.63	4.83	1.00	4.11	1.0	1.0	1.0	1	4.2	4.10	−1.00

Values listed top row are re-fitted values and original values are in bottom row. The equilibrium nearest-neighbor distance $R^{(0)}$, the exponential decay factor for the universal energy function α, the scaling factor for the embedding energy A, the exponential decay factors for the atomic densities $\beta^{(l)}$, and the weighting factors for the atomic densities $t^{(l)}$.

Table 5
Experimental data and calculated lattice parameter and elastic constants for vanadium at 300 K using the re-fitted Baskes' MEAM potential

	Re-fitted	Original [11]	Experiment [21]
Lattice parameter (nm)	0.30271	0.30546	0.30274
C_{11} (100 GPa)	2.248	2.073	2.234
C_{12} (100 GPa)	1.148	1.011	1.146
C_{44} (100 GPa)	0.433	0.373	0.425
C' (100 GPa)	0.550	0.531	0.544

done, the lattice parameter and the elastic constants agree closely with the experimental data at 300 K. Other elastic constants, such as Young's modulus and Poisson's ratio, can be derived from the three elastic constants. These values also showed close agreement with experimental data.

4. Summary

Validation of the selected interatomic potentials for vanadium published in the literature has been carried out. Numerical simulation of the stress–strain response was adopted to calculate the elastic constants by means of MD simulation. The MEAM potential derived by Baskes was selected for further optimization. After the re-fittiing of four of the 11 parameters to finite temperature elastic constants, the calculated elastic properties now agree closely with the experimental data.

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