The reconciliation and validation of a combined interatomic potential for the description of Xe in γU-Mo

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Abstract

1. Introduction

The angular dependent potential [1] is a generalization of the embedded-atom method [2, 3]. The total energy E_i of an atom i is given by:

$$E_i = F_\alpha \left(\sum_{j \neq i} \rho_\beta(r_{ij}) \right) + \frac{1}{2} \sum_{j \neq i} \phi_{\alpha\beta}(r_{ij}) + \frac{1}{2} \sum_s (\mu_i^s)^2 + \frac{1}{2} \sum_{s,t} (\lambda_i^{st})^2 - \frac{1}{6} \nu_i^2 \tag{1}$$

$$\mu_i^s = \sum_{j \neq i} u_{\alpha\beta}(r_{ij}) r_{ij}^s \tag{2}$$

$$\lambda_i^{st} = \sum_{j \neq i} w_{\alpha\beta}(r_{ij}) r_{ij}^s r_{ij}^t \tag{3}$$

$$\nu_i = \sum_s \lambda_i^{ss} \tag{4}$$

where F is the embedding energy and is a function of the electron density (rho), ϕ is a pair potential interaction, α and β are the element types of atoms i and j, and s and t = 1,2,3 and refer to the cartesian coordinates. The μ and λ terms represent the dipole and quadrupole distortions of the local atomic environment. This formalism extends the original EAM formalism (the first two terms in eq. (1)) by introducing angular forces. A similar approach which involves the modification of the EAM formalism to include angular forces is the modified-EAM (MEAM) [4, 5].

A given ternary potential requires interactions for each of the three species themselves, and interactions between each species, for a total of six pair descriptions. The U-U, Mo-Mo, and U-Mo interactions are taken from the 2018 version of the Starikov U-Mo ADP [6]. The descriptions of U-Xe, Mo-Xe, and Xe-Xe interactions have been parametrized, but for an EAM interatomic potential [7]. Thanks to the shared underlying physics between the EAM and the ADP type interatomic potentials, the EAM-based Xe pair interactions

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can be implemented into an ADP formalism, provided that appropriate scaling is performed. Therefore, the construction of a ternary interatomic potential in this work should not necessarily be considered a development step, but a reconciliation step, where multiple models have been unified under a single potential formalism, but the underlying descriptions have remained unchanged.

This work presents an interatomic potential for the U-Mo-Xe system, building upon prior separate descriptions for U-Mo and U-Mo-Xe interactions. The interatomic potential is validated, and prior results investigating the U-Mo-Xe system are investigated to demonstrate the differences from prior ternary descriptions.

2. Computational Details

This work makes use of the assumption that for Xe interactions no angular forces are required for the accurate representation of forces. Given the noble nature of Xe interactions and the lack of unpaired valence electrons, we believe that this is a reasonable assumption. Therefore, the EAM descriptions from Smirnova [7] can be taken as complete, and the third, fourth, and fifth terms in eq. (1) can be set to zero.

The first step in the fitting procedure involves an invariant scaling. The quantity $\bar{\rho}$ is given by the sum:

$$\bar{\rho} = \sum_{j \neq i} \rho(r_{ij}) \tag{5}$$

where ρ is the electron density. The energy and forces in the system are invariant to the scaling of the electron density and the embedding energy,

$$\rho(r_{ij}) = \alpha\rho(r_{ij}) \tag{6}$$

$$F(\bar{\rho}) = F(\frac{\bar{\rho}}{\alpha}) \tag{7}$$

where F is the embedding energy and α is a scale factor. This scaling does not change the properties of the pure systems, but does in fact change the behavior of the multicomponent system. The scale factor is applied only to Xe, and in such a fashion as to lead to the same electron density range as that employed for the U-Mo ADP.

3. Results

3.1. Interatomic potential validation

3.1.1. Phase stabilities and structural constants

3.1.2. Xe solid and liquid behavior

3.2. Surface and formation energies of voids and bubbles

$$E_{surf} = \frac{(E^* - E)}{A} \times N \tag{8}$$

$$E_f^{bub} = E^{bub} - \frac{N^{void}}{N^{sys}} E^{sys} \tag{9}$$

3.3. Xe bubble equation of state in U-Mo

The equation of state (EOS) can be determined by tracking the pressure inside the bubble and the bubble size as a function of the number of Xe atoms present in the bubble while the system is equilibrated in an NVT ensemble, which provides a pressure versus density relationship. This data can be fit to an EOS that provides a generalized function predicting the relationship between pressure, temperature and molar volume. In order to extend the applicability of the EOS, temperatures from 300 K to 700 K are analyzed, for all bubble sizes previously mentioned. No restrictions are imposed on the fitting parameters for each individual EOS.

In line with the previous study exploring the EOS of Xe bubbles in U-Mo [8], a virial equation of state is utilized, expanded to the third order with respect to volume and the second order with respect to temperature, as is shown in equation 10,

$$P = \frac{RT}{v}\left(A + \frac{B}{v} + \frac{C}{v^2} + \frac{D}{v^3}\right) \tag{10}$$

where A=1, and B, C, and D are temperature-dependent Taylor series of 1/T (B=b_0 + b_1/T + b_2/T^2, C=c_0 + c_1/T + c_2/T^2, and D=d_0 + d_1/T + d_2/T^2), leading to nine unique fitting parameters. The virial equation is a general function relating pressure, molar volume and temperature that can be directly derived from statistical mechanics [**?**].

The subsequent fit, with and without molecular dynamics data, is shown in Fig. 1 and Fig. 2, respectively. The MD data is removed in Fig. 2 and the optimized EOS is displayed on a log/linear scale to emphasize the differences between the individual isotherms. The optimized coefficients are shown in Table 1.

Table 1: UPDATE THISVirial equation of state (Eq. 10) parameters for Xe bubbles in U-Mo.

Parameter	Value
b_0	197.229 cm^3/mol
b_1	120307.145 cm^3-K/mol
b_2	60.555 cm^3-K^2/mol
c_0	-22038.723 cm^6/mol^2
c_1	2292.793 cm^6-K/mol^2
c_2	-117.564 cm^6-K^2/mol^2
d_0	1030015.045 cm^9/mol^3
d_1	-5.200 cm^9-K/mol^3
d_2	-280.677 cm^9-K^2/mol^3

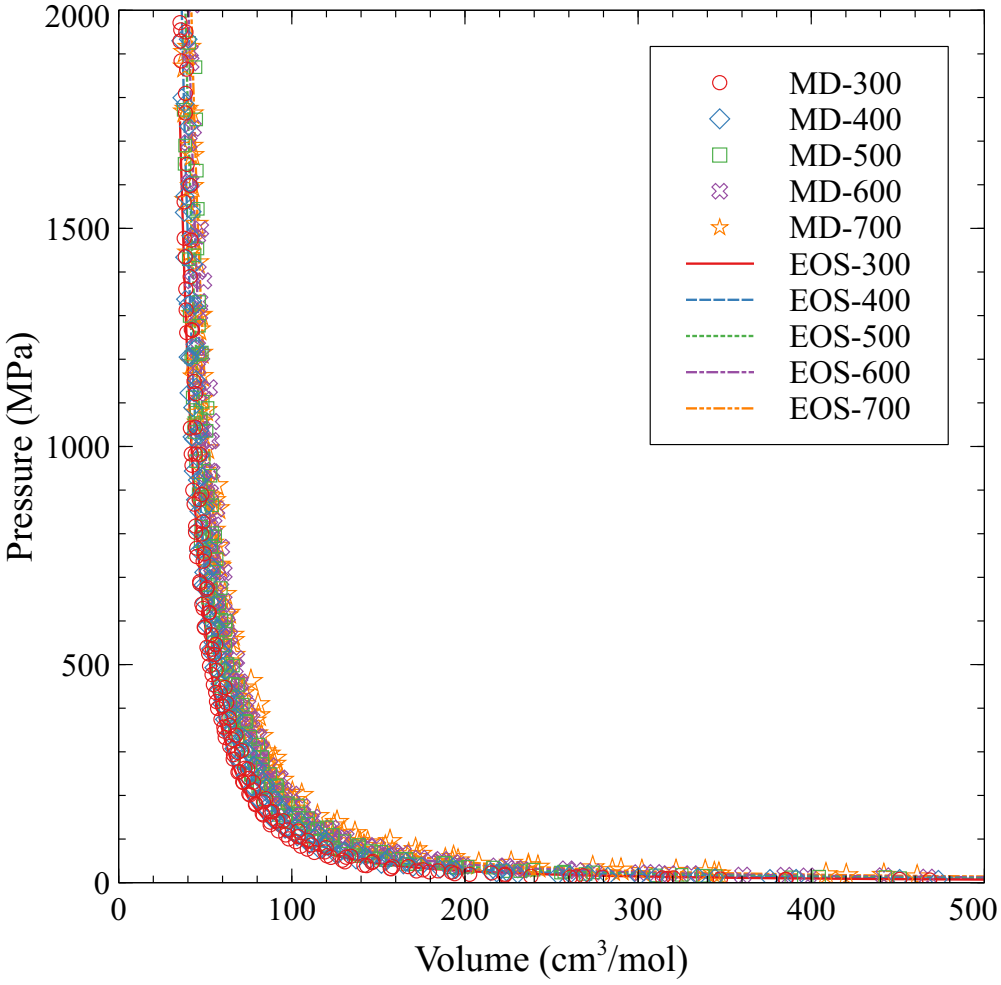

Figure 1: An equation of state (EOS) based on a virial expansion for Xe bubbles in U-10Mo from 400 K to 700 K (a) compared to molecular dynamics data and (b) without molecular dynamics data.

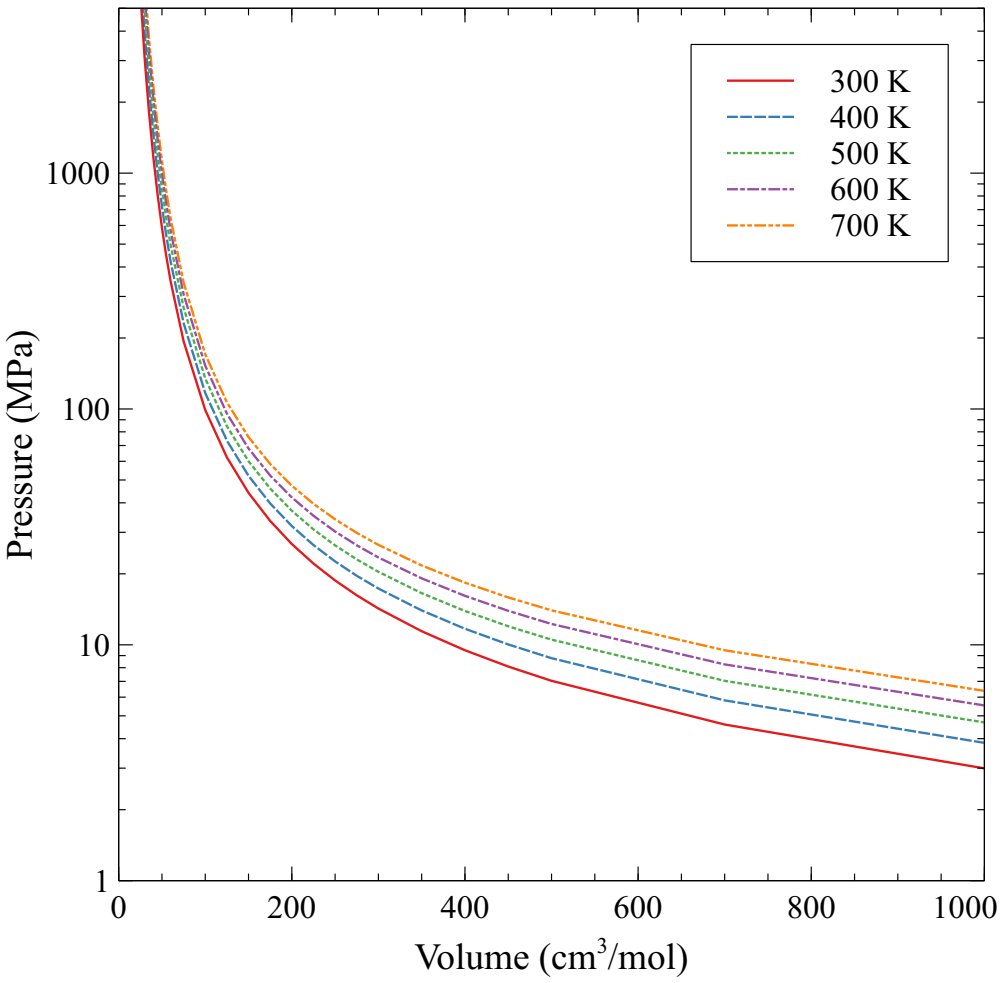

Figure 2: An equation of state (EOS) based on a virial expansion for Xe bubbles in U-10Mo from 400 K to 700 K (a) compared to molecular dynamics data and (b) without molecular dynamics data.

4. Conclusions

5. Acknowledgement

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