Frontiers: 1185448: The reconciliation and validation of a combined interatomic potential for the description of Xe in gamma U-Mo

Reviewer 1:

The authors reconciliated the interatomic potential for U-Mo-Xe and tried to validate it by comparing with EOS. The research work showed the importance and meaningfulness in evaluating the fuel performance based on the MD method. Comments and suggestions are listed below.  
1. The reconciliated interatomic potential should be clearly described in the main texts.

The steps taken to combine multiple potential are identified.

2. Check the subtitles with the serial numbers.

Don’t know what they mean.

3. Check the captions of Figure 6 and Figure 7.

Captions has been modified to state 300 K instead of 400 K.

4. Check the abbreviations of EOS. Do not repeat the full name and abbreviations.

The abbreviation EOS is now only defined once.

5. Detailed comparisons between MD and EOS should be carried out to evaluate the proposed interatomic potential. More supporting data are necessary.

To do…

6. MD results should be given with error bars, indicating statistical fluctuations due to initial atomic configurations and other factors.

To do…

1. Is the quality of the figures and tables satisfactory?  
   - No  
     
   b. Does the reference list cover the relevant literature adequately and in an unbiased manner?  
   - Yes  
     
   c. Are the statistical methods valid and correctly applied? (e.g. sample size, choice of test)  
   - No  
     
   d. Is a statistician required to evaluate this study?  
   - Yes  
     
   e. Are the methods sufficiently documented to allow replication studies?  
   - No

Reviewer 2:

This manuscript presents the development and validation of a combined potential to described the Xe behavior in gamma-UMo fuel alloy. Such a knowledge is important because of the critical role of Xe behavior at the atomic scale on such nuclear fuel performances and safety in reactors.  
This paper is well written and the scientific knowledge reported here is of broad interest to the Nuclear fuel community, as it may serve as an example to develop other interatomic potentials for other fuel systems. The paper is, in my opinion, perfectly in the scope of the Frontiers in Nuclear Engineering, section Nuclear Materials.  
As stated in the title and being the main goal of the paper, the validation of such new potential constitutes the core of the paper. However, my main concerns is about the validation itself, which is somehow lacking direct comparison to the available experimental data. Indeed, the validation is here essentially performed towards the previously available potentials or other calculations methods and approaches, while one should always compare whenever possible to experimental data. The corresponding data are obviously scarce and quite limited, but they do exists, especially when considering the validation against phase stability and structural considerations.  
Nevertheless, the current paper is a nice review paper I would suggest to consider for publication with moderate revisions. In particular, I invite the authors to assess the following points.

• Please add the units for all terms described in all equations. For example, are energies in equation (1) given in J, eV, or kg.m2.s-2? Same applies to all other equations within the paper.

To do….

• On page 3, line 43: please explain what is behind the concept of embedding energy to help the non-expert reader.

To do…

• On the potential adjustment section (2.2, page 3-4): Authors stated that scaling affects the multi-component behavior. How much elemental behavior is sensitive to small scaling? What is the impact of such scaling on the behavior of Xe in the studied system? Do the authors can elaborate and/or justify on this? What was the scaling value used in the reported reconciliation?

To do…

• On phase stability section (2.3.1, page 4): recall of the main structural characteristics of the gamma phase of U-Mo alloy would be welcome here. At least, one may expect the space group, and cell parameters, as available in literature.

To do…

• On the vacancy formation section (2.3.2, page 5):  
• What is the energy difference in creating a vacancy by removing one U atom compared to removing a Mo atom?

To do…

• Similarly, when calculating the substitutional Xe, is there differences when substituting a U atom compared to a Mo atom.

To do…

• How the chemical potentials are calculated for U and Mo, and also for Xe? Indeed, the choice of the pure Mo and U reference compounds may have an impact on the calculated values of the chemical potentials, and thus on the deduced defect formation energies using equations (8) and (9). The statistical approach as described in reference 25 and used in this paper can solve the above-mentioned issue. More details would be welcome.

To do…

• On page 5, line 121: the authors state that the voids were created large enough to ensure that the surface energy sufficiently converged.  
• What was the minimum size studied here?

To do…

• This approach de facto excludes the first steps of the formation of Xe bubbles. How does it affect the validation of the potential, given that there is no clue on the potential capability to describe properly those important initial states?

We acknowledge that the nucleation of bubbles is not explored in this work. It is assumed that nucleation can occur, as observed experimentally. The relationship between the internal bubble pressure and the surrounding matrix is deemed the critical component for utilization in mesoscale models. A study on nucleation could indeed be performed, but was deemed beyond the scope of this manuscript. Such a study it not expected to change any of the conclusions of this work, but would provide additional information to aide in the understanding of fission gas bubble behavior in U-Mo.

• On section 2.3.3 (page 5):  
• have the authors evaluated the impact on the results reported here of inserting all Xe at once and then letting the system to relax, compared to the one Xe at a time approach ?  
• Is there any difference between the distance calculated using two atoms from the matrix and two Xe atoms to evaluate the bubble size? Have the authors crosschecked that they are similar? In case Xe is solid or liquid, would it be possible (in a very naïve view) to have Xe occupying only a part of the void?  
• Often, bubbles are stabilized by the presence of Xe occupying vacancies. Have the authors considered or tested the formation of bubbles by randomly inserting substitutional Xe and vacancies, and let the system relaxing?  
• Why the pressure inside bubbles was evaluated as stated? Is there any benefits compared to for example using the known pure Xe equation of states, and Xe-Xe distances?  
• On page 6, section 3.1.1:  
• Please add phase diagram and other experimental relevant information to compare the calculated phase stability with.  
• It would be interesting to compare all conditions tested in this study to the reported phase diagram.  
• On figure 1: there is (a) and (b) in figure caption, but there is no mention of a and b in the figure itself. Worth to change for (left) and (right).  
• On figure 2:  
• This is a nice 3D plot, but hard to read and the 3D is not significantly important here. I would suggest keeping only a contour plot for sake of clarity. This would also allow for showing directly where both phases are situated, and also a potential comparison to the experimental phase diagram.  
• On page 8, line 172-175: More details on the distribution would be very useful to help understanding the impact of local distortion (if any) as a function of the local environment (i.e. number and nature of 1NN and 2NN). Are there any differences between U-U, U-Mo and Mo-Mo distances? The distribution shown in Figure 3 may give further insight if one consider the local difference responsible of the energy variation. Are the lower energy corresponding to more U atoms as first neighbors as suggested by the figure 4? Similarly, Figure 4 is a bit unclear. Plotting those very interesting results using a distribution for each case (i.e. in the form of multiple line distribution like in figure 3) can help to see the differences more clearly. If possible (statistic wise), the mean and standard deviation would be very interesting to share.  
• Figure 6 and Figure 7:  
• Captions of Figure 6 and 7 are exactly the same and contain (a) and (b) which is seemingly correspond to figure 6 and 7.  
• Figure 6 would require log scale in y-axis as done in Figure 7.  
• Dots in Figure 6 are probably too big to make the figure readable easily.  
• The deduced EOS is compared to Kaplun calculated data. The author claims that one can use their updated EOS in the Xe-U-Mo system. There is slight difference in between those two EOS. However, without comparison to experimental data, one may wonder which one is the most appropriate. Please elaborate on this, and if possible/available provide experimental data from literature to compare with both EOS.  
• On the conclusion, authors stated “This work has served to demonstrate the need for an updated interatomic potential, and provided that tool, which will serve to inform mesoscale and engineering scale fuel performance simulations, providing increased accuracy and reducing known errors, allowing for the exploration of critical phenomena in U-Mo fuels.” This statement is not correct as the paper is providing no comparison to experimental data supporting this statement nor the demonstration of actual improvement. At least, the accuracy and known error should have been mentioned and discussed in the light of the reported calculated results.

a. Is the quality of the figures and tables satisfactory?  
- No  
  
b. Does the reference list cover the relevant literature adequately and in an unbiased manner?  
- Yes  
  
c. Are the statistical methods valid and correctly applied? (e.g. sample size, choice of test)  
- Not Applicable  
  
d. Is a statistician required to evaluate this study?  
- No  
  
e. Are the methods sufficiently documented to allow replication studies?  
- Yes