We thank the reviewers for their time and their comments. We believe that all suggestions/comments have been appropriately addressed below, and correspondingly included in the manuscript.

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

Reviewer 1:

The authors reconciled 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.

Given that limited modifications of the potential were actually made, the full descriptions of the potential fitting processes, tools utilized, fitting data sets, etc., were left to the original manuscripts. All steps performed to modify the potentials are outlined, and additional information has been provided such that this process should be replicable.

2. Check the subtitles with the serial numbers.

Subsection titles have been lightly modified for consistency and clarity.

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.

There is limited data available for comparison, and limited atomistic-level comparisons that can be made to quantify the differences between the MD data and the fitted EOS beyond what was performed. Additional comparisons have been made to the experimental EOS to demonstrate areas of agreement, and importantly, areas where there is disagreement. The presented EOS is designed for Xe in a UMo system and has been validated against all applicable data. Finally, this equation of state is being implemented into mesoscale models for the description of fission gas swelling behaviors and is being compared against other EOSs for Xe gas. Thus, we believe that any spurious behavior in the EOS will be identified through the utilization of the EOS.

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

Where appropriate, error bars have been added, or the statistical error has been discussed within the text.

    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)
- 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.

The units can be modified to fit the appropriate system. Frequently, the units within LAMMPS denoted as "metal" are quite convenient, which provide energies in eV, forces in eV/Angstrom, etc. However, this equation is not limited to a specific unit set, as long as the units are appropriately consistent. Text has been added to denote that commonly the units of this equation are in eV.

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

A clarifying parenthetical has been added: F is the embedding energy (the energy required to place an atom type alpha into the electron cloud).

• 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?

Additional sentences have been included to ensure that the scaling procedure is reproducible and to provide clarity. This was a small scaling and should provide minimal impact on calculated ternary properties. The accurate solution energies of Xe (to the best knowledge available), provides confidence in the ability of the potential to predict properties of the ternary system.

• 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.

A reiterative statement has been included in section 2.3.1 to ensure the reader is aware of the crystal structure (body centered cubic) and the solid substitutional nature of the gamma phase.

• 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?

There should be no difference in the vacancy formation energy computed by removing U and Mo atoms because the reference and the defected cells have exactly the same numbers of U and Mo atoms. The formation of a vacancy in a real material system does not involve removing an atom, although it's seemingly that way in atomistic calculations. Rather, the total mass should be conserved, as Eqs. 8 and 9 imply. Vacancies can be generated from sinks such as free surfaces and grain boundaries without removing any atoms, and they do not feature chemical types. In atomistic calculations, as the reference cell usually has one more atom than the defected cell, the chemical potential of the removed atom is added into the defected cell to conserve the total mass and remove the dependence of the result on the type of the removed atom. As such, there is no distinction between "U vacancies" and "Mo vacancies". There is a slight difference in composition between cells having a U atom removed and those having a Mo atom removed. Here, we assume such a negligible change in composition won't cause a substantial change in vacancy formation energy. The insertion of Xe follows the creation of a vacancy, and the resulting Xe solution energies are independent of the type of the removed atom as well.

A paragraph has been added in Section 3.2 to clarify this point.

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

Please see the response to the comment above.

• 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.

A brief description has been added in Section 2.3.2 to show the process of computing the chemical potentials. Indeed, the purpose is to avoid using pure Mo and U reference compounds, and this is critical as using inaccurate chemical potentials will lead to a dependence of the resulting vacancy formation energy on the type of atoms removed to creating vacancies.

• 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?

The minimum void size was approximately 8.5 Angstroms in radius, and thus a 1.7 nm diameter void. The minimum size is not noted specifically in the text but is shown in Fig. 5. Such voids are stable, even without Xe atoms present, but do exhibit a higher surface energy than larger voids, hence the need to increase the void size to ensure a converged surface energy.

• 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. A paragraph in the methods section has been added to address this point.

• 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 ?

Yes. This was performed in the original study, reference 14. If too many Xe atoms are inserted at once, the pressure will increase too dramatically, causing Xe ejection from the bubble, interstitial loop formation, or, in extreme cases, strong radiation damage cascades. This behavior is unphysical to the described systems and is important to avoid. Additionally, there is a minimum energy state that can only be found by adding in Xe atoms in steps, as adding all the Xe at once would dramatically inhibit the ability of the Xe to move, relax, and reduce the system energy.

• 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?

The differences should be negligible, but significantly more difficult to execute if analyzing Xe atoms. Atoms on the surface are stationary at these temperatures and timescales, while Xe atoms move. Thus, extreme caution would need to be utilized to accurately obtain two Xe atoms on opposite sides of the bubble. Arguments could still be made on which method is more accurate. Additionally, it has been observed that Xe effectively occupies all of the void space, where in very low-pressure bubbles Xe atoms fluctuate and diffuse significantly. Thus, Xe has been observed to exist in a gaseous state. In theory, if Xe could exist as a solid or liquid in the given system, yes, then it could partially fill the void volume. However, it is in a gaseous state, as governed by the interatomic potential itself, and so does not exist as a liquid or solid for these systems.

• 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?

This effectively indicates a bubble nucleation study, or a Xe-vacancy binding energy study. This has not been explored here, but the attraction of Xe and vacancies has been established in similar systems, and the energetic relationships known and presented in this manuscript point to a near certainty of binding of Xe and vacancy atoms. Further studies on Xe-vacancy binding could be pursued, but the suggestion of the reviewer where a system of vacancies and Xe atoms are distributed in U-Mo and relax to form bubbles is very likely beyond the timescale for which MD can sample.

• 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?

The pressure is calculated from the potential and the molecular dynamics calculations. The Xe equation of state itself may not hold for Xe bubbles in a U-Mo alloy, as there is interaction along the surface of the void with the Xe bubbles, adding additional forces which impact the bubble. In the large bubble limit, Xe should have a relatively low density and should act effectively as a pure Xe gas. Thus, the EOS for this system and the experimental EOS for pure Xe gas should converge at low densities, which is shown to occur. However, at high densities, the role of the matrix is amplified, leading to deviations from the experimental EOS, which are observed here.

• 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.

In response to the previous two points: Some prior comparisons have been made, including the ADP potential paper itself: https://doi.org/10.1016/j.jnucmat.2017.11.047, and a recent paper from those same authors: https://doi.org/10.1016/j.jnucmat.2023.154265. However, the phase stability of interest concerns the thermodynamically stable gamma phase, and a tetragonally distorted metastable phase which does not appear on the phase diagram but is known. The ADP potential is developed to simulate the bcc U-Mo solid solution phase, and it does not do as a good job for other U-Mo phases. This means that it does not reproduce the phase diagram, making the comparison somewhat irrelevant. The EAM potential fails to stabilize the U-Mo solid solution phase, raising the need of reconciling the ADP potential with Xe for simulating Xe behavior in bcc U-Mo alloys. We have added a few sentences in Section 3.1.1 to discuss this.

• 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).

Caption and associated text have been changed to left/right nomenclature.

• 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.

We used the 3D plot to show the c/a ratio in addition to the boundary between bcc and bct phases. To improve the clarity, we have changed the axes scales and rotated figure 2 to better show the region in which the bct phase is more stable. As described in the above, the ADP potential focuses only on the bcc solid solution phase and does not permit a valid comparison on the whole 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.

We appreciated the suggestion and added two more figures (3 & 4) to show the impact of local atomic environment on atomic pressure and volume and bond lengths. Indeed, there are noticeable differences between U-U, U-Mo, and Mo-Mo bonds in terms of length and distribution. There are also noticeable differences in atomic volume and pressure of U and Mo atoms. The dependence of atomic volume on 1NN Mo atom seems to be consistent with the Xe solution energy, providing a plausible explanation for the preference of Xe for U-rich environments. In addition, we have expanded figure 6 to include vacancy formation energy and the difference between Xe solution energy and vacancy formation energy. The corresponding descriptions have been updated to have more discussion on their dependence on atomic environment.

• 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 captions in Fig 6/7 have been updated accordingly. We believe that showing the data as it in Fig 6 on a lin/lin scale is the best representation of the actual raw data, illustrating the somewhat minor differences with respect to temperature and the sharp increase with pressure. The adjustment to a log/lin scale in Fig 7 is provided to aid the reader to identify temperature trends. The markers in Fig 6 have been adjusted down but provide no real benefit to readability. The data set included is large, for completeness, and inhibits the ability to discern the optimized lines from the markers. However, the associated statistics included within the text provide the necessary context, and we still believe that this figure demonstrates the intended purpose of displaying the nature of the data which is being fit.

• 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.

Unfortunately, there is no known experimental data for which to base a comparison. There has also been disagreement in mesoscale researchers about the utilization of the Kaplun EOS, or modified van der Waals EOSs, or utilizing some other form. Kaplun's data itself contains some statistical errors. Added discussion has been included, emphasizing that at low Xe densities, the differences are negligible, being exaggerated by the log/lin scale, but at high pressures, where the U-Mo-Xe interactions come more into play, significant differences are present.

• 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.

The reviewer is correct that for many of the computed properties direct comparisons are not made. This is either caused by the limitation of the capability of the potential or the lack of experimental data. By reconciling the ADP potential and the Xe-metal interaction from the EAM potential, atomistically studying Xe behavior in stable U-Mo solid solution using molecular dynamics becomes possible. This will help us understand gas behavior and gas bubble formation and give more reliable data to support mesoscale and engineering scale modeling when obtaining experimental data is beyond reach. By no means we should claim that the potential is ready for making quantitative predictions. We have modified the conclusion Section to clarify this.

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