Reviewer 1

The authors present atomistic simulations of Xe gas bubbles in gamma U-Mo alloy. The results may be regarded as novel and interesting. The estimated equation of state way be used in the macroscale codes describing an evolution of nuclear fuel at the operating conditions. However, several points need to be addressed:

1. The authors state that "there is a region below a Xe/vacancy ratio of 0.15 where additional Xe atoms inserted into the bubble produce no noticeable change in the relative bubble energy". What atomic density of gas does this ratio correspond to?

This Xe/vacancy ratio corresponds to a molar volume of approximately 80 cc/mol. This information has been included in the manuscript. The choice of molar volume is to maintain consistency with results appearing later in the text.

1. The reference state in equation (3) is substitutional-vacancy pair. However, concentration of such defects is proportional to [Xe][Vac]exp(-E/kT) where [Xe] and [Vac] are concentrations of Xe substitutional and a single vacancy, respectively, and E is a binding energy. In this case, concentration of such sub-vacancy pairs is much lower than concentration of Xe substitutionals. Thus, Xe substitutional should be used as a reference state in equation (3). Also, I strongly recommend to collect all energies in a one table.

We thank the referee for the comment. We ourselves had disagreements on the proper way to establish a reference state. We acknowledge the referee’s comments and have modified the reference state the be the substitutional configuration. Fig. 4 has been updated to reflect this change, as well as the text.

In addition, the referee’s comments prompted us to go back and refine our defect energies to make sure we were analyzing the correct configuration. This study on defect energetics has been included into the manuscript. With the formation energies added as a table. We feel this strengthened the manuscript.

1. In my opinion, data from figure 5 should depend on size of the calculation cell. What is the size of simulated system? Did authors perform an analysis of size effect?

The quantitative relative volume as a function of Xe/vacancy ratio will change with changes in supercell size volume, but the qualitative relationships will not vary. Supercell size effects were investigated in limited cases to ensure the qualitative relationships held. This information has been included in the manuscript.

1. Please, add units to “predicted-calculated” axis on figure 6.

Units have been added onto Fig. 6.

Consequently, this work may be published after considering the above comments. I recommend major revision for this work.

Reviewer 2

Review: JNM\_2019\_841

An improved equation of state for Xe gas bubbles in gamma U-Mo fuels

B. Beeler et al.

The authors present a study on Xe gas bubbles in U-Mo fuels, an alloy selected to replace current highly enriched uranium fuels. The problem of Xe gas bubbles created in the fuel is important, since the induced swelling can dramatically reduce the fuel performance and lead to failure. The authors successfully argue that, since current models are based on small bubbles only, mesoscale models require further input from atomistic scale to better predict the fuel performance and stability.

The study is relevant, well-written, and scientifically sound. The extension of the Young-Laplace equation is interesting, and the authors provide two forms of EOS that can be used in higher-scale models. As such, this reviewer would support publication of the manuscript, granting the comments listed below are addressed, especially regarding the validation of the model and validity of its predictions.

1. The value of the study is only as good as the reliability of its predictions, and in the case of empirical interatomic potentials, the latter must be addressed. The authors used and EAM form potential developed by Smirnova et al., which should be discussed, even briefly. How was the potential parameterized? Is it expected to do well against defects (Xe interstitial and U-Mo surfaces)? Is there any validation against DFT calculations or experimental measurements? For instance, the propensity of Xe to reach a bubble rather than an interstitial site is strongly dependent on the binding energy, which thus needs to be accurately reproduced. Even if the justification for using this EAM is “that’s the best we have”, something of the sort should be stated, along with the expectation of reliability of the results.

A discussion of the applicability of the potential is included below as well as in the manuscript.

This potential is capable of describing the body-centered cubic phase of U-Mo alloys, and is presently the only potential capable of describing the U-Mo-Xe ternary system. The potential is able to reproduce the stable structure, modulus of elasticity, room-temperature density and melting temperature of U–10Mo. Additionally, this potential is able to reproduce a number of properties of pure Xe gas and face-centered cubic Xe. However, it is unknown the level of accuracy of the defect properties of Xe in U-Mo with this potential, as no such experimental data is available, and the inherent mechanical instability in density functional theory simulations makes such examinations untenable. The authors would recommend validation of this potential with *ab initio* molecular dynamics simulations; however, such a study was beyond the scope of this project and additionally no such study of the kind has been performed.

1. Additionally, what about size effects? If Figure 2 is meant to represent the entirety of the system, it appears that the bubble (not the biggest one considered in this work), represents a significant fraction of the system, and may lead to significant interaction with image system under periodic boundary conditions.

Size effects were considered and larger supercell sizes were investigated to ensure no simulation artifacts were present that related to the size of the supercell. It was observed that the observable stress fields around low-pressure bubbles are localized to less than 10 Angstroms from the edge of the bubble for all bubbles investigated in this work. There were no observable changes in equilibrium bubble properties, such as energy and pressure, with changes in supercell size. A comment on analyzing different size supercells has been added to the computational details section.

1. Missing caption in Figure 6.

We apologize for the mistake and the caption has been inserted.

1. For the EOS part, the authors list comparison with results from Hu et al.. Some details should be given about that study (if it was, it was missed by this reviewer). It seems both studies used the same EAM potential, with would motivate the one-to-one comparison. Otherwise, comparing EOS fitted with different data set (from different interatomic potentials) would not add much value to the paper.

Compare more thoroughly Shenyang’s work. Same potential. His focused on high pressure systems.

1. Page 3, lines 56-57: “Temperatures of interest are (…), which span the realistic operating temperatures of U-Mo fuels”. Citation?

We thank the referee for the comment, and a citation has been provided.

1. Figure 3: Is there a clear reason why larger bubbles show a much steeper increase in relative bubble energy than the smaller ones?

This relates to the nature of the X-axis, in that it is Xe/vac ratio. Comparing Xe/vac ratio against pressure shows nearly identical curves for each bubble size.

Explain qualitatively reason for curves. Pretty sure it is Xe/vac ratio vs # of Xe