We thank the editor and the reviewers for their patience in receiving a resubmission. Developments in lammps allowed for a new implementation of random solid substitutional alloy construction which gave us the ability to precisely prescribe the composition, which removed a significant amount of statistical fluctuation in the dataset. However, this demanded a full rework of all the simulations, which is what has been performed. The conclusions and the magnitude of the key data did not change, which provided us a great deal of confidence in our previous simulations, but being able to provide more statistically significant results in this manuscript was deemed a necessary step that we feel has strengthened the manuscript.

We have addressed all reviewer comments below in blue text.

Reviewer 1

First, I would like to apologise to the authors for the time it took me for various reasons. I think that this is a great research subject, and that the successful multi-scale approach with BISON is very nice. I also appreciate the way thermodynamical properties beyond energy are considered, beyond simple surface energies. However, I have one main issue with the size of the simulation boxes, which could significantly change everything related to surface energies. I understand that it is not practical to get complete box size dependences for all radii, compositions and temperatures, but this really needs to be discussed, with a plot of surface energy as a function of box size for one case to estimate systematic errors. I also have a couple of comments below.

- Box sizes are mentioned in terms of multiples of the unit cell. It would be better to add the rough equivalent in angstrom or nanometers for the readers who don't know off the top of their head the lattice parameter of metallic U.

Have added dimensions in angstroms in the computational details section to provide actual dimensions of supercells.

- No analysis of box size effect is shown. Whilst I can accept that 10x10x10 is enough to calculate bulk properties, I am not entirely convinced that 30x30x30 is enough for a void. Using ~3 Å for the lattice parameter, this means that the boxes are 90 Å across. This seems very small compared to the largest voids with a radius of 30 Å (so 60 Å across). I would expect surface energy to be very dependent on the box size, as most of it would be in long-range elastic fields.

A discussion of variation of surface energy on box size has been included. The text within the computational details has been modified as follows: “Utilizing the same NPT-ensemble details as for equilibrium properties, the system is equilibrated for 50 ps. Subsequently, atoms within a spherical region of a prescribed size are removed from the center of the supercell, generating a void, and the system is allowed to relax for a further 50 ps in an NVT ensemble at the bulk equilibrium volume. Equilibrating the void in an NVT ensemble mimics the void residing in an infinitely large bulk with an ideal equilibrium volume, which is a realistic approximation that has the added benefit of reducing cross-boundary interactions. Regardless, supercell size convergence studies were performed which showed less than 1% variance in the void surface energy for supercells greater than 30x30x30 unit cells. Finally, averaging of the system potential energy is performed over the final 25 ps of each individual 50 ps segment.”

- p.3, shouldn't equation 3 be Cp = (E2 - E1) / (T2-T1), a finite difference equation equivalent to the formal definition CP = ∂H/∂T?

We have changed the nomenclature to be more specific. The equation now reads (H2-H1)/(T2-T1), clarifying that potential and kinetic energy contributions are taken into account.

- p.3 & 4, the notations E, TE and U are used for the energy, which is a bit unclear.

In accordance with the previous comment, we believe the modification in notations have made the equations clearer.

- in general, although enthalpy and total energy are the same when the pressure is 0, I think that "enthalpy" should be used instead of "energy" in several places. In particular, equation 8 is wrong in general, and is only true here because U=H.

This has been modified to say deltaH, and in the text stating that this is equal to U since the P=0 in these simulations.

- p.5, the authors mention that "the Gibbs Formation Energy is negative over the entire compositional range" despite the enthalpy of mixing being positive for some compositions (discussing reference 11). This is true at a given temperature, but not at arbitrary low temperatures (with the limit case that the Gibbs free energy is equal to the enthalpy at 0K). The temperatures for which this is true should be mentioned.

We thank the reviewers for this comment, and have modified the sentence as follows, per Moore’s work and the phase diagram: “This corresponds with the findings of Moore, who also showed that if entropic effects are included, the Gibbs Formation Energy is negative over the entire compositional range at temperatures above 1100 K, in accordance with the phase diagram.”

- still p.5, the total energy needs to be considered in the calculations. Just adding a kinetic energy of 3 kB T per atom would be enough, but keeping around the potential energy is just asking for trouble. All the equations use the total energy (or the enthalpy), and some of them would give very different results without the kinetic term.

The total energy is considered where appropriate according to the equations laid out in the computational details, namely including potential and kinetic terms to determine enthalpies, and thus heat capacity, and subsequently the entropy. We believe that the modifications to the computational details equations have made this more clear.

- In figure 1, the difference between temperatures is much smaller than the difference between U and Zr, which makes the plot difficult to read (e.g., does the composition change the difference in energy between say 900 and 1200 K?). From my point of view the same information would be much more legible with a second plot showing the difference with the energy at 900K instead of the absolute energy.

We thank the reviewer for the comment and have accepted their recommendation. Fig 1 now has Fig 1a and Fig1b, which shows the original chart and the energy per atom with respect to 900 K, respectively.

- the trend in figure 5 is very unclear. Lines between points are useful, otherwise it would be very difficult to se anything, but they should not be splines. Splines give a sense of continuity which is simply not there in this case, where the data is noisy. I can (barely) see that Cp is slightly lower for Zr than for U, but it is a bit of a stretch to say that it increases with temperature, considering the dispersion of the results. Which itself is quite unexpected, one would expect anharmonic effects to make it increase with temperature measurable over a 400 K range, particularly just below the melting point. Is this related to my comment about equation 3?

With the modification in the configurational setup, this chart has become much clearer, and less noisy. We believe the data and trends in the chart are now readily discernible.

- Are void sizes measured, or as-created before relaxation? I could not see it in the manuscript, and this could change the discussion.

Void sizes are reported as-created before relaxation. Supercells are generated with approximately equilibrated lattice constants, as such, the void sizes should not change due to cell relaxation. We also did not see any evidence of void shrinkage during the simulations, so the void sizes should be accurate for the relaxed systems, within +/- 1 Å. A sentence was added noting that as-created void sizes were utilized in the determination of the surface energy.

Reviewer 2

I would like to recommend for a moderate revision to improve the paper on the following points:

1. As shown in Fig 11, the dependence of porosity on the surface tension (which is approximated here as the surface energy) of the void is rather significant. The surface tension should actually be approximated by the surface free energy rather than the surface energy (e.g. see Ref Acta Metallurgica 4, 576 (1956)), which I thought was the reason why the the surface free energy is specifically calculated for a special case of U-10Zr as shown as a function of temperature in Fig 10. However, in the porosity calculation (Fig 11), it is unclear which energy (enthalpy or the gibbs free energy) that is used as the value for the surface tension $\gamma$ in Equation 10.

The equations 9 and 10 from Olander [Ref 18] were derived using the assumption of the bubble being in mechanical equilibrium with the solid, and thus utilizes surface tension. This work approximates the surface tension as the surface free energy. The text has been modified to state that it is the surface free energy that is utilized.

1. If in point 1) the free energy is used, I would encourage to perform additional calculations to obtain free energy plot vs temperature (similar to Fig 10) for several higher concentrations of Zr since the internal region of the fuel is Zr-rich. This will make the MD data more complete to be used in future fuel modeling studies.

This has been performed to include entropy and free energy data for both U-10Zr and U-40Zr, in atomic percent, in addition to U-23Zr. These compositions span both potentially high and low Zr concentration that can be reasonably expected in metallic U-Zr fuel. This justification has been included in the manuscript.

1. How does the porosity vs burnup as shown in Fig 11 compare with experimental data? As Fig 11 is presented as a function of $\gamma$ (i.e. parametric study), it would be very useful to discuss which value is suggested for future fuel modeling studies.

The text on page 13 has been modified as follows to illustrate how the surface tension was utilized and how it is affecting future fuel performance modeling:

“Given that a primary factor in fuel pin failure is plenum pressure, which is directly related to fission gas release, obtaining accurate porosity accumulation is critical in predicting accurate fuel evolution under operation. However, surface tension is not the only key input parameter utilized to describe porosity accumulation. The initial assumed bubble number density can dramatically impact the rate of fission gas bubble swelling. It was found that given a computationally determined surface tension, the assumed values for the bubble number density were significantly lower than required to match experimental results. This refinement of fundamental materials properties via atomistic simulations has reduced the inherent uncertainty in the fuel performance simulations and provided a basis for further investigation of key properties.”

1. (minor point), please describe how the void surface area is defined in Eq 4, e.g. is it from the sphere used to remove the atoms to create the void, is it from the actual area formed by the surface atoms, is relaxation taken into account, etc. This would become important when others need to compare their atomistic data in the future.

Void sizes are reported as-created before relaxation. Supercells are generated with approximately equilibrated lattice constants, as such, the void sizes should not change due to cell relaxation. We also did not see any evidence of void shrinkage during the simulations, so the void sizes should be accurate for the relaxed systems, within +/- 1 Å. A sentence noting that as-created void sizes were utilized in the determination of the surface energy.

1. (minor point), it is mentioned in Section 2 that the MEAM potential used in this study is that developed by Moore [ref 11] in which properties were calculated with the DYNAMO code while here the LAMMPS code is employed. Is there any property discrepancy calculated between the two codes?

Exact reproducibility is not achieved between the codes, as has been observed previously for other interatomic potentials. Within statistical error, the results match and there are no observed properties that are significantly different. This has been included in the manuscript.