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 demonstration of surface energy on box size has been included.

- 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 authors for this comment, and have included the following sentence in sectionXX “dsfsdfsfsfssf”

- 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.

I think that clarifying the terms in the equations will suffice to justify this comment, but not sure. Will add some future comments.

- 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.

This plot has been modified to show only 1000 K, 1200 K and 1400 K.

- 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?

need to change to straight lines. verify I am using total energies. perhaps increase # of simulations to reduce scatter.

- 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

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.

Al, can you please provide a sentence in response? If we need to change the verbiage in the manuscript, please let me know and we will do so.

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.

I can do this, and will explore adding these simulations for a 40 atomic percent Zr system.

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.

Perhaps we should add a short discussion that the value of the surface energy actually led to greater deviation from experimental results, which in turn led to a re-evaluation of the number density of bubbles.

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

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.