**Comments from the editors and reviewers:**  
-**Reviewer 1**  
  
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This paper presents new research that determines the grain boundary and surface energies in bcc-U, bcc-Mo, and U-Mo alloys ranging from 600K to 1200K. This paper helps to explain why there is Mo depletion at the bcc U grain boundaries, which is commonly observed experimentally and may contribute to increased fuel swelling during irradiation. Overall, the manuscript is well written. There are only a few comments given below:

Fig.1 seems unnecessary.

Figure 1 has been removed.

In Figs. 12-14 – It is difficult to see all gb orientations. Perhaps slightly modifying the line weight will help to visualize all orientations.

Figures have been modified in an attempt to make it clearer to view all orientations.

In Fig. 17 it can be noted that there is also Mo depletion at the supercell boundaries (y=-90 and +90). What is the cause of this (e.g. are there artificial boundary effects)? It also appears that it is not periodic. This behavior is also exhibited in Fig. 18. How is this depletion explained in both Fig 17 and 18 at the boundaries?

We thank the reviewer for raising this point. In fact, there is a grain boundary at 0 and at +/- 90, with periodic grain boundaries applied to the supercell. More explanation has been included to ensure this fact is communicated clearly and explicitly.

-**Reviewer 2**  
  
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The authors employed molecular dynamics method to study the grain boundary energy and surface energy as a function of Mo concentration and temperature in U-10Mo fuels. One of the interesting results is that the lower grain boundary energy of bcc U, compared to bcc Mo, provides the driving force for Mo depletion at grain boundaries. This driving force diminishes with increasing temperature but is not eliminated. Also, Monte Carlo and MD simulations have been combined to demonstrate active depletion of Mo at grain boundaries. However, these results are expected based on energy criterion. Although some of the results and approaches were described in the previous publications, the manuscript does provide some interesting results. I believe that the studies should be interesting to nuclear materials community and could be published after considering the following suggestions.

-       One of the issues with the current version is that the authors used an energy criterion to derive the driving force for Mo depletion of GBs at high temperatures. It may be important to use free energy or Gibbs energy as the criterion because the entropy becomes important, particularly at high temperatures. This is particularly related to the different configurations of Mo atoms at grain boundaries and in the bulk. The definition of the grain boundary energy at high temperatures needs to be clarified. If the authors calculated the grain boundary energy at 0 K, the same conclusion should be achieved (see Fig. 3 for example).

We agree with the reviewer that the Gibbs free energy is the true determinant of the microstructural changes within this system. We also feel that the potential energy is a primary contributor to the free energy of the system. Although the change in entropy can be determined via integration of the heat capacity of a system [McNutt J Nanopart Res (2014) 16:2365], there is not an established method of determining the magnitude of the entropy of the system from molecular dynamics. Thus, it is very difficult to compare the magnitude of the free energy for different systems at the same temperature (or as a function of temperature). The MC/MD simulations should inherently account for configurational entropy and thus provide some substance to our claim that potential energy is a primary driver of free energy in these systems. Further discussion highlighting the importance of free energy has been included.

-       The authors compared the grain boundary energies at 600 K with that at 0 K in Fig. 3, and noted that the grain boundary energies are slightly lower than those predicted from the ab initio calculations, which was due to the ab initio calculations conducted at 0 K, whereas their work was at a non-zero temperature. Why don’t the authors carry out the calculations at 0 K, and make a proper comparison with the ab initio results?

Calculations are not conducted at lower temperatures due to the restrictions associated with the potentials. The gamma U phase is unstable at low temperatures and thus it is extremely difficult to conduct low temperature (or 0 K) simulations without imposing restrictions on the system that could possibly make the results of the simulation unphysical. Discussion for the choice of temperature has been included in the computational detail section of the manuscript.

-       In a number of places, the authors provided the simulation results, but did not explain why the results should be, as usually did in atomic-level simulations. For example:

Page 12, ‘It can be observed that there is a general trend of increasing grain boundary energy as a function of Mo concentration. This trend for each grain boundary plane is non-linear ….’. Why is this non-linear? Is this due to not enough samplings or are there some physics behind?

We thank the reviewer for their comment. The referenced comment was intended to describe the behavior of the observed trends. As this alloy system displays an asymmetric heat of formation curve, in addition to deviation from Vegard’s law, it was not expected that the trend would be linear. It should be noted that all of the results lie within the lower bound of the general rule of mixtures. The phrase: ”This trend for each grain boundary plane is non-linear, but can be accurately fit with a second order polynomial.” Has been modified to: “The trend for each grain boundary plane can be accurately fit with a second order polynomial.” In order to avoid any implied suggestion that the trend should be linear.

Page 14, ‘The results show that for high Mo content (above approximately 30 atomic percent), grain boundary energy is relatively unchanged as a function of temperature. For low Mo content, the grain boundary energy increases with increasing temperature.’ Please provide an explanation why this should be.

We thank the reviewer for their comments. The following has been added to further explain the results: “This is consistent with the behavior from each individual species, as bcc Mo shows very little variation in grain boundary energy with temperature, and bcc U shows a marked increase in grain boundary energy with increasing temperature. Thus, we would expect that as more Mo is present in the allow, there would exist less and less variance of the grain boundary energy as a function of increasing temperature.”

-       I noticed that the authors used the LAMMPS code to generate grain boundaries, but did not mention if the GBs were properly relaxed. Have the authors compared their GB structures with the previous simulations? I am very much doubt that their GBs had minimum energy structures before rising temperatures. The authors may want to firstly determine the gamma surfaces of these GBs from which the minimum energy structures can be obtained. This is important because it will affect the grain boundary energies with different concentrations of Mo in U-Mo alloys, even the grain boundary energies in pure U and Mo crystals.

The grain boundary structures for (210), (310) and (410) symmetric tilts were compared to literature results for bcc Fe, Mo and Ta and found to be in concordance with the observed relaxed structure. The grain boundary structures were relaxed at the target temperatures. Particle motion is greatly enhanced at high temperatures and simulations were equilibrated for a long enough time to ensure the reorganization of the grain boundary to the low energy structure.

Structures were verified against results from Tschopp [PRB 85, 064108 (2012)], Morita [MSE A234-236 (1997) 1053-1056] and Hahn [Scripta Materialia 116 (2016) 108–111].

-       Another important issue is how the simulations can be compared with experimental results. In addition to the depletion of Mo at grain boundaries, the recent experiments showed that when a homogenized U-10Mo alloy was subjected to annealing at 500 oC, discontinuous precipitation occurred along g-UMo grain boundaries forming lamellar products of Mo-enriched g-UMo and a-U, as early as within 1 hr of annealing. On longer term annealing, the discontinuous precipitation appeared to grow into the interior of the grains. These results demonstrated that the GBs is simply not depleted by Mo, but the local region at GBs could be enriched by Mo.

Thanks to the reviewer for bringing recent experimental facts to our attention. The phenomena mentioned in the comment, we would argue, is a likely thermodynamic step taken from the conclusions within this paper. The grain boundary precipitation observed is a direct result of the Mo depletion we predicted from thermodynamic point of view. Lower Mo content increases the likelihood for alpha U to form, either forming alpha U and U2Mo regions, or alpha U-which has essentially zero Mo solubility- forming and forcing the excess Mo from the parent phase into the surrounding gamma UMo. This work is intended as a bridging step to move from pure gamma UMo, to depleted Mo at grain boundaries inducing phase decomposition.