**Evaluation of the anisotropic grain boundaries and surfaces of α-U via molecular dynamics**

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We thank the reviewers for their comments, and we feel the paper has been strengthened and has gained in clarity due to their efforts. Comments to their revision requests are given below in blue.

Reviewer #1:

 Khadija et al. have submitted the manuscript entitled "Evaluation of the anisotropic grain boundaries and surfaces of α-U via molecular dynamics". This paper systematically studied the GB anisotropy of α-U by calculating and GB energies, surface energies, and work of separation. Such energetic calculations appear to be new for this materials and the rich computational data is collected. However, this paper may have some issues that required major revisions.

1) Equation 1 is only applicable to zero K. At elevated temperature, equation 1 may be wrong for calculating interfacial energy because entropy term cannot be ignored (Temp \* entropy is not zero anymore!). Authors must address the entropy effect and how it changed the interfacial energy. Without the discussion regarding to entropy effect, all the results about the effect of temperature on energetic calculations are quite questionable.

Thank you for your comment. You are correct in that the ‘interfacial energy’ used in equation 1, is the internal energy (potential energy plus kinetic energy) not the free energy. Considering that the temperatures are relatively low (below 600 K), and Beeler et al. (please cite my paper looking at surface energies in UZr) have shown that in similar metallic systems, and at higher temperatures, the effect of entropy on grain boundary energies will be on the order of 10 mJ/m2-K, the effect of entropy can be reasonably neglected here. Considering entropic effects would not affect conclusions or qualitative relationships within this work. Discussion along these lines has been added into the computational details section.

2) In Figure 4, why the GB anisotropy is considered as a function of GB type? This figure only shows the A\_type GB. The author should elaborate further on it.

Thank you for your valuable comment. In equation 3, it is noted that, ‘A\_type’ is the nomenclature for the GB anisotropy, not the ‘type A STGB’. We will change this confusing nomenclature. GB anisotropy is considered as a function of GB type to illustrate that GB energy in alpha U depends not only on misorientation angle but also in tilt plane and shear plane of GBs.. We have updated nomenclature for equation 3 and 4.

3) In page 11, author claimed that "Thu current work indicates that at lower temperature, there is less driving force for grain growth due to the lower grain boundary energy". Indeed, it is well known that the slow grain growth at low temperature can be also ascribed to slow diffusion of grain boundary. Hence, it is not persuasive to demonstrate that the slow grain growth is due to low GB energy.

We agree with the reasoning of slow grain growth by slow diffusion of grain boundary. But grain boundary migration is also a function of the driving force, and curvature driving force is proportional to the GB energy. So, at lower temperatures, due to the low GB energy, there will be less driving force for grain growth. A revision reflecting that slow GB diffusion also contributes to slower grain growth has been included.

4) In page 11, author mentioned stable fine-grained microstructure may hinder dislocation movement and lead to lower ductility. I suggest author to perform tensile study for some representative GBs (e.g., low GB energy vs. high GB energy) and analyze how these GBs response with external loadings at different temperatures. Otherwise, it is hard to understand the GB with low energy can lower ductility at lower temperature.

Thank you for your insight. As per existing literature, there must be some correlation between GB energy and elastic modulus of the bi-crystal system. The reason behind the current statement is, the low GB energy of STGB can potentially lead to a finer microstructure, with GBs acting as stress concentrators and dislocation motion inhibitors, which leads to lower ductility. This is effectively an extrapolation towards a nano-grained structure material, which can exhibit brittleness. Our goal here is not to determine the elastic properties of different bi-crystal or polycrystalline systems along with their temperature dependence. In order to thoroughly perform tensile testing in a bi-crystal system in alpha U, a significant amount of additional work would need to be performed. This includes tensile testing on single crystal alpha U via MD methods, of which no such studies have been performed. However, such efforts are the subject of future work. The authors would prefer to perform a thorough study on computationally determined stress-strain curves in alpha U, rather than a cursory study.

5) The legends labelled in some figures are too similar, e.g., Figure 5, which is hard to discern difference between them. I suggest using different line styles, colors, and maker styles to optimize all figures.

Thank you for your suggestion. We have updated the figures in the manuscript

6) Please show the crystal structures of A, B, and C type GB in main text. The Fig. 10 in appendix is hard to understand.

Thank you for your suggestion. Figure 10 has been updated.

Reviewer #2:

The article presents an interesting study of planar defects (surface and grain boundaries) in uranium. The investigation was based on classical atomistic simulation. The obtained results may be considered as the first attempt to shed some light on the properties of planar defects in alpha-uranium. The strong advantage of the work is clear description of the obtained results. However, several points need to be addressed:

1. The main reason for a concern is the used method of GB preparation. Here, I can point to three different possible sources of inaccuracy at GB simulation:  
A) First of all, the authors simulated strongly deformed structure in the bulk part of the calculation cell due to non-equilibrium lattice parameters. At the same time, they used equilibrium value of cohesion energy E0 in eq. (1). It should result in some error at the calculation of GB energy.  
B) The authors did not describe the minimization procedure for search of the equilibrium configuration of GB. The most common way is the "gamma-surface" method where one grain (i.e. half of the simulated system) is shifted along the GB plane in search of the position with the minimal energy. If the authors used such procedure, they should describe it.  
C) At consideration of some GB, "gamma-surface" method cannot give equilibrium ordered structure due to formation of defects inside the created GB. This happens when the average atomic density near the GB is different from the bulk. One of the possible methods to find the equilibrium structure of GB is the evolutionary algorithm [T. Frolov, W. Setyawan et al. Grain boundary phases in bcc metals, Nanoscale (2018)]. The other way is MD annealing with the open surface that should be used for creation of the equilibrium GB structure. The use of the open surface in such MD simulation is mandatory aspect as this surface is necessary for formation/removal of GB defects (for instance, see [T. Frolov, Y. Mishin, Phys. Rev. B 79 (2009)] or [S. Starikov, M Mrovec, R Drautz, Acta Materialia 188 (2020)]).  
Based on the comments given above I strongly recommend the authors to take a step back and to perform the validation of the GB construction method (at least, for several types of GB).

Thank you for your valuable comments:

1. It is correct that in order to construct the appropriate geometry for the GB system, we have utilized non-equilibrium lattice constants. In the determination of E0 in eqn 1 (energy of the pristine crystal) we have considered equilibrated lattice constant. However, sufficient relaxation of the system is performed to ensure equilibrated systems, with equilibrium lattice constants, for the analysis of stable grain boundaries. Due to the utilization of an NPT ensemble with periodic boundaries and independent relaxation of each lattice vector, the imposed stresses from the simulation setup are rapidly relaxed. Individual unit cells from the grain interior were analyzed to ensure equilibrium lattice constants were present. This information has been included in the manuscript.
2. We did not utilize ‘gamma surface’ method, as at temperature above 0 K, the gamma surface method does not always predict the ground state (Frolov *et. al.*2018). Period boundary calcul
3. In order to validate the GB construction, and per the reviewer’s suggestion, we have utilized another method (Frolov *et. al.*2018, Zhu *et. al.* 2018) named as method 2, for comparison. The method we have already utilized in our manuscript is named as method 1. Comparison in grain boundary energy between these two methods are shown in figure 1. The discrepancies in GB energy of normal STGBs between two methods are not significant (maximum by 10 %). Moreover, GB energy of the preferred twin GB shows a lower GB energy in method 1. This evaluation has led us to re-review all of the GB and surfaces studied in the manuscript in order to verify the symmetry and periodicity of our generated GBs. Through this process, we have found four mistakes, and we have corrected such errors in the manuscript. This discussion has been included in the manuscript, and we feel increases the credibility of this work. We thank again the reviewer for their suggestions.

Figure 1: comparison in GB energy calculated by method 1 and method 2

2. The authors used the potential (ADP) developed in 2015. Are there some special reasons explaining why the authors did not use more recent version of this potential [S. Starikov, L. Kolotova, A. Kuksin et al. JNM (2018)]?

Thank you for pointing out this. We have utilized the ADP developed in 2018 (S. Starikov, L. Kolotova, A. Kuksin, D. Smirnova, V. Tseplyaev, Atomistic simulation of cubic and tetragonal phases of u-mo alloy: Structure and thermodynamic properties, Journal of Nuclear Materials 499 (2018)). Unfortunately, we put the wrong reference and we apologize for that. This has been updated.

3. It would be interesting to add more detailed discussion about twins. There are several experimental works about twinning in alpha-uranium [18-20]. The authors only mentioned these works in the Introduction without detailed comparison with the simulation results in the main part of the work. I guess such comparison may add value to this work.

Thank you for your valuable feedback. Unfortunately, the GBs we have studied here, do not describe the experimentally observed twins. Hence, we could only provide an insight on some new probable twins, rather than an explicit comparison. But we will study additional temperature effect on those twins (observed from the current work).   
  
References:

1. Nanoscale, 2018, 10, 8253
2. Zhu, Q., Samanta, A., Li, B. *et al.* Predicting phase behavior of grain boundaries with evolutionary search and machine learning. *Nat Commun* **9,**467 (2018). https://doi.org/10.1038/s41467-018-02937-2