We thank the reviewers for their comments. The comments helped identify and rectify the shortcomings of the initially submitted paper.

We have now redefined GBs in a way that should address the concerns the reviewers had. Instead of only using mobile GB atoms for MSD calculation, we now take into account all the atoms in the GB region. Consequently, all the results have changed as well, albeit without significantly affecting the conclusion of the manuscript. The new figures and tables reflect this change.

Another major change was made in the reporting of the GB diffusion coefficients. Due to the presence of anisotropic diffusion, diffusion coefficients parallel and perpendicular to the tilt axis are now reported separately throughout the article. This avoids the issue of dimensionality in the diffusion coefficient calculations of some GBs. Also, the ratio of the parallel diffusion coefficients to the perpendicular diffusion coefficients for U, Mo, and Xe are included to enrich the discussion of anisotropy.

We have also noticed that the term "total diffusivity" was not defined in the article. It was used to mean species-agnostic diffusion of the GB. We have modified our discussions to only refer to the diffusion of individual species. Also, other types of diffusion coefficients including multiple species, such as binary diffusion coefficients, can easily be computed using the species GB diffusion coefficients reported in the article.

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Reviewer #1: This work evaluates the self-diffusion behavior of U, Mo, and Xe along different types of GBs. Both temperature and composition are considered in the evaluation. Very nice description and discussion of results.

\* One major issue that needs to be clarified and discussed is the definition of GB atoms. It looks like the GB atoms are chosen based on the MSD at different temperatures. In the discussion part, the authors mentioned that, given the criterion explained in Figure 3, some of the real GB atoms (atoms in the GB region) may not be selected to be GB atoms as not all atoms in the GB migrate. Therefore, are the real GB atoms underestimated? How does that factor change the quantitative results? More importantly, when evaluating the MSD for GB atoms, these atoms that don't move much should also be considered, since using Einstien's equation implies a probabilistic process (statistically, it does not mean all GB atoms have to move a lot). The authors should clarify this aspect and make corrections as necessary.

GB atoms are redefined to not exclude any static atom in the GB region. GB regions are now defined as rectangular cuboid zones that include lattice points participating in the diffusion process. (Page 4; Lines 111 - 127)

\* In alignment with the definition of effective GB width, Figure 4 is suggested to change to width versus inverse of temperature.

Figure 4 is updated according to the new definition. GB width is plotted against temperature (not inverse temperature) to showcase the linear correlation between them. (Page 7)

\* In evaluating Xe diffusion, since there are just 2 Xe atoms, the authors need to check whether Xe migrates to bulk during the simulation, which makes the diffusivity small. Is there any experimental data for comparison?

Xe atoms are observed not to migrate to the bulk. It might be because of the Xe atoms' strong affinity to the GBs. To the authors' knowledge, there is no experimental data for comparison. Prior molecular dynamics simulations have observed that Xe diffusion requires additional vacancies and proceeds extremely slowly (<https://doi.org/10.1016/j.jnucmat.2022.154137>). Thus, no Xe diffusion into the bulk is expected, and none is observed.

\* Page 13, it is noted that 1D pipe diffusion occurs for some of the GBs. How does this affect the conclusion since the dimensionality in Eq. 1 becomes 1 instead of 2?

Diffusion coefficients are now reported perpendicular or parallel to the tilt axis. It solves the problem with the dimensionality. Since assuming a dimensionality of 2 is equivalent to the assumption that GB diffusion is isotropic, the authors now report diffusion coefficients in directions perpendicular and parallel to the tilt axis of the GB plane. (Page 5; Lines 137 - 144)

\* Figure 6, is this total diffusivity? Should be mentioned in the caption.

Any discussion of total diffusivity has been omitted and is now species-specific.

\* Page 10, line 27, is there a typo "prevent comparsion"?

The phrase “prevent a comparison” has been changed to “prevent comparison”. (Page 11; Line 231)

\* Page 10, 2nd paragraph, the "high temperatures" is vague, and should be provided rough temperature values.

Specific temperature values are included to delineate low temperatures (600 K - 800 K) and high temperatures (900 K - 1200 K). (Pages 11 - 13; Lines 224 - 242)

\* Page 10, line 42, "GB diffusion is more important in the calculation of the effective diffusivity of the material." This claim could have more elaboration, by incorporating the width information in the first step and typical grain size.

The claim has been elaborated with a sample calculation incorporating GB width information and typical grain size. (Page 13; Lines 243 - 258)

\* Page 10, line 43, "The diffusion enhancement starts to wane at higher temperatures since the activation energy of GB diffusion is lower than that of bulk diffusion." May remove "since …".

The clause "since the activation energy of GB diffusion is lower than that of bulk diffusion" has been removed. (Page 13; Line 255)

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Reviewer #2: The authors use molecular dynamics simulations to calculate the grain boundary diffusion coefficients of U, Mo, Xe, and total species in BCC U-Mo alloys at different Mo concentrations, grain boundary types, and temperatures. The results may be used as input for mesoscale modeling of microstructural evolution in U-Mo fuels. Overall, I think this work is well executed and deserves the eventual publication at the Journal of Nuclear Materials. Below are my minor suggestions for improving this manuscript.

1. Page 2, please define "DART".

“DART” has been changed to “the Dispersion Analysis Research Tool” and is appropriately cited. (Page 2; Line 38)

2. Figure 1, I suggest labeling X, Y, and Z in the figure.

Figures are now labeled with coordinates. (Page 3)

3. Page 3, I suggest specifying U-Mo weight percentage together with atomic percentage, as this is an atomistic modeling.

Atomic percentages are now reported for reference. (Page 3; Lines 75 - 76)

4. Page 4, the authors used a time step of 5fs for Xe diffusion. This is extremely large for a typical MD simulation, although I understand the authors try to reach a simulation time of 100 ns. I suggest a brief discussion about the stability of the simulation using this large time step.

We verified the stability by running two systems for the same simulation time but with different timesteps (2 fs and 5 fs). It is observed that the effect of a 5 fs timestep is minimal on the movement of the Xe. A discussion is included in the computational details section. (Page 4; Line 108 - 110)

5. Page 4, in the Method section, grain boundary atoms are not defined. I suggest the authors to make a note that the definition is in Section 3.2.

GB definition is now provided in the Method section. (Page 4; Lines 111 - 127)

6. Page 4, the author should mention how to construct a reference system for calculating the grain boundary energies. The distribution of Mo atoms in a bulk U-Mo alloy could affect the reference energy by a few eV, which could be large enough to affect the grain boundary energy.

A discussion about the reference system is included in the Method section. 25 simulations with different random number seeds are performed to get the average reference energy. Different random number seeds account for different distributions of Mo atoms in the system. (Page 4; Lines 86 - 87)

7. Section 3.2, the authors used a temperature-dependent grain boundary width (d) to define grain boundary atoms. In literature, many studies use P = D\_gb \* d to minimize the grain boundary width effect. The authors may briefly discuss this.

The double product P = D\_gb \* d is discussed in section 3.2. Also, the difference between diffusional width and structural width of GBs is discussed briefly. (Page 7; Lines 162 - 166)

8. Section 3.2, at high temperatures, grain boundary migration could occur, which can affect the boundary width according to the authors' definition. A brief discussion about this could be beneficial.

GB migration is discussed in the method section. The authors have observed GB migration in only a handful of simulations at high temperatures. The simulations showing GB migration are not used in the diffusivity calculations. (Page 4; Lines 123 - 127)

9. The units should be clearly specified in all table captions.

Units are now specified in all table captions.

10. Page 9, In the sentence "the total GB diffusion coefficients for different tilts and temperatures ……… Tables A1 and A3", but there is no temperature in the two tables.

“and temperatures” has been omitted. (Pages 20 - 21)

11. Table 3, "orientation-averaged" should be mentioned in the caption.

"Orientation-averaged" is now mentioned in the caption of Table 3. (Page 11)

12. Figure 8, the grain boundary diffusivity might have a positive correlation with the grain boundary energy (Figure 2), although the uncertainty is large. The authors might mention that if it is the case.

The positive correlation between GB diffusivity and GB energy at low temperatures is mentioned. A table has also been added in the section to report GB energies at 600 K and 1200 K from the literature. (Page 14; Lines 270 - 277)

13. Page 13, I think the statement of "a pure tile GB is essentially an array of parallel edge dislocations" is not accurate. This should be only valid for low-angle tilt boundaries.

The sentence has been modified to indicate that low-angle tilt boundaries can be thought of as arrays of parallel edge dislocations. (Page 15; Line 294)

14. Figure 9, I don't know how the authors calculated the MSD for the low-angle boundaries. Did they ignore the grain boundary atoms that do not have large MSD? In my opinion, they should be included as long as they are within the boundary thickness d.

In our previous definition, we did ignore GB atoms that did not make at least one lattice point jump. So, the MSD of only the mobile GB atoms was calculated before. The definition of GB atoms has been changed to include all atoms within the boundary thickness. (Page 4; Lines 111 - 127)

15. In Appendix, the numerical format should be consistent with the main text, e.g., e-10 should be 10^-10.

The numerical format in the appendix has been changed to make it consistent with the main text. (Pages 20 - 21)