**The authors would like to thank the reviewers for their time, effort, and thoughtful comments and suggestions. Apart from the reviewers’ comments, here are few things changed:**

* **The self-diffusion coefficients of U and Mo atoms were recalculated. The number of atoms (U and Mo) was multiplied into the mean square displacements to calculate the self-diffusion coefficients of U and Mo atoms. The interdiffusion coefficients were also recalculated using the newly calculated self-diffusion coefficients.**
* **In table 2, the ratios of the Mo self-diffusion coefficients to the U self-diffusion coefficients were added.**
* **Table 3 and table 4 were recalculated.**
* **In figure 5(d), the experimental results (interdiffusion coefficients) were removed. Instead, the experimental results were compared to the interdiffusion coefficients calculated in this work in figure 6.**

**Reviewer #1: The manuscript, "An atomistic study of defect energetics and diffusion with respect to composition and temperature in γU-Mo and γU-Mo alloys," calculates vacancy and self-interstitial formation energies, and self-diffusion coefficients using interatomic potentials. This work presents a lot of calculations, but is missing the organization. The paper reads more as a report then actually providing physical insight into their calculations. I recommend that the authors provide a more cohesive flow to the paper and provide more discussion on comparing the two systems. This work is potentially publishable and so I recommend major revisions. Below are my comments.**

**1. The authors' state, "high performance research," in line 3 and I would recommend to the authors to further explain or change verbiage as this is too vague.**

**Response:** More explanation is added as follows in lines [2-7]: “Uranium-molybdenum (U-Mo) alloys are considered as one possible candidate for nuclear fuels for future fast reactors, as well as high-performance research and test reactors (HPRRs) [1, 2, 3]. For example, HPRRs such as the Advanced Test Reactor at Idaho National Laboratory and the High-Flux Isotope Reactor at Oak Ridge National Laboratory, require a higher uranium density (> 6.5 g/cc) due to flux requirements, as compared to other research reactors.”

**2. Lines 9-10 has an awkward sentence structure, as in is the "isotropic swelling behavior" is more evident in αU than γU and by how much?**

**Response:** “Isotropic swelling behavior” is more evident in γU due to its crystal structure (bcc) as opposed to αU. This is due to αU, having an anisotropic crystal structure (orthorhombic), which shows anisotropic swelling behavior. In addition, the magnitude of the swelling depends on a number of the in-pile characteristics, and the crystal structure is merely one of the factors affecting swelling behavior. The sentences are edited for clarification as follows in lines [10-14]: “During reactor operation, pure αU fuel exhibits anisotropic growth and anisotropic swelling under irradiation, which has historically limited the usage of unalloyed αU [5, 6]. On the other hand, γU exhibits isotropic swelling behavior, therefore displays comparatively better irradiation performance than αU [6,7].”

**3. Lines 31-39 is motivated by studies in αU and then discussions of O impurities and I do not understand how this is related to γU and the authors do not provide the numbered value from Lund et al.**

**Response:** The data reviewed focused on metallic U with an emphasis on γU. The findings by Lund et al. [18,19] were discussed since they provided insight into the variability observed within the experimental studies, showing that oxygen suppressed the vacancy formation energy in U. The authors believe that the variation on the vacancy formation energy in γU could be the same reason. We have clarified this section to state in lines [34-38], “Lund et al. [18, 19] attributed the variation in experimentally determined vacancy formation energies to differing specimen geometries, which contained varying oxygen (O) impurity concentrations. This was further supported by first-principles calculations in αU, which showed that O defects reduced vacancy formation energies [19].”

**4. At the end of line 42, should you add "respectively?" Why is there the .30 eV difference between exchange-correlation functional?**

**Response:** The sentence does not mean that 1.08 eV is from the PW91 and 1.38 eV is from the PBE, respectively. Rather, it means that the three vacancy formation energy values ranged between 1.08 eV and 1.38 eV [20. 21], and they were obtained using the two different exchange functionals. Specifically, 1.08 eV was obtained using the PW91 [20]/ 1.32 eV was obtained using the PW91 [21]/1.38 eV was obtained using the PBE [21] as represented in Table 1. Thus, “respectively” should not be added in the text. Different exchange-correlation functionals produce different results. Also, the discrepancy of 0.3 eV is due not only to the different exchange-correlation functionals, but also the different methodologies. A brief explanation regarding the 0.3 eV difference is added as follows in lines [48-49]: “The discrepancy of 0.3 eV can be attributed to the use of different exchange-correlation functionals, as well as different methodologies.”

**5. Line 44-46, I thought that the <110> interstitial are more favorable in bcc metals and <100> for fcc metals?**

**Response:** That is usually true for the bcc materials where bcc is the ground state structure. In this case, the bcc phase is a high-temperature structure, which is unstable at lower temperatures. Defect behaviors have been observed to be unique in these types of systems. However, the self-interstitial formation energies of the <100> dumbbell and the <110> dumbbell are comparable (only vary by 0.05 eV) from γU based on first-principles calculations [21]. Since both are much less than 1 eV, both <100> dumbbell and <110> dumbbell are favorable in γU.

**6. Line 69, What is the accuracy and confidence in the U-Mo-Xe EAM potential?**

**Response:** The interatomic potentials describe the interaction between atoms, and thus they are utilized to predict the properties of material. Thus, the accuracy and confidence in the potential cannot be discussed numerically. More explanation on the accuracy and confidence in the potential is added in lines [80-82] as follows: “This potential was selected since it has successfully reproduced characteristic material properties, such as lattice parameter, Young's modulus, room-temperature density and melting temperature of U-10Mo alloys [37].”

**7. For the γU-Mo systems, as discussed in Line 75-77, what are the positions of the Mo in the lattice? Are the Mo-Mo repulsive?**

**Response:** U atoms were replaced with Mo atoms at randomly distributed lattice sites, generating a solid substitutional alloy. Regarding the repulsion of Mo-Mo, the nearest environment neighbor was not considered in the construction of the random substitutional solid solution alloys. More explanation regarding position of Mo atoms is added as follows in lines [91-92]: “The U atoms were replaced with Mo atoms at randomly distributed lattice sites, generating a solid substitutional alloy.”

**8. Line 79, what was the motivation for the elevated temperature?**

**Response:** The elevated temperature regime was evaluated since U-Mo alloys are one promising candidate for future fast reactors which its operating temperature is above 500 oC. In addition, diffusion coefficients in γU-xMo calculated in this work can be compared to the existing experimental results. A brief motivation for both the low and elevated temperature range is added as follows in lines [95-97]: “The low and high temperature ranges correspond to the operating temperatures for research reactors and fast reactors, respectively.”

**9. Line 82, what does equilibrium mean? Equilibrium in temperature, locations of atoms?**

**Response:** It means equilibrium of the system (simulation box) with respect to energy. More explanation on equilibrium is added as follows in lines [100-102]: “Equilibrium of the system was confirmed by investigating the potential energy of the system as a function of time. The potential energy stayed nearly constant after 10 ps, indicating that the system reached equilibrium at this time.”

**10. Line 84, is it only one defect then why the large supercell? Or is it one defect type? This would imply a defect concentration. Did you consider both U and Mo defects? The authors seem to answer this question in Line 112-114, but I would recommend them to include this earlier to avoid confusion.**

**Response:** A single point defect (vacancy or interstitial) was created in a large super cell to calculate the point defect formation energy. Both U and Mo were considered as a defect. Line 112-114 moved into line [104-108] as recommended. More explanation regarding why the large supercell is used is added as follows in lines [87-89]: “A large system was utilized to approximate the random substitutional solid solution alloys and thus avoided the interaction of the point defects with periodic boundary conditions.”

**11. Line 103, what was the reference crystal structure for the Mo?**

**Response:** More explanation on the reference crystal structure for Mo is added as follows in lines [127-128]: “Note that the crystal structure of Mo is also bcc.”

**12. Line 128, what about thermal defects?**

**Response:** Thermal effects are included in the equations 11 and 12 ( . The equilibrium defect concentration increases with increasing temperature.

**13. Line 246-247, what was the equilibrium defect concentration? Can you estimate this from your calculations?**

**Response:** The equilibrium defect concentration was calculated as a function of temperature and composition.The equilibrium vacancy and self-interstitial concentration in U-12Mo at 1200 K are now provided in the text as an example. The added sentence is as follows in lines [273-275]: “The equilibrium concentrations of vacancies and self-interstitials were found to be 2.4 x 10-5 and 2.5 x 10-3 in γU-12Mo at 1200 K, respectively, as an example.”

**14. The authors could provide some further insight into why the self-diffusion decreases with increasing Mo concentration and why their calculated values are less than experimental results (Line 261-262).**

**Response:** More explanation is added, and the paragraph was reorganized as follows in lines [333-340]: “Since the self-interstitial formation energies were lower than the vacancy formation energies in γU-xMo at the investigated temperatures, the self-diffusion coefficients were dominated by the self-interstitial terms in Eq. (9) and (10). An exponentially decreased equilibrium concentration of self-interstitials, which is primarily attributed to an increase in self-interstitial formation energies, resulted in a decrease in self-diffusion coefficients with increasing Mo concentration. The vacancy terms in equations (9) and (10) were found to be negligible due to the higher vacancy formation energies, which reduced the concentration of vacancies exponentially.”

In addition, the discrepancy between experimental results and computational results could be due to the interatomic potential used and short time scale in molecular dynamics simulations. The possible reasons about why the calculated diffusion coefficients are less than the experimental results are added as follows in line [301-304]: “The discrepancy between experimental results and computational results could be due to a number of factors, including the interatomic potential and the comparatively short time scale of molecular dynamics simulations.”

**15. The discussion is not very clear or organized in the first paragraph (Line 275-289). The thoughts are not cohesive. They begin with vacancy formation energies, then discuss mixing enthalpy, then jumps back to Mo vacancies, and finishes with self-interstitial formation energy. I would highly recommend to rewrite this paragraph.**

**Response:** The paragraph was re-written in lines [305-319] as follows: “The vacancy formation energy in γU-xMo decreased with increasing Mo concentration. Note that the vacancy formation energies in pure Mo, obtained via PAS and first-principles calculations, ranged from 1.6 eV to 3.6 eV, and tend to be greater than that of γU [61, 62, 63, 64, 65, 66]. This seems to indicate that the bond energy between U and Mo atoms is lower than the bond energy between U and U atoms. Thus, it might be easier to create a vacancy if more Mo neighbors exist in the alloy. The mixing enthalpy calculated both experimentally and computationally support this proposed hypothesis. The mixing enthalpy in γU-Mo was found to be positive over the whole composition range at 1000 K using MD simulations [67]. The positive mixing enthalpy in γU-Mo was also experimentally observed when the concentration of Mo was greater than 7 wt.% at 1100 K [68]. The mixing enthalpy is positive when the internal energy following mixing is greater than the internal energy prior to the mixing. This indicates that the U-Mo bond is weaker than the U-U bond, and thus the U-Mo bond is not favorable. Vacancy formation energies in alloys are dependent on the types of atoms present and their concentrations surrounding a vacancy [69, 70, 71, 72, 73].

**16. Line 305 Reference is needed.**

**Response:** The reference is added in lines [339-340] as follows:“The interdiffusion coefficients in γU-xMo, calculated using the self-diffusion coefficients in this work, compares well to previous observations [59].”

**17. Figure 3 could the authors comment on the scattered in the data due to the different potentials? Is one more accurate (or better) over the other?**

**Response:** More explanation on the scattered data is added in lines [232-234] as follows: “Since each interatomic potential has different parameters that are more or less accurate compared to the experimental data and the different range of applicability, the entire scope of data for different potentials was included.” This has also been addressed via the responses provided in part to comments 6 and 14.

**18. The authors don't touch on the anomalous self-diffusion in γ-U and this does deserve a discussion in the introduction.**

**Response:** A brief discussion is added in lines [53-55] as follows: “In addition, an anomalously high self-diffusion coefficient was observed in γU when compared to other bcc metals, which is attributed to the dominance of self-interstitials in the diffusion process [24, 25, 26, 27].”

**Reviewer #2: In this work the authors use EAM-potential MD to study vacancy and self-interstitial thermokinetics in the γU-Mox binary system as a function of temperature and Mo concentration. Key formation and diffusion parameters are provided and compared against known theoretical and experimental trends. These contributions are valuable for higher time/length scale models and understanding possible materials degradation processes. There are a few (likely minor) points that should be addressed before publication:**  
  
**(1) Comparison to binary phase diagrams could be potentially useful given the random alloy approximation applied. While no additional phases were observed based on the authors' analysis, this could be an artifact of sampling statistics (cell size, simulation length, number of simulations, etc.). In particular, diffusion properties of U vs. Mo as discussed in lines 237-245 would seem to be consistent with clustering of Mo. Some analysis of clustering dynamics (e.g., Mo-Mo RDF as a function of time) should easily demonstrate whether or not Mo clustering is occurring. If it is, that may well alter interpretations.**

**Response:** As the reviewer commented, it is possible that local Mo clustering is occurring in the alloy, especially at low temperature regime (400 K - 800 K). However, experimentally, the γ-phase is quenched in during the manufacturing process, and the γ-phase has been observed to be stabilized under irradiation at low temperatures (400 K - 800 K) as a substitutional solid solution alloy. Even though we are investigating low temperatures where other phases are present by the phase diagram, we are introducing defects, and assuming that we have random substitutional solid solution alloys (γ-phase) with defects, which matches the experimental observations. The clustering of Mo was not observed in the MD time scale based on the RDF analysis as shown in Fig.1.

Chart, line chart

Description automatically generated

**(2) In Figure 4/Ref. [25]/lines 228-230, there is a very large discrepancy in calculated self-interstitial formation energy for the γU-Mo10 system at 400K between this study and a previous study using the same potential (~0.32 eV vs 1.1 eV). It is stated that there is qualitative agreement but assuming x=9,10 behave similarly, Refs. [24] and [25] would seem to indicate self-interstitial formation energy drops significantly with increasing temperature which is the opposite trend to what is reported in Figure 4a. Is this a mistake or is there really a qualitative difference? If the later, it is important to note instead of saying there is qualitative agreement.**

**Response:** There is qualitative agreement in the sense that the self-interstitial formation energy was lower than the vacancy formation energy in the studies. A discrepancy in the self-interstitial formation energy between the current work and [28] may come from the different methodologies. Both U and Mo atoms were considered as a defect for both works. However, in [28], the number of simulations that a U atom was introduced was proportional to the concentration of U atoms in γU-xMo. On the other hand, in the present work, the number of simulations that a U atom was introduced as the defect was the same as the number of simulations that a Mo atom was introduced. In addition, since [26] and [28] evaluated the point defect formation energy at single temperature with different methodology, it is hard to conclude that the self-interstitial formation energy decreases with increasing temperature. The current work is the first study investigating the point defects properties in γU-xMo as a function of composition and temperature. More explanation on the discrepancy in the self-interstitial energy is added in [251-256] as follows: “A discrepancy in the self-interstitial formation energy between the current work and Beeler’s work [28] could originate from the different methodology. In Beeler’s work [28], the number of simulations that a U atom was introduced was proportional to the concentration of U atoms in γU-xMo. On the other hand, in the present work, the number of simulations that a U atom was introduced as the defect was the same as the number of simulations that a Mo atom was introduced.