The authors would like to thank the reviewers for their thorough reading and insightful comments. All the comments have been very useful, and by addressing them the paper has been greatly improved. In what follows all the comments are addressed.

Editor and Reviewer Comments

Reviewer #1: This paper is overall well-written. It contains a solid examination of the two potentials for UN. Various aspects are discussed. There are some important details and discussions that should be added.

1. The comparison should include what has been done using this potential. For example, the author calculated the thermal expansion, elastic constants, and some other properties. Are some of these already calculated in previous work developing and/or applying this potential? Are the results comparable or why recompute them?

The thermal expansion coefficient as a function of temperature had not been calculated by the updated version of the Tspelyaev and Starikov potential that we use in this study prior to submission. It was calculated by Li and Murphy using the Tseplyaev potential, but they used a different definition than the one used by Hayes et al. to report experimental thermal expansion coefficients. Since submission, one of the co-authors, Galvin et al. (<https://doi.org/10.1063/5.0177315>) have published thermal expansion. Their work is now cited here, as well as in the section on heat capacities.

The elastic constants as a function of temperature were calculated by Kocevski et al. using their potential but they assumed an isotropic material. That’s why we recalculated the elastic constants using both potentials for consistency. To make this clear, the 6th paragraph of the introduction was updated as follows: “Kocevski et al. [...] developed an embedded-atom method (EAM) potential fitted to experimental thermal expansion and single crystal elastic constants, as well as formation energies of stoichiometric point defects from DFT calculations. The potential was used to calculate the elastic properties and temperature-dependent heat capacity. However, the authors assumed UN to be an isotropic material, which is not true as will be explained in Sec. …, and the heat capacity was underestimated at high temperatures. Also, the potential was used to study Xe diffusion in UN.”

2. “Kurosaki et al. mention the values of potential parameters without units, which makes it hard to further assess its behavior.” Why not get in contact with the original authors for confirmation?

We reached out to the corresponding author, but they did not reply.

3. “Li and Murphy [26] have later used this potential to study self-diffusion in hypo-stoichiometric UN” Does this paper share the same conclusion regarding the stoichiometric issue? Discussion would be helpful.

By stating “the stoichiometric issue” we assume the reviewer means the inability of the Kocevski potential to describe U-rich systems. If that is not the case, we would ask the reviewer to please clarify.

The manuscript by Li and Murphy only explored hypo-stoichiometric UN, and only used the Tseplyaev potential. Since the Tseplyaev potential is capable of describing both hyper- and hypo-stoichiometric systems, there should not be any evident issues that would present. Also, the paper by Li and Murphy does not elaborate on the formation energies of defects, but only on lattice constants and diffusion. Thus, we are not able to compare our results/conclusions to their findings.

4. Since 𝛼- and 𝛽-U2N3 are stable at different temperature ranges, it is not clear why the calculations consider the ranges when they are not stable. Elaboration is necessary.

Each phase was explored over a temperature range to identify whether the temperature may stabilize certain phases or to identify trends in stability. 𝛼-U2N3 was only considered within its stability range (i.e., 0-1400 K). 𝛽-U2N3 was examined from 0 K to 1600 K to examine the stability ranges of this phase, because the temperature range over which 𝛽-U2N3 is stable is quite uncertain: the phase diagram in the literature review by Uno *et al.* implies that 𝛽-U2N3 is stable in the range 1324-1648 K, whereas that by Okamoto implies that 𝛽-U2N3 is stable in the range 1214-1625 K. Also, we wanted to examine if the potential can stabilize 𝛽-U2N3 at lower temperatures.

5. “This free-energy barrier can be eliminated by using the void-induced melting method. Voids (introduced in the supercell in the form of Schottky defects) form pockets of a liquid-like structure.” Why are voids liquid-like? Also, does this method depend on the void size?

The voids are described as liquid-like because of the following: With the introduction of vacancies at those very high temperatures, the magnitude of the atomic vibrations around the vacancies are so distorted from the ideal that the local structure is liquid-like. Additionally, the diffusion by the vacancy mechanism in this temperature range is so rapid that the diffusion is effectively liquid-like. We observed this liquid-like diffusion by introducing a single vacancy in an otherwise perfect crystal at temperatures near the melting point. It should be emphasized that single vacancies are inserted, although this method is referred to as the void method. Thus, no actual voids are inserted, only vacancies, up to a specified concentration.

6. “The Kocevski potential shows a good prediction of the UN Poisson's ratio compared to the experimental value at RT.” Based on the figure, it is hard to claim that the prediction is good. Same sentence, “predicts a slight increase with increasing temperature, which is the expected trend.” needs reference; also, at high T, it is not increasing.

The error is Poisson's ratio predicted by the Kocevski potential is 10%, whereas that predicted by the Tseplyaev potential has an error of 20%. We think that ‘good’ is an appropriate description for this comparison. The figure range is quite small, which is why the 10% error may appear quite large.

The sentences have been changed to: “The Kocevski potential shows a good prediction of the UN Poisson's ratio compared to the experimental value at RT with an error of about 10% (Fig. …) and predicts a more or less constant Poisson's ratio as assumed by Hayes *et al.* [...]. However, the Tseplyaev potential overestimates the RT Poisson's ratio by about 20% and predicts a decrease of the Poisson's ratio with increasing temperature, which is related to the significant softening of the bulk modulus.”

7. “The only experimental measurements of the temperature variation, …, with no experimental data to support it.” There is an inconsistent description here.

The phrase “with no experimental data to support it” has been deleted.

8. “To avoid the possibility of defective supercells converging to metastable energy states by static minimization at 0 K, the 0 K formation energies are averaged over many defect configurations” Why are there many defect configurations during the minimization at 0K? Has the minimization procedure changed that leads to the variation?

There are 8 cubic interstitial sites in the UN unit cell. In theory, these should all be equivalent. However, we noticed slight changes (a few tenths of eV) in the converged energy minimization between the different sites. That’s why we took the average. The reason for this variation is that for each configuration the defective system reaches a slightly different metastable state (local energy minima). The same minimization procedure has been used for all defects. We warn against using the 0 K data for the Tseplyaev potential due to these peculiarities.

9. The defect notation should use a non-italic font.

All capital letters have been changed to non-italic.

10. “Bonded defects at 1 K indicate that these defects relaxed to the defect-free crystal structure.” Is this because they are placed too close to each other?

Yes. The nomenclature has been changed from “bonded/unbonded” to “bound/unbound”, as this is the more typical description with regard to Schottky and Frenkel defects. The definition of the bound/unbound defects is included in the manuscript.

11. “It can also be concluded that in U-rich conditions, VN and UN accommodate most of the off-stoichiometry, whereas, in N-rich conditions, VU and N*i* accommodate most of the off-stoichiometry.” This sentence is confusing and needs to be rephrased.

The sentence has been changed to: “It can also be concluded that, in UN, hypo-stoichiometry is accommodated by VN and UN, whereas hyper-stoichiometry is accommodated by VU and N*i*.”

12. Figure 6, are they bonded or unbonded? Need to specify.

The figure caption has been changed to: “Defect formation enthalpy as a function of temperature for the unbound U Frenkel defect, unbound N Frenkel defect, and unbound Schottky defect in UN as a function of temperature as calculated by the Tseplyaev potential. Error bars correspond to one standard deviation.”

13. Why are the Dulong-Petit values close to room temperature results? Discussion would be helpful. “Dulong-Petit values around room temperature and deviates from it at higher temperatures, which is the theoretically expected trend.” Needs reference.

The following sentences have been added at the end of the paragraph: “In general, the specific heat reaches the Dulong-Petit value at temperatures near and below room temperature because of the inability of MD simulations to model the quantum effects that would otherwise lead to the decrease of specific heat with temperature down to 0 K according to the Debye model [...]. Thus, the classical thermodynamic Dulong-Petit value holds at low temperatures. However, at high temperatures, the specific heat is dominated by phonon dynamics which can be modeled by MD to some level of accuracy.”

A reference has been added.

14. “The Kocevski potential predicts that the 𝛽-U2N3 structure can be mechanically stable at very low temperatures.” What is used to assess the “mechanically stable”?

The nomenclature has been changed from “mechanical stability” to “crystal structure stability”. The crystal structure stability of a solid is assessed by observing the radial distribution functions as calculated by OVITO.

The following sentences have been added to the second paragraph of Sec. 2.3.: “The stability of the solid crystal structure is also judged by observing the radial distribution functions (RDFs) calculated by OVITO. For a specific crystal structure, the RDF is characterized by distinct peaks. A phase transformation to a different crystal structure is characterized by the change of peak positions, whereas for a liquid, the RDF is characterized by a single peak after which the RDF assumes a value of about 1 [...].”

15. “The Tseplyaev potential predicts with reasonable accuracy the UN phonon band structure, although the predicted optical branches coincide near the Γ-point due to the absence of long-range electrostatic interactions in the ADP model.” Why due to the absence of long-range interactions? Both potentials do not have long-range interactions. Discussion would be helpful.

The following phrase has been removed from the manuscript: “The optical branches predicted by the Tseplyaev potential coincide only for some portion of the *k*-path because the potential lacks the long-range electrostatic interaction, which is responsible for splitting the longitudinal and transverse optical phonon branches in ionic materials [...].”

Also, the following phrases in the Discussion section: “The Tseplyaev potential predicts with reasonable accuracy the UN phonon band structure, although the predicted optical branches coincide near the Γ-point due to the absence of long-range electrostatic interactions in the ADP model.” have been modified to: “The Tseplyaev potential predicts the UN phonon band structure with reasonable accuracy, which makes it preferable in the evaluation of thermal conductivities.”

Reviewer #2: The article presents a comprehensive comparison of the two available interatomic potentials of the U-N system. This work has high methodological value for the materials science community. The authors examined many interesting aspects of atomistic modeling of UN. Thus, this study deserves publication in the JNM. However, a few things need to be addressed:

1. Despite the wide range of simulated properties, many potential tests for UN have not been conducted. Here I have listed several important properties that can be calculated relatively easily using classical potentials and compared with available DFT or experimental data:

A) Vacancy migration barrier for VU and VN; references:

- [24]

- [L.G. Fonseca, M. Hedberg, L. Huan et al. JNM 536 (2020)]

- [E. Kotomin, Y. Mastrikov, S. Rashkeev et al. JNM 393 (2009)]

We believe that the fundamental inability of the Kocevski potential to describe U-rich conditions makes a thorough analysis of the defect behaviors between both potentials somewhat moot. Thus, we restricted this study to more structural and thermodynamic analyses. The activation energies for migration of both VU and VN are part of our future work on the diffusional creep in UN, where they have more relevance.

B) pressure-volume dependency at low temperature; possibility of phase transition in R-3m structure at P ~ 35 GPa; references:

- [J.S. Olsen, L. Gerward, U. Benedict, Journal of Applied Crystallography 18 (1985)]

- [P. Modak, A.K. Verma, Physical Review B 84 (2) (2011)]

- [Z.-G. Mei, M. Stan, Journal of Alloys and Compounds 588 (2014)]

We think this is interesting to compare, especially since this transition hasn’t been studied by the Kocevski potential. However, our goal was to compare the properties that are more relevant to the in-service conditions of a nuclear reactor. Thus, the stoichiometry-driven phase change from UN to UN2/𝛼-U2N3 is likely more relevant than this pressure-induced phase change. We have alluded to this phenomenon as a potential area of future study in the discussion.

The following phrases have been added to the 3rd paragraph of the Discussion section: “Another interesting aspect worth exploring in future studies is the high-pressure phase transition that UN undergoes from the cubic Fm-3m structure to the rhombohedral R-3m structure [...]. Experimentally, this transition was observed to occur at 29 GPa [...], and the Tseplyaev potential predicted it to take place at a pressure of 35 GPa [...]. However, the Kocevski potential was never used to study this aspect.”

C) Thermal expansion of UN2; reference: [G.W. Chinthaka Silva, Charles B. Yeamans, Alfred P. Sattelberger et al. Inorg. Chem. 2009, 48, 22, 10635-10642]

We stated in the paper that Silva *et al.* reported lattice constants of UN2 and 𝛼-U2N3 as a function of temperature and that their samples were not of high purity but rather included the UN2/𝛼-U2N3 solid solution, and are not ideal for comparison. Additionally, the y-axis of the figures in Silva et al. includes some typos, which makes it difficult to accurately read the data from the figure. Thus, while experimental data does exist for comparison, we felt that the nature of the experimental results and the behavior of the potentials themselves did not warrant extensive comparison.

2. I believe that the term "vacancy concentration" is more appropriate than "void fraction" when describing the melting model.

The term “void fraction” has been changed to “vacancy concentration” throughout the manuscript.

3. Recently, an experimental paper on precise measurement of thermal expansion was published [J. Liu, C. Gasparrini, J. White et al. JNM 575 (2023)]. It will be useful to add this data to Fig. 1b.

The lattice constant and thermal expansion coefficient calculated by Liu *et al*. have been added to Fig. 1a and 1b, respectively.

The caption of Fig. 1 has been changed to: “(a) UN lattice parameter calculated by both potentials and compared to the experimental data of Hayes *et al*. [...] and Liu *et al*. [...]. (b) UN linear thermal expansion coefficient calculated by both potentials and compared to the experimental data of Hayes *et al*. [...] and Liu *et al*. [...]”

The first paragraph of Sec. 3.1 has been updated to: “... As can be seen in Fig. …, the Kocevski potential is closer in comparison to the experimental results of both Hayes *et al*. and Liu *et al*., whereas the Tseplyaev potential slightly underestimates it. The linear thermal expansion coefficient (LTEC) is shown in Fig. … The Tseplyaev potential predicts a temperature-independent thermal expansion up to about 1000 K and then predicts a fifth-order power-law expansion--a trend that does not agree with experimental observations. On the other hand, while the Kocevski potential underestimates the UN LTEC with a larger error than that of the Tseplyaev potential, it predicts a trend very similar to that given by Hayes *et al*. and Liu *et al*.”

4. The authors explained the change in energies for U*i* between 0 K minimization and 1 K, but not for NU (for both potentials).

Thanks for pointing out this aspect. We did a visual inspection of NU and found that at 1 K it transforms to a VU and an N-N dumbbell along the [111] direction. This defect reaction resulted in a 3-eV reduction in the formation energy from the 0 K calculation to the 1 K calculation in all cases. Because atomic vibrations are nearly nonexistent at 1 K, this movement could be easily observed visually.

This paragraph has been added to Sec. 3.6: “Lastly, it is interesting to note that the formation energy of UN at 1 K is nearly one-half of its value at 0 K for all cases. Based on visual inspection it was noticed that the following reaction takes place:

NU → VU + NN-D[111]

where NN-D[111] refers to the N-N dumbbell along the [111] direction. This reaction likely leads to a reduction in the formation energy. Although Kuksin *et al*. [...] did not consider NU they reported similar reactions for other defect types in UN as well as uranium monocarbide (UC).”