Reviewer #1: The present manuscript explores the use of molecular dynamics (MD) simulation to generate data for the mechanical behavior of UN. The authors employ a recently developed U-N potential for this study in conjunction with successful methods for generating cells and evaluating different creep mechanisms of this fuel material, such as Nabarro-Herring and Coble. The produced data is highly needed for UN fuel, as few controlled tests are available.

The manuscript is in good shape for acceptance. I would only recommend that the authors add a small paragraph to the discussion to compare the obtained results for the mechanical behavior of UN with those of UO2. This would provide perspective on whether the fuel would creep more or less than standard UO2 and highlight different mechanisms. Currently, the discussion only provides a comparison with empirical data of UN itself (which is a good validation), but comparing with UO2 would provide a better understanding of the fuel’s different behaviors.

In Figure 5 and the related discussion, we compared the Coble creep rate of our model to that of UO2 in the temperature range 1700–2000 K. Then, we pointed out that comparing the properties of different fuel systems within a temperature range does not accurately represent how the fuel systems behave in reactor conditions. Due to UN’s higher thermal conductivity relative to UO2, its center-line temperature and temperature gradients are expected to be lower than those of UO2 fuel rods under the same conditions. We then conclude that under the same early-life reactor conditions, the thermal creep of UN is expected to be less pronounced than that of UO2. Note that the effect of irradiation on creep is not considered here.

Here is the main text that addresses this issue:

We compare the simulation and parameter-based models of Coble creep in UN developed in this work to Coble creep in UO2 as calculated by Galvin et al. The prefactor in the simulation model was calculated by utilizing the values in Table [X] within Eq. [X]. The prefactor was found to have a numerical value of A = 3.424 × 10-26. For both fuels, a stress of 15 MPa and a grain size of 10 𝜇m were assumed. The result of such a comparison is shown in Fig. [X]. It can be seen that the two models of Coble creep in UN agree to less than an order of magnitude, especially at lower temperatures. Additionally, irrespective of the creep model, the Coble creep rate of UN is approximately one order of magnitude larger than that of UO2. It is also an order of magnitude larger than the Hayes et al. correlation for dislocation creep in UN. Although a stress of 15 MPa is outside the stress range of the Hayes creep correlation (i.e., 20-34 MPa), it is not very far from the range's lower limit, and we assume it can at least provide an estimate of the dislocation creep rate for stresses slightly smaller or larger than the reported stress applicability range. This comparison shows that Coble creep in UN can be more dominant than (or is at least competitive with) dislocation creep. This agrees with the qualitative analysis by Konovalov et al. It should be mentioned that Galvin et al. used a Coble creep prefactor AC = 42𝜋 for their UO2 Coble creep formula, whereas in this work, we use AC = 46.347. However, the difference between the two values is only a factor of 3, which would have little effect on the results in Fig. [X].

Merely comparing the properties of different fuel systems within a temperature range might not accurately represent how the fuel systems behave in reactor conditions. More appropriately, we should compare fuel behaviors at representative center-line temperatures. Considering the high thermal conductivity of UN, it is expected that the center-line temperatures and temperature gradients of UN fuel rods are much lower than those of UO2 fuel rods under the same conditions. For example, based on their finite-element analysis of the thermal-mechanical behavior of several advanced fuel-cladding combinations, Zeng et al. have estimated the early-life centerline temperature of the UO2-FeCrAl fuel system to be about 1300 K, whereas it is only about 900 K for the UN-FeCrAl fuel system. Thus, under the same early-life reactor conditions, the creep of UN is expected to be less pronounced than that of UO2. The effect of irradiation on this comparison, however, requires a separate study.

Reviewer #2: This manuscript presents a detailed and well-structured molecular dynamics study of diffusional creep in uranium mononitride (UN). The work addresses a topic where experimental data are limited and often inconsistent, and it provides valuable computational insights. The authors convincingly show that Coble creep is the dominant diffusional mechanism in the 1700-2000 K range and introduce a practical method for estimating grain boundary widths in polycrystalline UN. These findings suggest that existing creep models for UN may need to be revised to include a more prominent diffusional component.

However, I have a few minor comments which should be addressed before accepting:

1. While the manuscript explains that the Kocevski potential was excluded due to its limitations in modelling metallic U, it would strengthen the manuscript to provide a clearer rationale for selecting the Tseplyaev potential beyond the fact that it does not have these limitations. Specifically, the authors should briefly discuss the key features of the Tseplyaev potential that make it suitable for modelling diffusional creep in UN. This would better justify its use as the foundation for the simulations.

The following paragraph has been added:

“... Additionally, governed by the migration of vacancies and atoms along GBs, accurate modeling of Coble creep requires reliable predictions of the energetics of vacancies and interstitials. The Tseplyaev potential provides accurate formation and migration energies for both stoichiometric and non-stoichiometric point defects that closely agree with DFT values. On the other hand, the Kocevski potential fails to reproduce accurate formation energies for U-rich and stoichiometric conditions.”

2. The authors correctly discuss that MD requires the use of strain rates many orders of magnitude above experiment; however, I think this argument would be strengthened through the addition of citations of other MD studies with high strain rates to make it clear that this is a common limitation, which still leads to experimentally and physically relevant results.

The following sentence has been added with appropriate references:

“Ultrahigh strain rates have been traditionally used in MD modeling of Coble creep and tensile testing.”

3. I believe the authors mention that the data in section 3.1 was averaged over 3 initialisations. Is it straightforward to include error bars on plots 2(b,c,d)? This would help understand the error in these calculations.

Error bars corresponding to one standard deviation have been added to the mentioned plots.

4. In section 3.1, the Coble creep activation energy derived from MD (1.79 ± 0.33 eV) has a relatively high uncertainty. Could you briefly comment on how this variability might impact predicted creep rates?

Based on our tests, we found that this uncertainty in Q can lead to more than an order of magnitude uncertainty in the calculated creep rate. This further supports our choice to use the parameter-based model instead.

The following sentence has been added to the discussion of the activation energy:

“It should be noted, however, that the large uncertainty in the activation energy can lead to more than an order of magnitude uncertainty in the calculated creep rate.”

5. Some of the content currently presented in Section 3.1—and elsewhere in the results—might be more appropriately placed in the Methods section. For example, details regarding system size, energy minimization tolerance, and the use of OVITO modifiers would fit more naturally within the methodological description. Moving these elements would help streamline the results and keep the focus on the scientific findings.

We intended to reserve the Methods section for procedures that are standard in molecular dynamics (MD) creep simulations. In contrast, a few brief (typically one or two sentences for each subsection of the Results) methodological elements were included in Section 3.1 because they are directly motivated by, and most clearly understood in the context of, the results being discussed.

We felt that adopting a structure where these minor, sparse methodological details were placed in the Methods section would lead to a Methods section that felt poorly motivated and disconnected. In particular, because we conducted several MD simulations under varying conditions (e.g., 0 K vs. finite temperature, zero pressure vs. applied stress), consolidating all of these in a single Methods section would risk overwhelming the reader and make it difficult to track which procedures correspond to which parts of the analysis. Integrating a few minor context-dependent details within the Results section helped maintain clarity and logical flow.

We chose this structure to guide the reader through the progression of the study while highlighting the rationale behind each methodological choice at the point where it becomes relevant. We are encouraged by the reviewer’s recognition of the novelty of the work, as it suggests that our presentation achieved its intended effect. If the reviewer still maintains that it is inappropriate to include select methodological information within the results section, we will, of course, make these modifications.

6. Figure 3: The distinction between the two subfigures—bulk (a) and grain boundary (b)—should be made clearer directly within the figure itself. As it stands, a reader might assume that (b) is derived from (a), rather than representing a separate GB analysis. Although the caption clarifies this, it would be helpful to label the plots more explicitly (e.g., "Bulk Diffusivity (MSD)" and "Grain Boundary Diffusivity (Arrhenius Plot)") to avoid confusion at first glance.

Labels have been added as requested.

7. Table 2: The inclusion of Omega(atom) as a reference value is noted, but its purpose is unclear—particularly since it is approximately three times larger than the vacancy volume Omega used in the Coble creep model. If Omega(atom) is not used directly in the analysis, the authors should clarify why it is included and discuss the physical significance of the discrepancy between the atomic volume and the vacancy volume. This would help readers understand the rationale for selecting Omega(V₁) as the appropriate value and avoid potential confusion.

The following sentences have been added to the discussion of vacancy volume:

“The vacancy volume is nearly one-third of the atomic volume (13.909 Å3), indicating that the commonly used approximation of equating Ω to the atomic volume is not accurate for UN. The discrepancy arises from the local lattice relaxation and volume shrinkage induced by the formation of a vacancy, highlighting the importance of calculating Ω explicitly.”

8. Figure 5: The legend would benefit from more descriptive labels—specifically, replacing “UN 1” and “UN 2” with “Simulation model” and “Parameter-based model.” This change would make the figure more self-explanatory and improve readability.

The proposed change has been applied to the figure.

9. I believe AC is quoted as 46.374 and 46.347 at different points in the manuscript. Is this a typo or an alternative value?

Thanks for catching this typo. AC = 46.347 is the correct value according to our derivation, and it has been corrected.

10. Line 412: I think “approaches zero” is a better phrase than “dies out”. Also, in the caption to Figure 4, “In the limit of a very long time…” should be changed to something more rigorous.

The phrase “dies out” has been changed to “approaches zero”, and “In the limit of a very long time…” has been changed to “In the limit of infinite time…”.

11. Are the authors able to give some physical interpretation to N0 and C in equation 12? I think this would help the reader follow the logic of this saturation point.

We could identify their sum: N0 + C represents the asymptotic (saturation) number of atoms participating in Coble creep once steady-state grain boundary diffusion is fully established. This statement has been added to the text.

12. In section 3.2.2: If I'm interpreting correctly, the apparent discrepancy between simulations (which show a temperature-dependent GB width) and experiments (which report little to no temperature dependence) is, at least in part, due to the limited timescales accessible in molecular dynamics. That is, higher temperatures in MD simulations allow more atoms to diffuse within the short simulation window, making the GB region appear wider, whereas experiments reflect a long-time equilibrium state. If this is indeed the reasoning, it would be helpful to state this more explicitly in the manuscript to clarify the physical basis for the observed difference and the rationale behind using an extrapolated, temperature-independent GB width.

Thanks for pointing out that this part of the paper is not clear. We have added the following paragraph to make it more explicit:

“In other words, although atomistic simulations reveal a temperature-dependent diffusional GB width, this behavior is not attributed to the limited timescales accessible to MD. Rather, our analysis shows that the number of diffusing atoms within GBs saturates with time and increases with temperature, resulting in an Arrhenius dependence of the diffusional GB width. In contrast, experimental GB widths reflect a long-time effective value that is typically temperature-independent. Additionally, the experimentally measured GB width can vary depending on the measurement technique. High-resolution TEM yields the structural GB width, which is precise but not directly relevant to diffusional creep studies. In contrast, GB widths inferred from diffusion data are more relevant but are less reliable due to uncertainties associated with data fitting. To reconcile simulation and experimental perspectives, we extrapolated the temperature-dependent diffusional GB width to the melting point and defined this extrapolated value as the effective GB width to be used in the Coble creep formulation. This value (2.69 nm) reproduces the expected prefactor in the Coble creep equation and provides a consistent physical basis for incorporating MD-based GB width estimates into mesoscale models.”

13. Further to my previous comments. Section 3.2.2 is very dense with some novel findings contained within. However, the language is quite loose and leads to a section that is hard to parse. I recommend tightening up the language so that the novelty does not get lost.

This section introduces a novel definition of diffusional grain-boundary width based on first-principles analysis of atomistic trajectories in polycrystals—an aspect that, to our knowledge, has not been previously addressed in the literature. Given the technical depth and multi-step reasoning involved, a certain level of detail is necessary to convey the methodology, assumptions, and implications with precision.

The section has a logical progression: beginning with the distinction between structural and diffusional GB width, deriving a general expression for polycrystals, validating the underlying assumptions (e.g., temporal saturation and Arrhenius behavior), reconciling discrepancies with experimental observations, and concluding with a method for extracting a temperature-independent effective GB width. Each paragraph builds upon the previous, and the narrative is constructed to guide the reader through the derivation and its physical justification.

While we recognize that this material is inherently dense, we have ensured that the language remains technically precise and that the structure supports a coherent narrative. To further aid the reader, we have included explicit cross-references and transitional sentences—but we have not removed technical content essential to the novelty of the contribution.

14. Although your extrapolations are well explained, it may help to more explicitly discuss how results from 14-20 nm grains translate to typical UN fuel microstructures (5-30 µm). This could be a one-paragraph addition in the Discussion.

We have added the following paragraph to the discussion:

“While the simulations presented in this work utilize nanometer-scale polycrystals (14-20 nm) to achieve sufficient creep strain within accessible MD time scales, the fundamental deformation mechanism, namely, diffusional creep mediated by GB diffusion, is governed by well-established scaling laws. The Coble creep model inherently accounts for grain size effects, combined with the several hundred MPa stresses, through its *σ/d3* dependence, enabling extrapolation of MD-obtained parameters to realistic microstructural scales (on the order of micrometers) and typical creep stresses (tens of MPa).”

15. The manuscript would benefit from a clearer discussion of how irradiation effects—particularly defect generation and microstructural evolution—might influence diffusional creep in UN. While this is understandably outside the scope of the current work, acknowledging the potential impact of irradiation on GB diffusion and creep mechanisms would help contextualize the findings for reactor-relevant conditions.

The following paragraph has been added to the discussion section:

“While the present work addresses thermally activated diffusional creep in pristine UN, irradiation is expected to significantly influence creep behavior under reactor-relevant conditions. In particular, irradiation-enhanced diffusion can markedly increase atomic transport rates and enable creep rates far exceeding those predicted from thermal diffusion alone. Consequently, the results presented here should be interpreted as a lower bound on in-service creep behavior and motivate future studies that explicitly incorporate radiation damage.”

16. It would be helpful for the authors to briefly comment on the generality of their proposed method for estimating effective grain boundary width for other materials.

The following sentence has been added:

“It should be mentioned that the proposed method for estimating the effective GB width is general and transferable, as it relies solely on atomistic trajectories and makes no assumptions about the material's chemistry, crystal structure, or defect energetics.”