**Reviewer #1:** This manuscript presents a systematic study of Ce and Nd diffusion in body-centered cubic (BCC) Cr and V, using a combination of density functional theory (DFT) and self-consistent mean-field (SCMF) theory. By quantifying vacancy-mediated transport mechanisms for oversized lanthanide solutes, the authors provide much-needed atomic-scale diffusion data for candidate nuclear cladding materials, where such data are notably scarce. These results offer important theoretical insights and have potential relevance for multi-scale modeling of fuel-cladding chemical interaction (FCCI) in advanced nuclear reactors. Overall, the study is timely, methodologically sound, and contributes valuable data to the field. However, certain aspects of the manuscript require clarification or improvement before the work can be considered for publication. Specific points are detailed below.

***Reply:*** *We thank the reviewer for the positive comments and the constructive feedback.*

**Comment #1:** The authors employ different temperature ranges for diffusion coefficient calculations (625-1000 K) and for vacancy drag and segregation analysis (extending up to the melting point). However, the rationale for this temperature partitioning is not explicitly discussed. A clearer explanation of the physical basis for temperature selection in both contexts is warranted to ensure consistency and reinforce the credibility of the analysis.

***Reply:*** *We thank the reviewer for this helpful observation. For the diffusion coefficient calculations, all temperatures were considered but in Figure 3, we only plotted the intermediate range of interest from 625 to 1000 K around the operating temperatures of cladding liners to focus on comparing diffusivities at this range.*

*For the analysis of vacancy drag and solute segregation behavior, we extended the temperature range up to 2000 K—approaching the melting points of Cr and V. This was done to investigate whether a transition occurs from vacancy drag at low temperatures to the inverse Kirkendall mechanism at high temperatures, as previously observed for lanthanides in HCP Zr in our previous work. We have clarified this rationale in the revised manuscript in the beginning of sections 3.2 and 3.3.*

**Comment #2:** While the manuscript includes comparisons between computed diffusion coefficients and values from prior experimental studies, these comparisons are primarily presented in tabular format. To enhance clarity and impact, it is recommended that the authors incorporate representative literature data directly into the existing diffusion plots (e.g., as discrete data points or curves). This will allow readers to visually assess the agreement and trends across methods more effectively.

***Reply:*** *We thank the reviewer for bringing this to our attention. Including all of the comparisons in the tabular format may not be very clear to the reader. However, it should be noted that experimental diffusivities are very scarce and only exist for self-diffusion in BCC Cr and V and for other solutes (not lanthanides) such as Zr and U, which we used for comparison in Figure 5 in the discussion section.*

*Since the only direct comparison will be with DFT-calculated Ce and Nd diffusivities in BCC Cr by Yang et al., we modified Figure 3 to include the results from this reference for a clearer and more impactful comparison, as suggested by the reviewer.*

**Comment #3:** The authors mention that direct exchange between solute atoms and vacancies does not occur in the adopted model. Given this constraint, further clarification of how solute migration is realized under the nine-frequency model would benefit the reader. A more detailed discussion of the relevant indirect mechanisms—such as vacancy-assisted nearest-neighbor hopping—would reinforce the physical plausibility of the diffusion pathways and lend additional support to the simulation results.

***Reply:*** *The reviewer’s comments are deeply appreciated and we agree that more clarification will benefit the reader. We added a couple of sentences explaining the diffusion pathways of the solute via association/dissociation jumps.*

**Reviewer #2:** In the paper entitled "First-Principles Investigation of Cerium and Neodymium Diffusion in BCC Chromium and Vanadium via Vacancy-Mediated Transport" the Authors compute vacancy-mediated diffusion properties in these systems, using a well-established methodology: DFT calculations of vacancy-solute binding and migration energies, SCMF method with the KineCluE code, analysis of the resulting diffusion coefficients and flux coupling properties under irradiation. The study is well conducted and the presentation of the paper is satisfactory. I only have minor comments:

***Reply:*** *We thank the reviewer for the positive comments and the constructive feedback.*

**Comment #1:** Below Eq. 6: the Authors should provide some references to back up their statement that their assumption on attempt frequencies "may only alter the transport coefficients by a factor 2-2.5 at most."

***Reply:*** *Reference [Wu, H. et al. High-throughput ab-initio dilute solute diffusion database. Sci. Data 3:160054 doi: 10.1038/sdata.2016.54 (2016)] was added to this sentence. The assumption on attempt frequencies and the expected error of 2-2.5 factor is taken from this reference.*

**Comment #2:** In Eq. 8, I don't understand why it is a (3N-1) exponent instead of the usual (3N-3) where the 3 rigid-body translation degrees of freedom are removed. Moreover, there is not the same number of atoms in the perfect cell and in the cell containing the defect, so the Authors should expand a bit on how they computed this quantity exactly.

***Reply:*** *We thank the reviewer for pointing this out. Indeed, the standard form of vibrational formation entropy excludes the three translational degrees of freedom for a fully relaxed N-atom system, leading to a (3N−3) exponent.*

*In our work, however, we compute the vacancy formation entropy using the Vineyard harmonic approximation, but only for atoms in the 1st and 2nd nearest neighbor shells around the vacancy site to reduce the computational expense. This corresponds to N=14 atoms that are allowed to vibrate, while all other atoms in the supercell are fixed using selective dynamics in VASP. As a result:*

* *The number of vibrational modes is 3N=42 in both the perfect and defective supercells.*
* *No global translational motion of the system is possible, since the surrounding atoms are fixed. Therefore, the translational zero modes do not appear, and there is no need to subtract them (i.e., we do not use 3N−3).*
* *We ensured that the same atoms (in the same positions) were allowed to vibrate in both the perfect and defective configurations, so the vibrational entropy difference is computed consistently using matched mode sets.*

*We have updated Eq. 8 accordingly and added a clarification in the manuscript.*

**Comment #3:** Page 8 "… the stress tensor of relaxed supercells with substitutional atoms under a constant volume constraint was analyzed." Note that this data can actually be used to remove the elastic interaction between periodic images of the solute and defect, which is quite useful to reduce finite size issues in DFT calculation. Reference [10.1103/PhysRevB.88.134102] and subsequent publications by the same authors provide more information on this matter

***Reply:*** *We thank the reviewer for highlighting the utility of stress tensor data in correcting for elastic interactions between periodic images of defects, as described in Ref. [Phys. Rev. B 88, 134102]. We fully agree that this method can substantially reduce finite-size errors in DFT simulations by accounting for elastic image interactions through the elastic dipole tensor.*

*In the present study, we analyzed the stress tensor primarily to provide qualitative insight into the relative lattice distortion induced by different substitutional solutes (Ce vs. Nd) in Cr and V. We did not apply the correction scheme outlined in Ref. [88, 134102], as it requires a non-trivial implementation involving the elastic Green’s function and summation over periodic images, which is beyond the scope of the current work.*

*Nonetheless, we will revise the manuscript to acknowledge this correction method, cite the suggested reference, and clearly state that our stress analysis is not used to perform elastic corrections. We believe our conclusions remain valid for the trends discussed and that the correction method suggested by the reviewer presents an excellent opportunity for future refinement.*

**Comment #4:** Page 10, paragraph 3.2, "...the observed lower diffusivities in BCC Cr are expected for any substitutional solute…" I find this conclusion too affirmative given the fact migration and formation energies are overestimated in Cr due to magnetic order (the activation energy is 1.3 eV higher than in the references cited in Table 1).

***Reply:*** *We acknowledge the reviewer’s concern regarding the potential overestimation of activation energies in BCC Cr due to magnetic ordering. However, we would like to clarify that the discrepancy is not as large as 1.3 eV. Our calculated activation energy for Cr self-diffusion is 4.17 eV, which is approximately 0.5–0.6 eV higher than DFT results from previous studies that neglected spin polarization. Importantly, our value aligns more closely with experimental measurements (4.21 and 4.51 eV) listed in Table 3, indicating that the inclusion of antiferromagnetic ordering in our calculations results in a more realistic prediction of the self-diffusion activation energy.*

*Therefore, while the absolute values of diffusivity may be influenced by the choice of magnetic configuration in the calculations, the relative trends—both among solutes within the BCC Cr matrix and in comparison with other systems—remain meaningful. Even when considering activation energies from non-magnetic DFT studies, the values for BCC Cr remain relatively high compared to those in systems such as HCP Zr and BCC V. Nonetheless, we acknowledge that the original wording may have appeared overly definitive and have revised it to better reflect this nuance.*

**Comment #5:** Page 11, the discrepancy is fully attributed to the higher activation energy of vacancies in BCC Cr yet I wonder: did the previous studies account for all binding and migration energies up to 5th NN? Is it important to make such detailed calculations? A comment on this topic would help readers understand the pros and cons of the methodology applied in the paper.

***Reply:*** *We appreciate the reviewer’s thoughtful question. In our analysis, the primary source of the discrepancy in activation energies is indeed attributed to differences in vacancy formation and migration energies, largely stemming from the inclusion of antiferromagnetic ordering in BCC Cr. However, the scope of the thermodynamic interaction range—specifically the inclusion of vacancy–solute binding and migration energies up to the fifth nearest neighbor (5NN)—is also important but does not affect the activation energies.*

*To directly address the reviewer’s question, we tested the effect of limiting the interaction range to the second nearest neighbor (2nn), consistent with the nine-frequency model used in other studies. This test showed that our calculated activation energies changed by less than 1 meV in both BCC Cr and V, indicating that the extended interaction range does not significantly affect the computed diffusivities or explain the discrepancy with prior work.*

*Nonetheless, including interactions up to 5nn is essential for accurately computing off-diagonal Onsager coefficients and vacancy drag ratios. As shown in our previous study on HCP Zr, convergence of these quantities requires accounting for long-range solute–vacancy interactions. We have added a clarification to the manuscript to highlight this distinction and the rationale behind our extended interaction range.*

**Comment #6:** Page 15, paragraph 4.2, use "equilibrium state" instead of "standard state" (2 occurrences)

***Reply:*** *The word “standard” was changed to “equilibrium” in the two occurrences as suggested by the reviewer.*