**Reviewer #1:** The paper under current consideration, submitted for publication in Journal of Nuclear materials conducts a theoretical investigation of transport coefficients in dilute binary zirconium alloys Zr-(X = La, Ce, Pr, Nd) for the vacancy mechanism. The migration energies and pre-factors for the vacancy jump frequencies as a function of the local solute environment are calculated using DFT. These frequencies are then used as input data for the KineCluE code to compute the transport coefficients, particularly the flux couplings between the vacancy and solute atoms. The calculations are performed in the infinite dilute limit: no interactions between solute atoms are considered. However, the results appear to be accurate, of course within the limitations of the methods used. Besides, the diffusion data obtained are already very informative. Indeed, we have practically no experimental data and very few modeling results on these systems. The presented results are therefore essential for improving our understanding of the kinetic behavior of these lanthanide species in zirconium. The figures are clear and the article is well-written, although both are heavily inspired by Ref. 41 on the diffusion of solute atoms in iron. Consequently, we recommend the publication of this work with minor corrections.

Below are some more specific comments:

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

**Comment #1:** Page 8, Table 3

Table 3 shows very small values of migration energies for La and Ce. The authors should mention that transition state theory might not apply when kBT is of the same order of magnitude as the barrier.

***Reply:*** *We thank the reviewer for pointing out this observation. We agree that these barriers have a very low value that is comparable to kBT. However, we emphasize that the calculation of solute transport coefficients also depends on other migration barriers (other vacancy jumps in the solute-vacancy cluster). Even if we assume that these jumps are nearly instantaneous, this will not be the rate-limiting step in determining the diffusion behavior of these solutes. We added this comment to the text.*

**Comment #2:** Page 10, Equation (17) and below:

Be careful, the so-called D\_{self} does not correspond to the self-diffusion coefficient. Indeed, D\_{self-diffusion}= D\_{self}×f\_0 , with f\_0 being the correlation factor.

***Reply:*** *Equation 17 is removed from the manuscript since this is not the definition of the self-diffusion coefficient. Equation 18 is edited to define the self-diffusion coefficient by adding the correlation factor (f\_0). We calculate f\_0 using kineclue and the figures and tables are edited accordingly.*

**Comment #3:** Page 11, Figure 4:

The authors should also show previous theoretical DFT calculations of self-diffusivity in Zr in Figure 4 and discuss the comparison in the text.

***Reply:*** *Thanks to the reviewer for this recommendation. We added to this figure the theoretically calculated self-diffusivities from two previous studies in the literature. We commented on these in the text as well.*

**Comment #4:** Page 13, line 26:

The authors present the partial diffusion coefficient ratio as if it is a well-known quantity. It was introduced very recently by Messina et al. They should cite the publication when this quantity is introduced and explain better its meaning. This partial diffusion coefficient ratio is not a ratio of diffusivity. It is more a ratio of solute and vacancy fluxes (per solute atom) driven by a gradient in vacancy chemical potential.

***Reply:*** *The two papers by Messina et al. that introduced the partial diffusion coefficient (PDC) ratio are cited in this paragraph as suggested by the reviewer. Also, the description of this quantity was changed to match that given by Messina et al. The PDC ratio describes the diffusion speed (and direction) of solute atoms relative to matrix atoms.*

**Comment #5:** Page 15, line 13:

Averaging transport coefficients as a sum of transport coefficients along each direction of the crystal seems acceptable. They are the results of mean squared displacements, and if we assume that these mean squared displacements are independent of each other (which is incorrect in absolute terms), we may proceed to an arithmetic average. However, the partial diffusion coefficient ratio is a ratio of mean squared displacements, and averaging them in this manner is not correct.

***Reply:*** *This comment is very appreciated. We agree that this averaging may not be correct and as a result, the whole polycrystalline prediction is removed from the manuscript. Figure 7 was removed.*

**Comment #6:** Page 16, line 11, and Figure 8:

Before this work, there had been a single theoretical study of the diffusion of Ce in HCP Zr using the 8-frequency model. We would appreciate a quantitative comparison of this work with the published study. In particular, we would appreciate a plot of the 8-frequency result in Figure 8.

***Reply:*** *The diffusivity of Ce from that theoretical study was added to figure 8 (now this is figure 7).*

**Comment #7:** Page 16, Figure 8: There is not a significant discrepancy between the present calculation and the measured absolute value of the Nd tracer diffusion coefficient. Does the vibrational contribution play an essential role in the theoretical absolute value? If so, it should be emphasized.

***Reply:*** *We agree with the reviewer that there is a significant discrepancy between our calculations and the experimental results of Nd and Ce diffusion coefficients. The experimental Nd diffusion coefficient reported by Helmreich (ref. 27) is an interdiffusion coefficient and the discrepancy is expected because our calculations are for solute diffusion coefficients in the dilute limit (analogous to tracer diffusion coefficients for Ce in ref. 26). This is now emphasized in the manuscript.*

*It should also be emphasized that the discrepancy is in the activation energies and not just in the absolute values. A more accurate treatment for the vibrational contribution may change our result by at most 1 order of magnitude in the absolute values and will not affect the activation energies. Accordingly, we argue that the source of discrepancy cannot be attributed to the vibrational contribution. It may be attributed to different diffusion mechanisms not considered in this work such as grain boundary diffusion and/or interstitial diffusion. This is discussed in the manuscript. Also, impurity diffusion in the experimental samples may play a role in determining the activation energies. However, in the study performed by Paul et al. (ref. 26), it is implied that the samples are of high purity.*

**Comment #8:** Page 16, line 57:

When the authors discuss the discrepancy between their calculation and the experimental result, they should mention the infinite dilute limit assumption in their calculation and provide the experimental solute concentration. Interactions between the solute atoms and vacancies may be the source of the discrepancy. Also, we would appreciate a plot of the correlation factors. How much do they affect the diffusion coefficient? This property is very sensitive to the solute concentration…

***Reply:*** *We agree with the reviewer that solute concentration plays an important role in diffusion coefficient measurements. We also emphasize that the experimental Nd diffusivity in Zr reported by Helmreich (ref. 27) is an interdiffusion coefficient. This makes the comparison of our results with that measurement less meaningful. Therefore, we focus our comparison mainly on the Ce tracer diffusion coefficient in Zr measured by Paul et al. (ref. 26). In that study, the concentration of Ce-141 is not reported (we add this statement to the manuscript). However, tracer diffusion coefficient measurements are usually performed at dilute concentrations and are concentration-independent in principle.*

*Also, It should be noted that interactions between the solute atoms and vacancies are included in our calculations. What is not included is solute-solute and vacancy-vacancy interactions. This is a reasonable assumption in the dilute limit and should not be the source of the discrepancy with the experimental Ce tracer diffusivity.*

*Regarding the correlation factors, they are independent of concentration in this dilute model. They are only functions of temperature. A section was added in the appendix with plots of diffusion coefficients and the corresponding correlation factors.*

*Regarding the correlation factors, they are independent of solute concentration in this dilute model. They are only functions of temperature. We thus believe that adding the correlation factors to the main body of the manuscript does not add any new information. In the SCMF treatment used in KineClue, they are not used directly to calculate the diffusion coefficients. However, the correlation factors are included in the transport coefficients calculated by KineCluE. They can be calculated from the ratio of a transport coefficient to its uncorrelated part. A plot of the correlation factors versus inverse temperature was added in Appendix D.*

**Reviewer #2:** The Authors present a comprehensive study on vacancy-mediated lanthanides diffusion, making use of state-of-the art tools to address this topic. The paper is directly inspired by previous works on this topic (mostly Refs. 29 and 41) but the diffusion of lanthanides in Zr was not addressed previously, making it worthy of investigation. The paper is well-organized and well-written, but it contains some mistakes or inaccuracies which can be easily corrected. Therefore I recommend major modifications to fix these. Below are my comments.

***Reply:*** *We thank the reviewer for the positive comments and the constructive feedback.*  
  
 **Comment #1:** Abstract: l.27 "not yet unknown" instead of "no yet known"

***Reply:*** *This mistake was corrected.* **Comment #2:** Eq. 1: missing "j" under the summation symbol

***Reply:*** *The summation index “j” was added to the equation.* **Comment #3:** Below Eq. 2: the Ducastelle book is referenced for the cluster expansion of the Onsager matrix, but such topic is not treated in this book. It would be more appropriate to cite Refs. [41,45] instead of [46,45].

***Reply:*** *This book was removed from the references and Ref. [41] was cited instead.*

**Comment #4:** It is not very clear what variable C represents starting from Eq. 3. It depends on what the Authors consider to be a defect. I would rather define C as the total fraction of sites occupied by vacancies, to remove the ambiguity on the word defect. In the same paragraph, note that the cluster partition functions are not the only quantities required to evaluate fractions f\_V and f\_{VB}, equilibrium vacancy concentration and solute nominal concentration are required as well.

***Reply:*** *We thank the reviewer for this comment. We follow both suggestions for defining “C” and explaining how cluster fractions are evaluated. These edits have been included in the text.*

**Comment #5:** Fig. 1, 2 and 3: these figures are very much inspired from Ref. 29. It would be good to indicate it somewhere in the figure captions.

***Reply:*** *This is true. We add this comment in the caption of Fig. 1. We also comment about that in the text referring to Figs. 2 and 3 mentioning that we also extend that to visualize the case of oversized solutes exhibiting the two-half vacancies configuration (not in Ref. 29).*

**Comment #6:** The is a problem in Eq. 7. In Eq. 12, the Authors define binding energies in the convention that positive binding energies means attraction. Therefore, in the KRA approximation, it should be a minus sign in front of the fraction containing the binding energies. Moreover, migration energy is the difference between the saddle-point energy and the energy of the initial state. The right-hand side of Eq. 7 defines (once the sign is corrected) the saddle point energy, not the migration energy. Fortunately both errors cancel out in Eq. 8 which is correct.

***Reply:*** *We thank the reviewer for noticing this mistake in Eq. 7. Editing the sign before the fraction will yield the saddle point energy as mentioned by the reviewer. So, the sign before is edited such that equation 7 gives the migration energy (not the saddle point energy). Eq. 8 is left as it is since it is still valid.*

**Comment #7:** Eq. 14: it would be nice to discuss the validity of the approximation where only the phonon frequencies associated with the hoping atom are considered. The same comment holds for Eq. 15 where only the first nearest neighbors shell is taken into account for the computation of the vacancy formation entropy.

***Reply:*** *A brief discussion on that is added where we refer to a previous study in the literature by Wu et al. (ref. [50]) that showed that the error introduced by this approximation is minimal.*

**Comment #8:** Eq. 16: the [B] in this equation is note the same as in Appendix A (e.g. Eq A.3). In Eq. 16, it should be the nominal solute concentration, whereas in Eq. A.3 it is the isolated solute (monomer) concentration.

***Reply:*** *This is right. The solute concentration in Eq. 16 is given another symbol of “C\_B” to denote the nominal solute concentration. This does not alter any results because the solute concentration is assumed to be fixed to a nominal value ([B] = C\_B) in this work. This assumption is added to Appendix A after Eq A.3.*

**Comment #9:** Eq. 17: this is not the definition of the self-diffusion coefficient, which involves the introduction of a tracer atom, see e.g. the book by Allnatt and Lidiard (Ref. 34).

***Reply:*** *Equation 17 is removed from the manuscript since this is not the definition of the self-diffusion coefficient. Equation 18 is edited to define the self-diffusion coefficient by adding the correlation factor (f\_0). We calculate f\_0 using kineclue and the figures and tables are edited accordingly.*

**Comment #10:** Eq. 18: as a consequence of the previous equation being wrong, the self-diffusion coefficient is missing its correlation factor.

***Reply:*** *This issue was addressed in the previous comment.*

**Comment #11:** Eq. 20: it would be useful for the reader to emphasize the fact that G\_V is an intrinsic property of the vacancy-solute pair, i.e. G\_v = L\_{VB}^{VB} / L\_{VV}^{VB}.

***Reply:*** *This is now Eq. 19 because one equation was removed (comment #9). We thank the reviewer for this suggestion. The (VB) superscript is added in this equation to make it clearer for the reader.*

**Comment #12:** Eq. 21: related to a previous comment, is [B] referring to the monomer concentration or the total concentration of B ?

***Reply:*** *This refers to total concentration of solute B (C\_B). The [B] is replaced with C\_B and this should not affect the results since we assume that [B] = C\_B . Now this is equation 20.*

**Comment #13:** Below Eq. 21, l.36: "This quantity is closely related to the vacancy drag ratio". This is not obvious from the expressions. Is there a way to rewrite Eq. 21 to make G\_V appear somewhere? Also most of the paragraph below Eq. 21 (from l.34 to 48) is well-known and should be referenced accordingly.

***Reply:*** *We agree with the reviewer that it is not obvious from the expression that PDC ratio is closely related to the vacancy drag ratio. Our language was overly general here. Therefore, this statement is removed from the manuscript because the PDC ratio cannot be written directly as a function of G\_V. Also, we add the citation ref. 42 to the paragraph under the equation.*

**Comment #14:** Eqs. 22/23: I don't understand this polycrystalline average, even more I think that conceptually it is wrong. For instance the vacancy drag ratio characterizes the tendency for solute to be enriched or depleted at a sink. For the sake of understanding my concern, imagine that L\_{VB}^{axial}=-2 L\_{VB}^{basal}, both coefficients being non-zero. Then Eq. 22 would lead to L\_{VB}^{av}=0 i.e. there is no solute enrichment or depletion at sinks. Yet the reality is quite different: some sinks are enriched in solutes and others are depleted in solutes, depending on the position of the sinks wrt to the crystal orientation. Therefore, averaging over crystalline directions gives a rather poor description of what is happening in the system.

***Reply:*** *We thank the reviewer for this comment and for the enlightening example. We agree that this polycrystalline average is incorrect and therefore, we removed the whole part discussing polycrystalline average from the manuscript.*

**Comment #15:** Eq. 27: honestly I don't know about Ref. 27, but the fact that it is a diffusion couple makes me think that it was probably an interdiffusion coefficient which was measured, and not a solute diffusion coefficient as computed in this work. The Authors should elaborate on that to make sure that the comparison in Fig. 8 is meaningful.

***Reply:*** *The diffusivities reported in Ref. 27 are interdiffusion coefficients. This is different from the solute diffusion coefficient computed in our work. We add this elaboration after commenting on the difference between our results and Ref. 27. We also focus the discussion on the tracer diffusivity reported by Paul et al. This is a more meaningful comparison.*

**Comment #16:** P.17, l.8: "interstitial formation energies are at least 2 eV higher…" The Authors need to provide a reference for this statement.

***Reply:*** *This is according to our DFT calculations and not from a reference in the literature. We emphasize this fact and report the exact values of interstitial and substitutional formation energies for Ce and Nd.*

**Comment #17:** Fig. 9: I am not sure that surface diffusion is relevant here since it is completely different. The 1-2 orders of magnitude difference between the Author's work and Ref. 63, should be discussed with a previous comment made by the Authors about the vibrational contribution to the vacancy formation entropy, which could result in a factor 8 difference in the diffusivities (P.7, l.42).

***Reply:*** *Surface diffusion is relevant here because Nd can diffuse rapidly in the fuel through surface diffusion due to the presence of interconnected pores and bubbles. The relatively high Nd diffusivity in alpha U accelerates the fuel-cladding chemical interaction (FCCI). The relevance of surface diffusion in alpha U is added to the manuscript.*

*It should be noted that the results from ref. 63 are for alpha U and not Zr. So, the 1-2 orders of magnitude difference is not related to the vibrational contribution (from vacancy formation entropy). It is simply because it is a different system. The intent of the comparison being made here is for the assessment of Zr liners in metallic fuel systems to mitigate FCCI.*

**Comment #18:** P.18, l.16-17 and P.19, l.10-12: "This can be accomplished by controlling the grain texture…" This would probably lead to grain boundary diffusion as well (which is often faster than bulk diffusion). Therefore, wouldn't that be counter-productive? Moreover, solute depletion does not necessarily means that solute atoms do not reach the sinks, because equilibrium solute segregation plays a role as well, sometimes leading to the so-called "W" segregation profiles.

***Reply:*** *We thank the reviewer for pointing this out. Our statement on controlling the grain texture can be misinterpreted by the reader implying that smaller grain size is preferred. However, we mean controlling the crystallographic direction of the grains while maintaining or increasing the grain size. This is emphasized in the manuscript now. Also, we agree with the reviewer that solute depletion does not mean that lanthanides will not reach the sinks. We added a sentence emphasizing that solute segregation to grain boundaries may still occur. We also emphasize that controlling the crystallographic orientation of grains will increase the “tendency” of solutes to remain in the bulk.*

**Comment #19:** Appendix A: this whole Appendix is very much inspired from Ref. 41, and it should be stated somewhere.

***Reply:*** *This is true. A sentence is added to the beginning of Appendix A to mention that.*

**Comment #20:** Eq. A.2: Note that given the cluster partition functions used here (notably Z\_V=2), all of the concentration are computed as per primitive cell quantities, and not per substitutional site quantities. This fact should be either stated explicitly or corrected.

***Reply:*** *Thanks to the reviewer for pointing this out. This point is explicitly added after Eq. A.7 to explain that all concentrations are in per-primitive cell quantities. Also, the concentrations in the caption of Fig. 6 are edited and written in atomic % units.*