Response to the anonymous reviewer #1

Firstly, we would like to thank you once again for your insightful comments. You will find a list of individual responses to the comments below.

Comments by anonymous reviewer #1

>The study by Fukuda et al. provides an important microstructural contribution to the developing field of monazite fission track (MFT) thermochronology by investigating the nature of latent ion tracks using swift heavy ion irradiation and transmission electron microscopy (TEM). The primary objective of the paper is to determine whether fission tracks in monazite form amorphous domains, as is typical in minerals such as zircon, or whether they instead comprise defect-rich crystalline regions. Using Xe ion irradiation at energies of 80 and 200 MeV, designed to approximate the energy range of spontaneous fission fragments, the authors show that ion tracks in monazite-(Ce) exhibit reduced-density linear features while retaining an intact crystal lattice. In contrast, zircon irradiated under similar conditions displays clearly amorphous cylindrical tracks. The study also demonstrates that ion tracks in low-damage Quaternary monazite are etchable despite the absence of amorphous domains, supporting the interpretation that etching behaviour in monazite is controlled by the presence of point defects rather than amorphous material. The authors conclude that the non-amorphous nature of monazite fission tracks may explain the very low closure temperature of the MFT system, as recovery of point defects is expected to require lower activation energy than recrystallisation of amorphous zones.

The study addresses a key unresolved question in MFT thermochronology, which is what the structural nature of latent tracks and the implications for annealing kinetics is. The use of irradiation energies comparable to spontaneous fission fragments represents an important strength of the study, as many previous ion track investigations have relied on much higher electronic stopping powers that may not accurately reproduce natural fission track formation conditions. The TEM imaging approach, including the use of underfocus and overfocus imaging conditions and tilted irradiation geometries, provides convincing qualitative evidence for the presence of linear damage zones in monazite that do not exhibit clear amorphisation. The comparison with zircon irradiated under identical conditions provides an effective internal benchmark demonstrating that the analytical approach is capable of resolving amorphous track cores when present. The etching experiment on Toya monazite also reinforces previous observations that etchability is strongly dependent on radiation damage level, and supports the emerging view that etch selectivity in monazite arises from defect-enhanced dissolution rather than removal of amorphous material. Collectively, the study contributes useful experimental evidence supporting the conceptual model that monazite fission tracks consist primarily of linear defect arrays within an otherwise crystalline lattice.

Thank you for reviewing our manuscript and assessing the significance of our findings.

>Despite these strengths, several targeted improvements would substantially strengthen the paper. This includes firstly integrating the TEM observations with independent quantification of radiation damage (most practically through Raman spectroscopy), would allow the authors to directly link microstructural observations to a measurable damage proxy. This would also enable assessment of whether track morphology varies systematically with pre-existing damage, which is central to interpreting annealing kinetics. What is meant by this is pre-existing radiation damage in monazite might influence how new fission (or ion) tracks form, which in turn affects how those tracks anneal over time and temperature. In a more damaged crystal, the lattice is already disordered, so newly formed tracks may differ in structure, such as defect density, continuity, or geometry when compared to those formed in a relatively pristine crystal. If annealing behaviour depends on the physical nature of the track (e.g., how defects are distributed and how easily they can recover), any variation in track morphology caused by differing damage states could potentially lead to differences in annealing kinetics. This is important because many models assume a single, uniform relationship between time, temperature, and track shortening, but if pre-existing damage influences track structure, then annealing behaviour may vary between grains. Therefore, understanding how track morphology changes with pre-existing radiation damage is essential for accurately interpreting and modelling monazite fission track annealing.

We would like to thank the anonymous reviewer for suggesting that we integrate our TEM observations with the quantification of radiation damage. Accurately evaluating and interpreting annealing kinetics requires knowledge of the presence or absence of pre-existing radiation damage during ion irradiation of monazite crystals. However, at this stage, we did not employ Raman spectroscopy and or XRD analyses for the following three reasons:

1) First, the purpose of our study is to observe the structure of ion tracks in monazite. Raman spectroscopy and XRD can estimate structural disorder (i.e., the level of radiation damage), which depends on the fluence of ion irradiation. While this would help to evaluate the change in damage level before and after irradiation, the main focus of this study is to provide the first clues to the mechanism underlying the extremely low closure temperature of the monazite fission track system through TEM observations of the nature

of tracks. We hope that future studies will investigate track formation in monazite in relation to the impact of radiation damage level.

2) Second, we aimed to prevent the recovery of ion tracks in monazite by an electron beam during Raman and XRD analyses. If they recover, the results of our TEM observations showing no detectable perfect amorphous regions will be discredited. Additionally, the samples we employed have already been analyzed by Raman spectroscopy. In Nakajima et al. (2024), every 50 grains per sample were analyzed using Raman spectroscopy to characterize the radiation damage level of the samples. The monazite samples in this study from EY137-21 and Toya-6b are used from the same fractions in the paper. Strictly speaking, Toya-6b is a different sample to Toya-5b in the paper, but they are part of the same series of volcanic pyroclastic deposits.
3) Third, there is a limitation to Raman and XRD analyses. Raman spectroscopy has a spatial resolution limitation due to the beam diameter (generally several microns). For our TEM observations, the monazite crystals were finely ground using an agate pestle and mortar to prepare thin samples. Such thin (<100 nm) samples cannot be used in these analytical techniques.

>The study also does not consider crystallographic orientation effects on track formation. It implicitly assumes that all tracks behave similarly, however track structure may depend on direction within the crystal due to anisotropy in monazite. Without constraining crystallographic orientation, some of the observed variability in track morphology may be misattributed to intrinsic material behaviour. This represents a limitation of the current study and is an important consideration for future experiments.

We thank the reviewer for highlighting the possible influence of crystallographic orientation on ion-track formation. First of all, two definitions of crystallographic orientation effects can be considered. The first is whether ion tracks are formed in an amorphous or crystalline phase depending on the crystallographic orientation. The second is whether the track diameter varies with crystallographic orientation.

The first definition, rather than the second, is of greater interest from the perspective of our study because our focus is on whether ion tracks in monazite are amorphous. To the best of our knowledge, no previous study has reported that tracks formed in the same material change from amorphous to crystalline solely as a function of crystallographic orientation. Therefore, we believe it is unnecessary to consider the first type of crystallographic orientation effect in the present study.

Regarding the second definition, several studies have reported that track diameter can depend on crystallographic orientation. However, it should be noted that the primary parameter governing ion track formation is the electronic stopping power, whereas crystallographic orientation is only a secondary factor that modifies the dependence on electronic stopping power.Moreover, the oval-shaped track cross sections reported for Bi-based layered oxide superconductors can be attributed to the anisotropy of the thermal conductivity inherent in their layered crystal structure (Zhu et al. 1993, Phys. Rev. B, Fig. 7). In contrast, monazite itself exhibits only modest crystallographic anisotropy, and its thermal conductivity shows little directional dependence. Therefore, any orientation effects under the present irradiation conditions are expected to be negligible.

Nevertheless, we agree that a dedicated orientation-resolved study—e.g., using EBSD-guided ion implantation on single crystals or cross-sectional TEM of selected grains, embedding selected grains in epoxy or aligning a chosen crystallographic plane parallel to the ion beam—would be valuable for confirming this expectation and could reveal subtle effects in low-symmetry phases.

>The etching experiment appears to be designed primarily as a proof of concept to demonstrate that ion tracks, used here as a proxy for fission tracks, can be etched in a similar manner in monazite. In this context, the results are appropriate and support the authors' interpretation that etchability does not require amorphization. Etch rate in monazite is likely controlled by radiation damage, where higher defect densities associated with greater damage enhance dissolution and reduce etching time, whereas low-damage crystals with fewer defects require significantly longer etching durations to reveal tracks. However, the experiment remains limited in scope, as it only compares two etching durations and does not attempt to systematically characterise etching kinetics or variability between grains. As such, while the results are consistent with previous work and sufficient to support the qualitative claim being made, they do not provide a detailed understanding of the relationship between radiation damage, defect structure, and etch behaviour. Expanding the etching dataset or integrating independent measures of radiation damage would strengthen the conclusions, but this is not essential for the primary objective of the study.

We thank the reviewer for the constructive comments on the scope of our etching experiments. The primary objective was to demonstrate that ion-irradiated, low-damage monazite grains can be revealed by chemical etching, thereby supporting the notion that amorphization is not a prerequisite for track visibility. For this reason, we limited the study to two etching durations (90 min and 450 min) based on the etching duration reported by Nakajima et al. (2024). In our samples, the 90-min treatment produced only faint contrast, whereas the 450-min etch clearly exposed abundant pits corresponding to the ion tracks.

>Overall, this study represents a useful and timely contribution to the understanding of latent track structure in monazite and provides experimental support for the hypothesis that MFT annealing behaviour is controlled by recovery of defect-rich crystalline domains rather than recrystallisation of amorphous track cores. The findings are consistent with recent experimental observations of annealing kinetics and etching behaviour in monazite and help strengthen the emerging conceptual framework for interpreting ultra-low temperature thermochronological behaviour. Future work integrating TEM observations with Raman spectroscopy, controlled annealing experiments, and systematic variation in radiation damage state would significantly improve the ability to quantitatively link microstructural damage characteristics with annealing kinetics models. As such, the paper provides an important foundation for further experimental work aimed at refining kinetic models for the monazite fission track system and improving confidence in the interpretation of thermal histories derived from MFT data.

Thank you again for highlighting the importance of our study and findings. We also hope that this study will encourage further research into the formation and annealing processes of fission tracks in monazite, with the ultimate goal of establishing the MFT method as a new thermochronometer.