Geochron 2023-13

On etching, selection and measurement of confined fission tracks in apatite

RC2: *comments by Raymond Donelick, August 1, 2023*

AC3: replies by Raymond Jonckheere, August 2, 2023

General Comments

It is an honor and pleasure to review this excellent paper. Well done Dr Jonckheere! You have quantified the mental picture in my head of the many nuances of confined fission track lengths in apatite. I highly ***recommend*** *this paper be accepted for publication and I leave it to the author to choose to make the minor clarifications I request below. I have not read the comments that have been posted for this paper (as of 31 July 2023) and may not do so.*

I consider it a privilege to have my manuscript reviewed by Dr. Donelick. This has nothing to do with his favourable comments. More than one recent submission has been the target of malicious reviews, which have nothing to do with the scientific content (review #1). I receive no reward and get no promotion for peer-reviewed papers. Publishing has, in my experience, become a disagreeable and exhausting process, and there are easier, perhaps fairer, ways to put out data or ideas for the next generations to run with, ignore, or dismiss.

I am shocked at some of the manuscripts submitted for publication, and reviews that seem like there is an all-out war raging.

However, this review restores my faith in the scientific process. **It is worth it** when someone of the stature and unmatched competence of Raymond Donelick takes the time to have a civilized discussion about one's scientific work.

Specific Comments

Introduction: The biases discussed do not include mention of other decisions facing an analyst such as: 1) Is that a naturally etched fission track? Sample TI of Carlson et al. (1999) exhibits many naturally etched fission tracks near natural grain surfaces. 2) Is that feature a fission track at all or some outlier that should be removed from the dataset? Like potentially naturally etched fission tracks, some potentially non-fission track features can etch like fission tracks but be too long (I use 19 microns as a cutoff for low-temperature natural and laboratory samples). Or perhaps an etched feature looks like an etched fission track but it is too short for the current fission track population(s) under study. Human analysts are smart enough to make these decisions and these decisions should be done openly.

I did not know about the Tioga apatite, but I have also observed naturally etched tracks in some samples. The present work is however carried out on induced tracks Durango apatite, and, with at most a handful of exceptions, all the confined tracks are TinT's. Therefore I did not want to further complicate this manuscript. I also intend to put all the tracks and all the measurements on a suitable server. Carolin Aslanian is also re-measuring the KTB with the intention of creating a virtual KTB (isothermal holding?) profile, showing each measured confined track, allowing everyone to select and measure the tracks according to their insight.

Lines 46-50: I can never produce such a plot as discussed here. The notion of measuring sufficiently-etched fission tracks (apples) and contaminating those data by also measuring under-etched fission tracks (oranges) is not appealing to me. My plots would instead have no data (shorter etch times), then very little data, then sufficient data (longer etch times). I do not mix apples and oranges when it comes to measuring confined fission tracks in apatite.

I understand. I once measured confined tracks etched for 10 s (5.5M HNO_3; 21 °C) with the idea to get at information about the track structure that is erased at longer etch times. Step-etch data show however that the etchant progresses micrometres further along an underetched track than can be seen with a microscope. That makes such measurements meaningless, except perhaps in terms of observation biases. I believe that from the beginning the common-sense approach has been to etch the tracks to their ends, or slightly beyond, and to wrest what information we can from collections of tracks etched to their **intrinsic full lengths**.

Equations: It is these equations that can be used to show that non-elliptical, polar coordinate plots (length, angle to c-axis) of horizontal confined fission track lengths from a single population represent a mixture of apples (sufficiently etched tracks defining the ellipse, usually at higher angles to c-axis) and oranges (under-etched tracks falling short of the ellipse, usually at lower angles to c-axis).

I know those plots, and I admit that our group has produced some as well. We propose to define an effective etch time window to weed out underetched and overetched tracks, and are considering other measures as well.

Lines 172-177: Thank you for reproducing my work and that of Dr. Bill Carlson. The agreement between our works is truly independent and it is not accidental, but instead demonstrates that we as analysts can behave like machines with the ability to maintain biases within ourselves and share those biases with other machines.

I hesitated to discuss the lengths as it had been done before. I now believe that it is perhaps the most important outcome of this work, in particular in a climate where each measurement is considered to be the result of one bias or another, and therefore meaningless. The fact that three investigators separated by two decades and two oceans agree on a substantial set of measurements without conferring is indeed **not accidental**.

Lines 184-187: My first submission of Donelick (1991) was to EPSL and a reviewer rejected the paper on the basis that "they [polar-coordinate plots of fission track lengths] are not ellipses", because his/her published mixtures of apples and oranges did not match my apples-only data.

I am curious who, in 1991, had both the data or the confidence to disagree with the elliptical model, but I can make a guess.

Lines 199-209: Excellent discussion of this interesting issue. As we are after the mean and the mean ranges vary accordingly, it seems it should be possible to prove mathematically this is precisely how it should be.

I am grateful for the support. I am also conscious that, not without genuine hesitation, I am trespassing on your own work. But I think it was worth commenting if it helps someone to better understand it or to make use of it.

*Lines 248-261: In Donelick et al. (1999) I proposed a surface energy model for fission track annealing in apatite. In this model, converging track sides (during annealing) parallel to **c**-axis are flat and perpendicular to **c**-axis are rough with pyramidal faces. In this model, at high angles to **c**-axis and high degrees of annealing you get the results of Donelick et al (1999) – systematic accelerated length reductions - with the occasional Green et al. (1986) – segmentation where one or more opposing and 'rogue' pyramids intersect – along with the tips pinching off and appearing to diffuse into the bulk crystal as in Paul and Fitzgerald (1992). Surface energy minimization explains anisotropy and all experimental observations.*

Lines 270-271: This statement is totally consistent with the surface energy minimization model above.

I agree that the surface energies control the track boundaries during annealing, and, I would add, during track formation and etching. I'd need convincing, however, that the walls of tracks perpendicular ***c*** are made up of pyramidal faces. I see how this is suggested by the terminations of apatite crystals, but why do we not observe a pyramidal texture when etching basal faces? I guess that these ideas go back to Nichols and Mullins (1965)? I found them too difficult to follow but I would be interested to discuss them sometime.

I believe we are all agreed on unetchable gaps at the level of our observations. I commented on accelerated length reduction because it is confusing that both terms have existed next to each other for so long. If they do not mean the same, accelerated length reduction must be more than, or different from, gap formation. If it is different and continuous (no gaps) then is it anisotropic length reduction? Or is it something else?

Lines 346-348: I hazard to say that only analysts willing to mix apples and oranges have this problem as your (and my) experiments demonstrate. This is especially true for samples with low confined fission track densities such as highly annealed experiments or the many, many, many low-fission-track-density natural samples studied without using ^{252}Cf/particle accelerator ion implantation.

I wish we hadn't measured half the low-density samples that we did. It not only tempts the analyst to measure underetched tracks but also to count and measure tracks in faces that are far from prismatic. As Carolin Aslanian will show at the next conference, we combine deep ion implantation with etching for 40 s (Ito, 2004); this produces numbers of confined tracks in common grains of the same magnitude as the that of the surface tracks; we then use our width measurements to select those within a predefined etch time window.

Line 424: I see this minor but significant effect for l_{a0} and l_{c0} and a correlation between these and Dpar and Dper, respectively. I presented these data in Amsterdam (2004) but do not have the reference.

It makes me wonder why I haven't plotted the ellipse axes against effective etch time. It is a great idea; it would be the right graph for seeing if there is a change of the anisotropy during etching. Likely not, but worth checking.

The Amsterdam reference is: O'Sullivan P.B., Donelick R.A., Ketcham R.A. (2004) Etching conditions and fitting ellipses: what constitutes a proper apatite fission-track annealing calibration measurement? 10th International Fission Track dating and Thermochronology (Conference), 8-13 August 2004, Amsterdam, DVL-10-O.

Lines 461-463: Track shapes are due to surface energy ratios, the surfaces being different apatite crystal planes in contact with the etchant of some molarity and at some temperature. For a given apatite, we expect surface energy ratios to change – thus changing track shape - with changing acid strength and temperature (and possibly pressure).

Doubtless. I think that at low enough concentrations diffusion rates (stirring) begin to have an effect as well.

Lines 510-511: I have observed huge differences among unidirectional, low angle-of-incidence ^{252}Cf tracks in apatite. The differences are much more than those observed for confined fission tracks from ^{235}U or ^{238}U but keep in mind that confined tracks include the other half of the track that is missing when using ^{252}Cf. It seems likely that ^{252}Cf length correlates with nucleus energy.

I have never worked with a ^{252}Cf source but with accelerator ions; I cannot say that I observed such an effect.

Lines 544-546: Thank you for showing this. Thank you also for making available your imagery for posterity. In the current state-of-the-art, measurements for fission track experiments should be performed using such imagery and such imagery should be made available to anyone freely who requests it. The days of "just trust me" should end.

This was not the aim of this investigation but I believe it is perhaps the most important result. A colleague wrote in relation to confined track length measurements: "*It all depends on how we look at things and not how they are in themselves (Jung, 1941)*". I cannot think of more depressing (woke) principle in science. It is important to prove that we **cannot** measure whatever we want. I believe our work has reconfirmed that there is a truth, and that it is accessible and meaningful. Why else would we bother? And it does not take much to find it; to also end with a quote: "*There is one thing even more vital to science than intelligent methods, and that is the sincere desire to find out the truth, whatever it may be*" (Charles Sanders Peirce).

Clarifications Requested

Lines 193-195: Do you mean the standard deviations of ***c****-axis projected fission track lengths? Please clarify.*

Yes; I will check if the symbol (s_{PM}) is consistent with its use elsewhere in the text and in the Figures, and explain.

Lines 237-239: I assume you mean "...shorter projected mean lengths" "...longer projected mean lengths" "...order of projected mean lengths". Please clarify.

Yes; I will correct it.

Lines 241-242: It might be useful to cite here Jensen and Hansen (2021) and related comments https://gchron.copernicus.org/preprints/gchron-2021-8/#discussion.

I can add the reference although I must admit that I did not understand much of that paper or the discussion.

Lines 427-421: It is worth noting here that I measured the Carlson et al. (1999) data using transmitted light only

Yes. I think this proves that conscientiousness is the mark of the scientist, i.e. attempting to measure an unbiased, representative sample, and resisting the temptation to go for the low hanging fruit (picking the fat tracks that light up in reflected light) or a shortcut to a quick result (measuring nice tracks in non-prism faces).

Thanks for taking the time to review,

Raymond Jonckheere
Freiberg, august 2nd 2023.