



Review of gchron-2020-31

Confined fission track revelation in apatite: how it works and why it matters

Richard A. Ketcham and Murat T. Tamer

We should declare that we have a competing model, and thus a possible prejudice against, or in favour, of the reviewed manuscript. It was our intention not to review it, but we accepted at the insistence of the authors, and because of its interest to those in our group involved in related studies. Our evaluation is critical but we believe not unfair. The public comments will set us right, if need be.

General comments

This manuscript reports calculations and numerical simulations of fission-track etching in apatite based on a variable track etch rate $v_T(x)$, isotropic apatite bulk etch rate v_B , and certain track selection criteria. It concludes with a discussion of the implications for apatite fission-track dating and modelling.

An astounding fact considering the first author's impressive publication record is that the manuscript is almost unreadable. Even with foreknowledge of the concepts involved and of earlier publications, we struggled to understand certain sections. This was not helped by the constant use of ill-defined or undefined notions and meaningless word strings (e.g., semi-track penetration calculation). There appears to be no attempt to be understood, let alone to be exact, and readers less familiar with the subject can make heads nor tails of the manuscript, much less evaluate its significance.

A second striking fact is that the manuscript presents a track etch model that appears to exist in a vacuum, disconnected from all existing knowledge of latent tracks, etching of tracks and minerals, and from all published step-etch data, except for those of Tamer and Ketcham (2020), e.g., for the 5.5 M etch (Carlson et al., 1999; Jonckheere et al., 2007, 2017; Tamer et al., 2019), the comparable 5.0 M etch (Laslett et al., 1984; Green et al., 1986; Barbarand et al., 2003; Moreira et al., 2010), and, given that the model is a first-order approximation, for lower etchant concentrations as well.

One respect in which we found the manuscript lacking is a *post-factum* assessment of what it all signifies. The known statistical properties of surface tracks and confined tracks are a basic ingredient of the model. A second ingredient is the principle that to etch a track-in-track, the etchant penetrates down a host track, crosses to the confined track and etches it from their intersection point outwards. It seems not unlikely that this explains the broad traits of Figures 2-4, and, with scaling based on measured lengths, some experimental results as well, without the need for a specific $v_T(x)$ model. A foreseeable consequence is an excess of "under-etched" tracks which is culled down to $\ll 10\%$ based on for the most part unexplained *ad hoc* selection criteria. This results in etch rates (v_{Tmax}) ranging by a factor of ~ 3 and core lengths (Δx_{Tmax}) by a factor of > 50 between samples. Dazed by the mathematical acrobatics, the reader is left wondering what it all means. Is this a hard numerical result, and which of the several factors above weighed most on the outcome, the fundamental geometrical configuration, the etch rate model, or the selection criteria?

From our standpoint, the proposed model is too approximate and dependent on questionable assumptions to be confident about the numerical results. On the other hand, it highlights the paramount importance of etching protocols and selection criteria for apatite fission-track dating and modelling.

Specific comments

4.1. *Etching structure.* Given the title: "*Confined fission track revelation ...*", and Figure 1, showing a confined track with two endpoints, one could be forgiven for thinking that the calculations refer to etching of confined tracks. It turns out, after a full page of baffling equations suspended in mid-air, that the calculation was not that of the lengths of confined tracks at all but of those of a semi-tracks. The confined track length is then obtained as the sum of complementary semi-tracks. One needs to inform the reader from the start about what is going on, i.e. how one is going about solving a problem. With that information and a full explanation of all the symbols (and simpler notation) one need not bother about (1)-(5) as the conclusions are rather obvious from the outset.

Figure 2 suggests a more interesting observation that is not discussed: the results for the "Constant-core model" (A, C) and the "Linear model" (B, D) are identical. It is possible that this is due to a conservation principle, in the sense that if A and B are far enough separated, then the time to etch from A to B is independent of the etch rate function $v_T(x)$ between A and B, but only depends on the area under $v_T(x)$. A consequence of this observation is that if the two end-member models predict the same evolution of (semi-)track length with etch time (cf. Figure 2), then how are step-etch mean-length data going to distinguish between them, or between them and other models?

4.2. *Semi-track penetration and confined track revelation.* Much as the preceding discussion of the track etch rate and apatite etching, the lead up to Figure 3 (line 150) reads as a perfunctory dismissal of the accomplishments of earlier scientist as too trivial to cite. This entire section could have read: "*Semi-tracks have a $\sin(2\delta)$ dip angle distribution and a homogeneous etchable length distribution (Dakowski, 1978; Laslett et al., 1982)*". Then some modelling happens and something is plotted in Figure 3: *relative semi-track penetration and relative confined track revelation against depth* (below the initial or etched surface?). Even with foreknowledge it is not possible to guess at the significance of Figure 3. What is concluded from Figure 3 does not seem to require modelling at all: "*~10% of tracks reach $\leq 1 \mu\text{m}$ and ^{252}Cf tracks are shorter, ... but efficient*". Yes, see Dakowski (1978): $\frac{3}{4}$ of semi-tracks reach $\leq \frac{1}{2}$ a full track length deep, and most have near-zero depths.

4.3 *Confined track intersection.* Line 158: "*weighted as $\cos \delta$* ": see Dakowski (1978). It is hard to comment; on the one hand the concept is simple (hence likely the shorthand reporting) but, on the other, it is impossible to puzzle out what *exactly* has been done or what *exactly* is shown in Figure 4 (CDF?). Its discussion again refers to issues that seem to require no modelling at all: "*only about half of the tracks remain after surface intersecting track are excluded*". Yes, a slab of thickness L below a unit of surface contains $L N$ tracks of which $\frac{1}{2} L N$ intersect it (Fleischer et al., 1975).

4.4 *Confined track selection.* Fleischer et al. (1969), Paretzke et al. (1973) and a great number of other scientists published calculations of etch-time dependent track profiles for varying $v_T(x)$ and isotropic v_B .

Line 185: the isotropic- v_B case is not "simplified" compared to Aslanian et al. (in press), but contradicts it, as well as fundamental etching theories (e.g., Heimann, 1975). The definition of "bulk etch rate" is different.

Line 198: "*we constructed ... an operator bias function*". Despite the reserve one has to ask: where does it end? It brings to mind Murphy's variable constant, which multiplied with anything gives the desired result. How decisively does Figure 6A differ from a straight ramp, or even a step function? If there is an actual basis for it, please discuss it; if not, all that follows from 6A is a possible artefact.

Figure 5 is to all intents and purposes identical with Figure 4 of Fleischer et al. (1969); the equations are also not dissimilar. It is worth noting that the assumed gradual increase of track length is contradicted by the step-etch data of Jonckheere et al. (2017, in part measured by the present co-author), demonstrating that confined track lengths increase in fits and starts, reflecting a discontinuous latent-track structure, as reported in numerous apatite studies (Paul and Fitzgerald, 1992; Paul, 1993; Li et al., 2010; 2011; 2012; 2014, etc.), as well as nigh the entire literature on latent ion tracks (not counting the amusing contributions from the Canberra group who posit a cylindrical track and then shoot their ions straight through the crystal so that the track ends are

missing). One can of course think of $v_T(x)$ as averaging over a certain section, but this should be stated explicitly.

Figure 6. It is worth commenting on the true significance of the observed "excellent agreement". It would appear that the invasive, *ad hoc* filters (6A and 6D) would transform just about any underlying distributions (light blue in 6B and 6E) into almost negligible residual fractions (dark blue in 6B and 6E) with the general Gaussian appearance of the measurements presented for comparison.

Figure 7 is interesting, but, of course, underlies the combined reservations concerning the steps leading up to it.

A puzzling fact about section 4 is that 4.1 sets up two competing *ad hoc* models for the variation of the etch rate $v_T(x)$ along the tracks (in fact one end-member and an generalized member of a single model, which is assumed to be the same for tracks produced by all fission fragments and energies). Sections 4.2 to 4.4 then present modelling results, which seem not to require the equations in 4.1, and never again mention which model (or parameter fit) was used, much less how the models differ.

4.5 *Fitting step-etching data*. This section presents a complicated account of different modelling issues. It would help to reproduce Figure 1 of Tamer and Ketcham (2020) to present the reader with visual map of the data. It is worrying that there appears to be a need to exclude one data point, that the iterative fitting procedure does not seem to converge towards unique best-fit solutions, and that such solutions appear to not be strictly reproducible. Please discuss what that actually signifies.

5. *Results*. One cannot help thinking that the discussion of Figures 8-11 could be so much better structured if, instead of the artificial opposition of "two models", one of which is an end member of the other, there would have been just one constant-core model with two end members, a linear- $v_T(x)$ model ($\Delta x_{Tmax} = 0$) and a constant- $v_T(x)$ model ($\Delta x_{Tmax} = L_{lat}$). The latter is at least relevant to the ceremonial dismissal of the model unfairly attributed to Laslett et al. (1984) in Tamer et al. (2019).

6. *Discussion*. "It is remarkable that we are able to reproduce ... using these simple models of etching structure". Yes indeed, on condition (1) that the predictions in the first place depend on the assumed model, and in negligible measure on the geometrical framework and assumptions concerning track selection, (2) there is a negligible self-referral between the data used for calibration of the models and those adduced for testing them. Perhaps the careful term "reproduce" reflects concern about the fact that these overlap, which, in our view, affects the significance of the results.

"We take these successes as an indication of the overall correctness of our characterization of confined track etching". This is too facile; it could be a Ptolemaic model, which, admittedly based on false assumptions, nevertheless manages to account for isolated appearances (individual experiments) given suitable eccentricities and epicycles. The authors need to make a serious effort to make transparent the network of mutual influences between the geometrical framework, the actual etch model, the various selection criteria, calibration data, and the data used for verification, so as to reassure the reader that their model is not in effect a self-sustaining mirage hovering in mid-air.

It could be argued in favour of the model that, following annealing and etching together, pre-annealed induced tracks are longer than fossil tracks (Wauschkuhn et al., 2015; Figure 15). Perhaps they etch faster?

Line 358: "A thorough re-evaluation of this biasing based on measurements...". This appears to overlook the fact that the (length) bias function implemented in modelling programs is based on experimental data.

Lines 370-380. "As v_T/v_B rises the proportion of accepted tracks increases ...". Do we interpret this accurately as: "Including shorter tracks decreases the mean length and increases the standard deviation (range)"?

Line 386: "... the mean latent track length of 17 μm indicated by our data and model". More reliable estimates bracket the average latent track length in apatite between ~ 18 and ~ 21 μm (Jonckheere, 2003a; 2003b; Jonckheere et al., 2017). A discussion of established concepts such as *etch-*

ing threshold and range deficit relevant to this issue. On a related point: one could cite Figure 1 of Jonckheere (2003a) in support of the linear model, on the assumption that $v_T(x)$ is proportional to dE/dx .

Line 406: "*tip appearance depends on ... mount preparation, polishing, and cleaning technique, microscope optics, and captured image quality*". This should include etching protocol and apatite composition.

Lines 450-455: "*These differences in latent versus measured lengths ...*". The discussion omits established facts such as the *etching threshold and range deficit* (Fleischer et al., 1975; Iwano and Danhara, 1998).

Lines 450-455: "*By stopping etching as soon as the curve of length versus time in step-etch experiments was reached ...*". No, ... as soon as the data showed a constant rate of increase of the mean track length, taken to be the average bulk etch rate. The same principle used for calculating your bulk etch rate.

Lines 460-462: "*... a rate defined by v_B , which may be obtainable from etch figure measurements parallel and perpendicular to the c axis (Tamer and Ketcham, 2020a)*". Long-established etching theories hold that measurements of surface features are not suitable for estimating etch rates (Jonckheere et al., 2019).

Lines 497-498: "*Figure 17 shows the near-surface portion of the penetration model ...*": Figures 3 and 17 remain obscure; they could be etch-time-dependent depth distributions, but a better explanation is needed.

Lines 524-525: "*If ... geological annealing results in different etching rates than laboratory annealing ...*". It could be argued that, after simultaneous annealing, pre-annealed induced tracks are longer than fossil tracks (Wauschkuhn et al., 2015; Figure 15), perhaps because they do etch faster?

Lines 530-549: The outlook section reads as an appeal for more and better data and community involvement, but is rather short on scientific detail and tends to drift off into a dreamy utopia. It can be deleted. It is at this stage more important to look back and re-evaluate the results, than to look forward.

Line 551: "*a range of detailed step-etching data ...*". This is a rather exaggerated designation of a handful of mean track lengths (Table 1) and some intersection depths. The imbalance between the "*comprehensive*" model and the limited data for calibration and testing is a conspicuous weakness of this work.

Line 554: "*Along-track etching velocity ... varies ... among fission tracks ...*". The intended meaning is probably among different *types of tracks* (spontaneous, induced, annealed), not between individual tracks.

Line 563: "*Most variation ...*". Barring sample preparation accidents, does one need an etch model to conclude that lower mean lengths and higher standard deviations result from including shorter tracks? The v_T/v_B criterion is not progress, because it applies to isotropic detectors, not to apatite.

Lines 575-613: "*Appendix: $v_T(x)$ Etching Model Equations*". This appendix contains a long list of repetitive piecewise integrals of simple functions, that are not used to obtain the results in this manuscript, instead produced by numerical simulations. The appendix thus seems superfluous to the manuscript, and may be either deleted or included as a supplement. We have not checked these equations. It is on reflection possible that the modelling uses the equations but this is nowhere made clear.

Matters of detail relating to the writing, figures and captions are too numerous to be addressed at this stage.

Freiberg, 17 November 2020,

R. Jonckheere
B. Wauschkuhn