TECHNISCHE UNIVERSITÄT BERGAKADEMIE FREIBERG

## Two remarks related to the author-reviewer discussion around gchron-2020-31

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

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The new comments on the above manuscript prompt us to respond to two issues connected to our work.

1. P. Green (21.01.21): "*One issue that still puzzles me is the repeated assertion, in the paper under review and in earlier papers in the chain, that* $V_B$ *is not anisotropic. Surely the etch figures in a prismatic surface show that the etch rate is higher along the* ***c****-axis than perpendicular to it. Can anyone explain this conundrum?*".

Although it is indeed an issue in several reviews, we believe that it is not a major problem. The isotropic bulk etch rate ($V_B = 0.022 \pm 0.004$ μm s$^{-1}$) comes from (anneal)-etch-anneal-etch experiments (Tamer and Ketcham, 2020)[1]. After a first 10 s etch, the remaining unetched damage was annealed, after which the tracks were re-etched (+10 + 5 s). The rate of track length increase was assumed to be twice the bulk etch rate in the direction of the confined track. This is right by existing models of fission track etching. It is however wrong by established theories of crystal growth and dissolution. In contrast to $V_B$, a growth or dissolution (or etch) rate $V_R$ is the rate of displacement **of a crystallographic plane as a whole** in a perpendicular direction. It follows that concave forms, as in this case the ends of confined tracks, become bounded by the slowest-etching planes. In general, these correspond to the low-index planes, in apatite to the basal and prism planes (Figure 1). The basal and prism face both have an etch rate $V_R \approx 0.5$ μm min$^{-1}$ (Aslanian et al., 2020). Some geometry reveals that the rate of length increase of an *etched-annealed* track is then between (2×0.5) and (2×√2×0.5) μm min$^{-1}$ (0.017-0.024 μm s$^{-1}$; for Durango apatite etched in 5.5 M $HNO_3$ at 21 °C), depending on orientation. This is consistent with the minimum rate of length increase of *unannealed* tracks (Aslanian et al., 2020; Figure 7c). The higher values reported there are evidence of damage beyond the endpoint of the first etch step (30 s; 5.5 M $HNO_3$; 21 °C), i.e., damage which was annealed in Tamer and Ketcham (2020). It is also worth noting that, according to crystal growth and dissolution theories, convex forms like track-surface intersections become bounded by fast-etching faces (hence the etch pits). It follows that:

(1) One cannot use the length increase of fission tracks for measuring the bulk etch rate in the track direction.

(2) One cannot use the dimensions of track-surface intersections (*Dpar*; *Dper*) for estimating surface etch rates.

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[1] EAE1-3; SE4-6 exhibit a similar trend between 20 and 30 s etching, which is however more difficult to understand.

(3) We leave aside the question of the significance that can be attached to the "roundness" of the track tips.

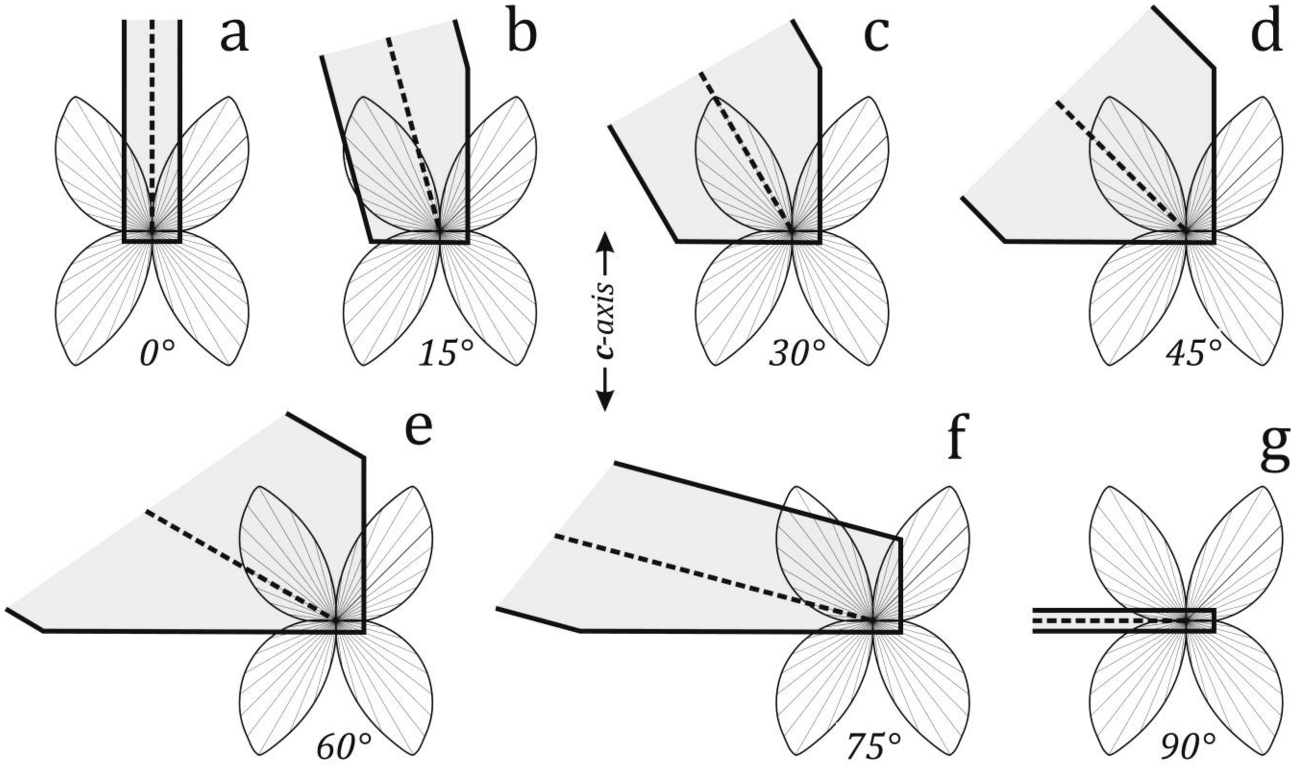

**Figure 1.** Calculated ends of etched confined tracks with various ***c***-axis angles in an apatite prism face. The clover leaf is the envelope of the anisotropic etch rates $V_R$ of the crystallographic planes perpendicular to the etch rate vectors (leaf radii); the apatite ***c***-axis runs from top to bottom (after Jonckheere et al., 2019; Figure 11).

2. P. Green (21.01.21): *"In regard to the evidence in Figure 15 of Wauschkuhn et al. (2015), the data presented there appear to fully support the validity of equivalent time. My reading of that Figure is that the induced tracks that were pre-annealed do not begin to start shortening again until heated at a temperature above that used in the initial treatment. At higher temperatures, both induced and pre-annealed induced populations give similar track lengths, which is just what is predicted from equivalent time.*

In our opinion, this reflects an increasing resistance to annealing but not the effect of equivalent time as such. Duddy et al. (1988, p. 25) write:

> *"[...] the 'principle of equivalent time' [...] assumes that at any moment, a track which has been annealed to a certain degree [...] behaves during further annealing in a manner which is* ***independent of the conditions which caused the prior annealing****, but which depends only on the degree of annealing that has occurred, and the prevailing conditions of temperature and time".*

The authors refer to Goswami et al. (1984,), who, about the independent pathway principle, as applied to track densities (s), write (p. 124):

> *"We [...] assume that once a value s is reached, then* ***all annealing pathways that can lead to the value of s are equivalent****".*

Both these formulations are expressions of the Markov property that a future state depends on the present state and the future conditions but not on how the present state came about. Thus no conditions at all can be attached to the past, because the purpose of equivalent time is that the past may be ignored, *whatever it was*. In the case of the Wauschkuhn et al. (2015) experiment, the pre-annealing of the induced tracks might just as well have been for 10 s at 500 °C.

That said, the experiment does not contradict the Duddy et al. (1988) data or their interpretation (except that the principle is formulated in terms of *"a track"* - i.e., each track - whereas its proof rests on the mean track lengths). Wauschkuhn et al. (2015) also do not call into question its application to geological annealing of fossil tracks in T,t-path modelling. All that the result shows is that a population of induced tracks of a certain mean length undergoes less shortening than a population of fossil tracks of the same mean length. Strictly, this is indeed a violation of equivalent time, but not one that precludes its application to fossil *or* to induced tracks.

Wauschkuhn et al. (2015) admit that their fossil track population is not in all respects identical to that of pre-annealed induced-tracks. However, the equivalent time principle is expressed in terms of the (mean) track lengths, nothing else. In particular, the different origins of fossil and pre-annealed induced tracks (irradiation followed by one isothermal annealing step as opposed to accumulation, a track at a time, during variable geological annealing) are immaterial as long as the track lengths | mean lengths | length distributions (?) are the same. The pertinent empirical fact is that two *almost* identical track populations nevertheless do not anneal at similar rates.

Its significance is that it suggests a difference between fossil and induced tracks (Price et al., 1973; Gleadow et al., 1983; Durrani and Bull., 1987; Tamer and Ketcham, 2020). This could be pertinent to the length-density relationship of fossil tracks, the reliance on curvilinear equations to explain geological track lengths, and, failing sufficient curvature, the world-wide exhumation. At the outside, one could speculate that the apparent increasing resistance to annealing, producing fanning Arrhenius diagrams, is in fact a side-effect of an increasing track etch rate $v_T$. Before dismissing the Wauschkuhn et al. (2015) result we should at least repeat the experiment.

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