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Discussion

Comment on: "Experimental evidence for the pressure dependence of fission track annealing in apatite" by A.S. Wendt et al. [Earth Planet. Sci. Lett. 201 (2002) 593–607]

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Abstract

The pressure experiments reported by Wendt et al. are cast in such a way as to question the fundamental basis underpinning apatite fission track thermochronology, implying the possibility of a large systematic error in previous interpretations. We find however, that the study is severely compromised by fundamental errors in both experimental design and execution, as well as lacking in consideration of a substantial body of previous work in the area of fission track annealing studies. Wendt et al. have not attempted to extrapolate the results of their experiments to geologically relevant heating times and temperatures. This has been the fundamental test that all previous annealing models have had to pass. Their study clearly fails this test. Moreover, if significant pressure dependence does exist then it is implicit in existing, deep borehole-consistent annealing models. Whilst such models are interpolated in terms of temperature, the borehole test means that they also accommodate any other factors, which correlate with temperature, including pressure. The implications of the results reported by Wendt et al. have been overstated. Far from being "intimidating", we find that they have little relevance either to previous studies of apatite fission track annealing and extrapolations based on them, or to the routine application of fission track analysis to elucidate thermal history information under geological conditions.

1. Introduction

More than 25 years have elapsed since experimental studies carried out to evaluate the effects of pressure on the stability of fission tracks in different minerals [2–6] suggested that any effects in geological conditions were insignificant when compared to those due to temperature. Hence, the paper by Wendt et al. [1] investigating the possible pressure effects on fission track annealing in apatite represents a timely reassessment of this issue.

Because the occurrence of track fading is cen-

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tral to the understanding of fission track systematics and its application to fundamental problems in the Earth Sciences, a considerable number of annealing studies have been carried out on different minerals. Two approaches have been employed. One based on laboratory experiments, where fading of freshly induced fission tracks was investigated under varying time and temperature conditions and the results extrapolated to geological time scales on the basis of an understanding of the physical processes governing track fading of the mineral studied. The second involved the observation of spontaneous track fading under well-documented geological conditions, such as in deep boreholes (where tracks may be fading under the present-day thermal regime) or in samples with a well-constrained geological history. For apatite, the most common mineral used in fission track studies, the kinetics of track fading are considered to be the best understood, and broad agreement has been apparent between the two approaches. This observation immediately suggests that any effects due to pressure must be relatively minor in relation to temperature.

The experiments reported in [1] are presented in such a way as to question the fundamental basis underpinning apatite fission track (AFT) thermochronology, which the authors' assert, (p. 605) have both "significant and intimidating" implications for the application of AFT studies to geological problems. As such the study casts doubt on a wealth of previous research related to the results of laboratory annealing experiments and their extrapolation to geological conditions, implying the possibility of a large systematic error in previous interpretations. The work of Wendt et al. therefore promises new insights into the understanding of track formation and annealing in apatite. However, their study is severely compromised by fundamental errors in both experimental design and execution, and a lack of consideration of a considerable body of previous work in the area of fission track annealing studies. For this reason, we suggest the study does little to further our understanding of the influence of pressure on AFT annealing or its interpretation. It does however raise important issues, which invite further, careful experimental investigation to revisit this question.

2. Evidence from deep boreholes

The reported findings by Wendt et al., and their implications that tracks in apatite should be stable to much higher temperatures than suggested by existing kinetic models of fission track annealing, are difficult to reconcile with studies from deep boreholes. These repeatedly show that AFT ages decrease to zero at temperatures between ~100 and 120°C, in agreement with predictions based on laboratory annealing models [7–11] (if the influence of the known high chlorine apatite grains within the Otway Basin [8] is allowed for, then the appropriate range for apatites with compositions similar to those on which commonly used kinetic models are based is ~100–110°C). The borehole data cover a range of thermal gradients and thermal histories, and therefore provide strong constraints on the magnitude of any pressure effect on annealing. It should be noted that in one case [7] total annealing occurs at 135°C, but this is in deep drillholes penetrating the flank of the Valles Caldera, New Mexico, which is a Pleistocene volcanic centre. The high temperature gradient (~60°C/km) in this well is related to a recent thermal heat pulse (~1 Ma) and the higher present-day temperature required for complete annealing is consistent with this thermal history.

Of particular relevance for the Wendt et al. study are AFT data reported from deep boreholes in regions of relatively low thermal gradient, an environment where any pressure effect should be maximised. Studies have been reported from two deep boreholes drilled in the Baltic shield [9,10], i.e. Gravberg-1 and the Superdeep Kola SG3 with average geothermal gradients of ~16–17°C/km and ~18°C/km respectively. AFT data from Gravberg-1 are reduced to zero at ~6.63 km at a present-day temperature of ~113°C [9]. This was later refined to between 100 and 110°C [10], while those from the Kola borehole yield virtually zero ages at ~7.06–7.66 km where present-day temperatures are ~122°C. These depths are approximately double that found in boreholes in much higher gradients. Not only are these temperatures similar to those reported from most wells in higher heat-flow environments, but they provide strong evidence that any pressure effect (in these cases approaching ~200 MPa) can only have a very minor, second-order influence on track annealing. A significant influence of elevated confining pressure on the stability of fission tracks in zircon has also been ruled out by a recently published study involving both field and laboratory work [12].

Given the importance of the borehole data to the interpretation of AFT measurements and their broad agreement with the extrapolation of the fanning model [13] based on laboratory annealing studies to geological time scales, e.g. [14], it is unfortunate that these were not discussed by Wendt et al. This oversight is difficult to understand given that, as reviewed above, there are numerous well-documented examples from deep drillholes that unequivocally show that pressure has very little effect on the thermal annealing of fission tracks at temperatures of ~110°C or less.

3. Experimental design

Wagner and Van den haute [15] comprehensively reviewed the theory and applications of the fission track technique, and summarised a great deal of previous research (see references therein). In doing so they pointed out a number of prerequisites necessary for rigorous annealing experiments, many of which had been outlined in more detail elsewhere [16]. These include: known and homogeneous crystal chemistry, defined crystallographic orientation, and in particular the use of freshly induced tracks rather than partially annealed natural tracks. This last point was acknowledged by Wendt et al. (p. 595) but nevertheless for their experiments they selected apatites from a variety of locations and provenances, presumably having quite different thermal histories, apparently heterogeneous in chemical composition and unlike all major fission track annealing studies, used spontaneous fission tracks. These surprising choices mean that no direct or reasonable comparison can be readily made with the very extensive array of published annealing studies.

The choice by Wendt et al. of natural spontaneous tracks for their experiments has a more profound implication. Consider a sample containing 100 natural track lengths of demonstrable length variability such as illustrated in their fig. 3. An annealing experiment on such a sample is actually attempting to assess the response of 100 *different* kinetic indicators. This is because the response of fission track fading to temperature varies non-linearly with track length as demonstrated by the concept of ''equivalent time'' [17,18]. This concept postulates that ''...a track which has been annealed to a certain degree behaves during further annealing in a manner which is totally independent of the conditions of temperature and time which caused the prior annealing, but in a way determined only by the amount of annealing that has previously taken place ... and the current time and temperature'' [18]. The principle of ''equivalent time'', validated by laboratory experiments involving variable annealing temperatures [18], clearly creates a problem in comparing the annealing properties of spontaneous and induced tracks. So, for an equivalent thermal exposure over a fixed time interval, a long track will initially anneal at a greater rate than a partially annealed track. Thus, the observed slower annealing in [1] of the Madagascar sample (original mean length = 13.24 μm) compared to the Mexican sample (original mean length = 14.21 μm) (p. 600), both of similar chemical composition, can be understood in terms of the presence of shorter tracks in the Madagascar length population with their increased resistance to annealing.

Induced tracks, traditionally used in annealing studies, have a narrow, nominally Gaussian length distribution and are all the same age, i.e. formed at the time of irradiation. Therefore, they have a very similar response to time–temperature conditions and as a result produce the observed narrow length distribution. By contrast, natural spontaneous tracks are unlikely to result in a simple Gaussian distribution about the calculated mean because individual tracks of varying length will respond differently during experimental runs.

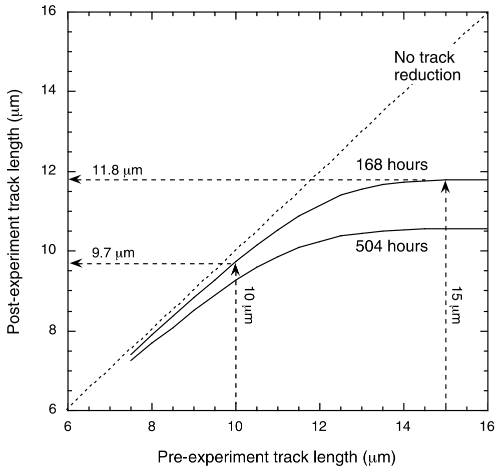

Fig. 1. The evolution of individual pre-experimental track lengths compared with their predicted track length reduction at 250°C for annealing experiments over 168 and 504 h. For an equivalent heating time at the same temperature spontaneous tracks of varying length will fade in a non-linear fashion. For example, spontaneous track lengths of 15 μm and 10 μm annealed for 168 h at 250°C will result in track lengths of 11.8 μm (~21% reduction) and 9.7 μm (~3% reduction) respectively. Calculations are based on the Laslett et al. [13] "parallel" annealing model.

As such, reporting the 'mean' length (as in their table 1 and fig. 2) is a pointless exercise. To do so without indicating the breadth of the track length distribution suggests a far greater precision than can be justified.

The erroneous treatment of track length distributions is further exacerbated by an attempt to compare the experimental results with a calculated theoretical curve purportedly based on the empirically derived model of Laslett et al. [13] (although our calculations do not reproduce the curve shown in their fig. 2). The "parallel form" is chosen [1] as opposed to the preferred "fanning equation" which provided a superior fit to the experimental data and is the form that is universally used. Presumably this choice was made because it allows more simply, the incorporation of their pressure term into the solution (their equation 3). But use of this equation (or the corresponding fanning model) for spontaneous tracks is only possible by first allowing for the appropriate "equivalent time" as discussed earlier. Thus, if a sample, with a bimodal distribution with peaks of 10 μm and 15 μm, is heated for 168 h at 250°C, the resulting distribution will be quite different to that from a sample with a unimodal population, even though the initial spontaneous mean track lengths may have been similar. Our Fig. 1 illustrates this point and shows how individual spontaneous tracks in a natural sample will evolve during a laboratory annealing experiment at 250°C over different periods of time.

In light of the above, both their figs. 2 and 8 must be regarded as questionable, as must any interpretation using correlations based on them.

Other indications of poor experimental procedure are suggested by their fig. 1, which seems to indicate that the samples have been etched to different degrees, a practice never used in fission track dating or annealing studies. That would render any direct comparison impossible either between the different runs or with previous experiments involving constant etching conditions. In addition, it has been shown that quite different track length distributions result for annealed tracks etched in this way [19]. This is a very serious departure from previous experimental practice, the significance of which is that no meaningful comparison can be made with previous annealing studies.

The paper [1] also suffers from shortcomings in basic data presentation. For example, in table 1 the number of measured track lengths is quoted as "> 110". It would have been more informative to present the actual numbers, but more importantly no errors are provided for the mean lengths. In section 5.2 (p. 601) it is stated that "Experiments performed at 500°C were only successful at 2000 MPa...". It is not clear what this means. Does that perhaps imply that other samples at lower pressures did not contain tracks? It is then pointed out that, "...due to the limited number of detected tracks, not all measured tracks were horizontally confined, which in turn biased the absolute mean length values". It is clear from their fig. 5 (showing the tracks produced by this run) that the fission tracks indicated in the photomicrograph are indeed not confined (only one end can be seen). This raises serious questions concerning what type of tracks Wendt et al. actually reported in their other results. In addition, track-density measurements at 0.1 MPa, 504 h shown in fig. 4 are not reported in table 2, and at least one point is incorrectly plotted in fig. 8, i.e. the 0.1 MPa, 504 h experiment.

4. Compositional effects

Wendt et al. suggest (p. 599) that differences in track fading for individual crystals may be due to compositional effects. A lower annealing rate is attributed to the low-chlorine (Canada) sample compared to the high-chlorine (Siberia) sample, despite both having the same mean initial length. Contrary to their interpretation it has been well established in numerous earlier studies that the presence of increased chlorine inhibits annealing (i.e. slows the diffusion process) over the range of F–Cl compositions commonly found in natural apatite samples, e.g. [19]. It is more likely that the observed varying thermal response in these samples is due to a combination of variable initial track length distributions (their fig. 3) and the influence of annealing anisotropy.

5. Anisotropy and crystallography

Previous research has very clearly established that tracks parallel to the crystallographic *c*-axis in apatite anneal at a slower rate than tracks at a high angle to the *c*-axis, e.g. [19–23]. The difference is largely attributed to enhanced diffusion due to the presence of anion channels parallel to the *c*-axis. However, it is argued (p. 600) that, "Tracks oriented parallel to the *c*-axis of the crystal shortened slightly faster, ...". They further suggest (p. 601) that this "confirmed earlier observations" by [23]. Contrary to this assertion, the very first line in the introduction to the article by [23] unequivocally states (p. 1224) the following premise on which the subsequent report is based. "Laboratory annealing experiments have shown consistently that fission tracks oriented at high angles to the crystallographic *c* axis are, on average, shorter than fission tracks at low angles to the *c* axis".

There could be several causes for the misinterpretation of the observations by Wendt et al. They may have measured track lengths on a misoriented crystal fragment or perhaps reversed the axes and/or confused the terms in the calculation of the polar coordinate plot (we note the reversal of the parallel and orthogonal axes between their fig. 3 and figs. 5 and 7). There would appear to be additional evidence for the former possibility. Wendt et al. reported that they cut or drilled parts of large ($\sim$15–30 mm) crystals, which were then treated during the experiments. It is stated that these crystal sections were cut parallel to the *c*-axis. But subsequently it is noted that samples were sometimes broken after experiments. Following this, $\sim$200 μm of material was removed prior to etching the fragments. In contrast to samples characterised by a population of unbroken euhedral grains, e.g. [22], it is often difficult to determine precisely the crystallographic orientation, with confidence, in broken apatite fragments. The presence of parallel etch-pits is frequently used as an indicator that a surface may be parallel to the *c*-axis. This is made more difficult, however, if not impossible, when samples have undergone ^{252}Cf irradiation. This is the case in the work here [1], but the authors have neglected to identify which samples had undergone this treatment. It was suggested in [23] that ^{252}Cf irradiation of their samples probably contributed to a divergence between natural and experimental results. In that work it was also emphasised that parallelism of etch-pits was an inappropriate guide to *c*-axis orientation in the case of ^{252}Cf irradiated samples and it was explicitly stated that "fragments of larger grains" (p. 1232) are a problem area.

6. The process of diffusion

In attempting to emphasise the importance of pressure in the annealing process, a solution to the diffusion equation is presented in equation 1 [1]. There are several problems with the assumptions and application of this equation in the annealing process. Firstly, in explaining the terms, the authors describe (p. 605) the pre-exponential (D_o) as "the diffusion coefficient at ambient temperature". More correctly, the term D is the diffusion coefficient at a given temperature (T). The value of D_o is the diffusion coefficient at an infinite temperature (or in reality, the melting point of a mineral) – this can be seen in that $D = D_o$ only when the exponential term approaches 1, that is, when the temperature (T) is very large. This confusion aside, the authors go on to imply (equation 3) that the diffusion coefficient can be incorporated into an empirically derived equation (equation 2) as the rate constant. However, as long ago as 1981, it was illustrated why attempts to use chemical kinetics and traditional rate equations need to be treated with extreme caution [24]. It was pointed out that chemical kinetic equations are based on "immutable" chemical species whereas tracks are a changing "entity". More recently, a detailed study of numerous annealing experiments led to the conclusion that the simple kinetic approaches implied by such equations were insufficient to describe the fission track annealing process [16].

7. Geological consequences

Drawing meaningful geological conclusions from laboratory scale experiments requires extrapolating a model of the laboratory experimental conditions over many orders of magnitude to those relevant to geological conditions. To do this reliably requires a sufficient range and quantity of laboratory data to control the extrapolation, and preferably also some natural data to test the reliability of the extrapolated model. To emphasise this point we show the experimental data and the "fanning" annealing model of Laslett et al. [13] in Fig. 2 compared with the experiments reported by Wendt et al. We draw attention to two aspects illustrated by Fig. 2. Firstly, the extrapolation of the fanning model [13] is controlled by an appropriate range of experimental conditions and by numerous individual experiments. Furthermore, the extrapolation of the "fanning" model is shown to be consistent with the observations made on the conditions producing complete annealing of fission tracks in apatite from deep boreholes (at pressures ranging from <100 to

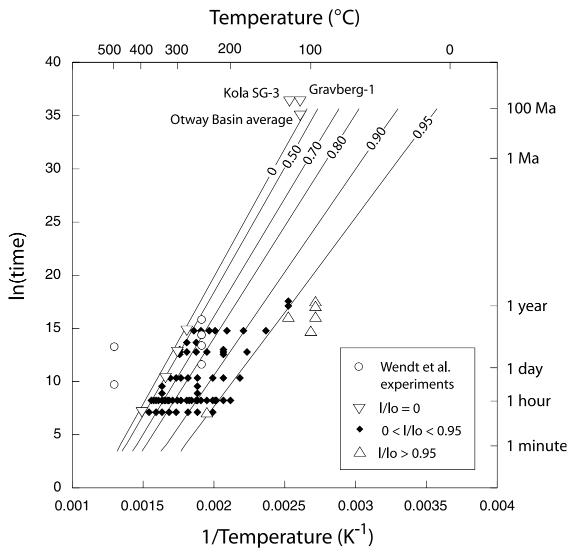

Fig. 2. A comparison of the experiments [19] used to constrain the AFT annealing models described by Laslett et al. [13] with the experiments of Wendt et al. [1] projected on a standard Arrhenius diagram. Note that the natural conditions for complete annealing predicted by the extrapolation of the preferred "fanning" model of [13] are quite consistent with observations from deep boreholes (over a range in pressures from <100 to ~ 200 MPa).

~ 200 MPa). Secondly, it is extraordinarily difficult to accept that a reliable model can be fitted to the data of Wendt et al. that would enable sensible predictions to be made concerning the effect of pressure on the kinetics of annealing of fission tracks in apatite under geologically reasonable conditions. And this is especially true given that Wendt et al. themselves indicate that (p. 605) "Annealing needs to be considered for what it is: a complex interplay of diffusional mechanisms, material properties...and the full range of physical parameters in the (natural) environment" and that therefore there is absolutely no reason why any pressure effect on the annealing kinetics should scale linearly with time or temperature as Wendt et al. imply. So, in our opinion the experiments reported by Wendt et al. do not allow any sensible extrapolation of their observations to geologically relevant conditions, and it is unacceptable to simply "anticipate" what the consequences of such an extrapolation might be without any considered analysis of the reliability of the extrapolation.

Furthermore, in addressing some of the supposed implications of their interpretations, Wendt et al. "...anticipate that the commonly accepted closure temperature for FT annealing...will be higher in most natural conditions..." (p. 605). We draw the authors' attention to the fact that the concept of a closure temperature [25] has had little application in fission track studies for many years. More than 20 years ago, it was recognised that a "partial stability zone" was a more appropriate concept in describing the nature of track shortening [26] and this term was later modified to a "partial annealing zone" [27], while routine application of AFT analysis in sedimentary basins relies on the detailed understanding of the "open-system" behaviour of the system, rather than any concept related to "closure", e.g. [14]. More recently, the concept of closure temperature and its inapplicability to fission track data has been specifically reviewed [28]. Not only does this concept appear to have been misunderstood and its inappropriate use for the AFT system ignored [1], but it has also been erroneously equated (p. 605) with "the temperature at which the rate of annealing tends to zero...", presumably meaning the total track annealing temperature. The misuse of the closure temperature concept for fission track studies is further highlighted in their fig. 9, where it is used to demonstrate the effect of pressure on the fission track system for the calculation of exhumation and erosion rates. We emphasise that the use of a closure temperature is inappropriate because it is the complex track length distribution that is modelled for thermal history studies rather than a unique temperature.

8. Conclusions

The experiments of Wendt et al. are replete with errors and their conclusions based on those experiments are totally inconsistent with the evidence from deep boreholes. Further, they have not attempted to extrapolate the results of their experiments to geologically relevant heating times and temperatures. This has been the fundamental test that all previous annealing models have had to pass. Their study clearly fails this same test. Moreover, if significant pressure dependence does exist then it is implicit in the borehole-consistent annealing models. Whilst such models are interpolated in terms of temperature, the borehole test means that they also accommodate any other factors, which correlate with temperature, including pressure.

It is our view that the implications of the reported results have clearly been overstated and, far from being "intimidating", have little relevance either to previous studies of AFT annealing and extrapolations based on them, or to the routine application of fission track analysis to elucidate thermal history information under geological conditions.

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