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Experimental study on hydrothermal annealing of fission tracks in zircon

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Abstract

Increasing temperature causes fading of fission tracks (FTs) in minerals, which has been quantified by many laboratory heating experiments in atmosphere. In order to elucidate whether or not FT annealing at hydrothermally pressured conditions is the same as that at atmospheric condition, we performed zircon heating experiments using a hydrothermal synthetic apparatus. For reliable comparison, we annealed the same zircon samples using the same temperature monitor and analyzed them using the same experimental procedure as the previous experiment at atmosphere [Yamada et al., Chem. Geol. (Isot. Geosci. Sect.) 122 (1995) 249]. The observed FT annealing characteristics are indistinguishable between atmospheric and hydrothermally heating. It is therefore implied that the annealing kinetics of FT in zircon previously established can be applied to thermal history analysis of rocks that were subjected to hydrothermal conditions in nature, such as those in fault and subduction zones.

Keywords: Zircon; Fission-track; Hydrothermal annealing; Confined track length

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1. Introduction

Increasing temperature causes fading of fission tracks (FTs) in minerals. Thus, we can inversely estimate the thermal history of rocks or minerals by quantitative analysis of FT length and density (Green et al., 1989; Gallagher et al., 1998). Especially, the lengths of horizontal confined tracks (HCTs) offer an accurate measure of track fading with minimum experimental biases (Laslett et al., 1982). Therefore, many artificial heating experiments in the laboratory and observation of geologically annealed tracks have been conducted to establish the accurate relationship between temperature, time, and HCT length reduction for apatite (Green et al., 1989; Laslett et al., 1987; Carlson et al., 1999; Donelick et al., 1999) and zircon (Yamada et al., 1995a; Tagami et al., 1998).

Most laboratory heating experiments have been performed in atmosphere even though those geological samples analyzed in applications may have been subjected to a variety of natural heating conditions. For example, some rocks may have been heated under hydrothermal conditions in natural settings, such as fault zones, sedimentary basins, and geothermal areas. The existence of fluid is thought to increase the rate of phase change and resolution of crystals: for example,

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it is known that a little fluid dramatically accelerates the phase change of Mg_2GeO_4 crystals (Shimizu and Shimobayashi, 1995).

Concerned with this issue, two contrary results have been published recently. Wendt et al. (2002) studied FT annealing in apatite chiefly using horizontal cold seal vessel and found the dependence of FT annealing on pressure. On the other hand, Brix et al. (2002) analyzed FT annealing in zircon using an undefined pressure vessel and found no such dependence. In a first consideration of pressure, Fleischer et al. (1965) employed a "belt" apparatus and NaCl or silicone oil as the high-pressure fluid and found that FT annealing in tektite depends on pressure, while annealing in zircon, olivine, and micas does not. One possible explanation for this contradiction is the difference in materials used. Another possibility is the influence of specific techniques upon the measured annealing data, as previously documented for apatite (Galbraith et al., 1990) and zircon (Yamada et al., 1995b). However, complete experimental conditions of FT heating and analysis were not given in detail in the two recent studies. For example, Wendt et al. (2002) gave no description about in situ heating temperature measurement as well as FT etching criteria. On the other hand, Brix et al. (2002) gave no details of experimental and analytical data of hydrothermally annealed tracks. In addition, they compared the observed FT length reduction in zircon under hydrothermal pressure with that at atmosphere measured previously on different zircon (Yamada et al., 1995a).

In order to elucidate whether or not FT annealing under hydrothermal pressured conditions is the same as that at atmospheric condition, we performed zircon heating experiments using a vertical externally heated cold seal pressure vessel (Tuttle, 1949; Luth and Tuttle, 1963). For reliable comparison, we annealed the same zircon sample using the same temperature monitor and analyzed them using the same experimental procedure as the previous experiment at atmosphere (Yamada et al., 1995a).

2. Sample

Zircon samples were separated from a dacitic welded tuff of the Nisatai Dacite, occurring in the northern part of Kitakami mountain range, northeast Japan. The rocks were collected exactly at the same locality, named NST, as those used by Yamada et al. (1995a,b) and Tagami et al. (1998). The NST was dated as 21.0 ± 0.3 (2 standard errors: S.E.) Ma by K/Ar biotite analysis and contains zircons having a spontaneous track density of $\sim 4 \times 10^6$ cm^{-2} (Tagami et al., 1995). The FT age of another zircon sample collected adjacent to NST is 21.8 ± 1.4 (2 S.E.) Ma. The zircon crystals separated through conventional heavy liquid and magnetic separation procedures are euhedral and clear so that their crystallographic *c*-axes are readily determinable. The variation in uranium content between grains is approximately less than ± 50%. Their spontaneous tracks have not been geologically annealed since their deposition, as confirmed by the concordance between the spontaneous (11.05 ± 0.08 μm with 1 S.E.) and induced (11.03 ± 0.10 μm with 1 S.E.) horizontal confined track lengths (Yamada et al., 1995a).

3. Experimental methods

3.1. Hydrothermal heating

For each heating run, an unannealed aliquot of zircon (~ 1000–1500 grains) was processed for measurement of fractional length reduction. The zircon grains were enclosed into platinum capsule (∅ 2 × ~ 15 mm) with ion exchanged water (~ 10 μl) for protection from pollution and loss. The capsule was put in a vertical cold seal pressure vessel connected (Tuttle, 1949; Luth and Tuttle, 1963) to pump, pressure gauge, and thermocouple and heated by external electric resistance furnace. The temperature and the pressure in the vessel can be monitored directly throughout the run. Time needed to reach plateau temperature from the start of heating is almost constant at about 1.5 h, regardless of final temperature. In contrast, cooling after the run is within ~ 15 min below 200 °C.

In the present study, we used the same voltmeter for monitoring temperature as in the previous experiment at atmosphere (Yamada et al. (1995a,b); they used a chromel–alumel (CA) thermocouple throughout). In the early stage of this study, samples were heated for 11 h after reaching plateau temperature.

Temperature was monitored with a CA thermocouple located at the subsurface of hot end of the vessel, < 3 cm from the sample. Later in the study, the heating duration was kept as 100 h for individual runs, for which we can ignore the annealing effects during the initial 1.5 h and last 15 min. Temperature was monitored with a Pt/12.7% Rhd (PR) thermocouple placed < 1 cm from the sample in the vessel (see Fig. 1). In this case, the outer CA thermocouple was used only for heating control of the furnace. The thermocouples used in the present and previous studies are made by Atlas, and their temperature calibration is certified to within ± 5 °C. The subsurface temperature of the vessel was found to be systematically higher than the in situ temperature for about 30–40 °C (uncertainty about ± 10 °C), slightly depending on plateau tem-

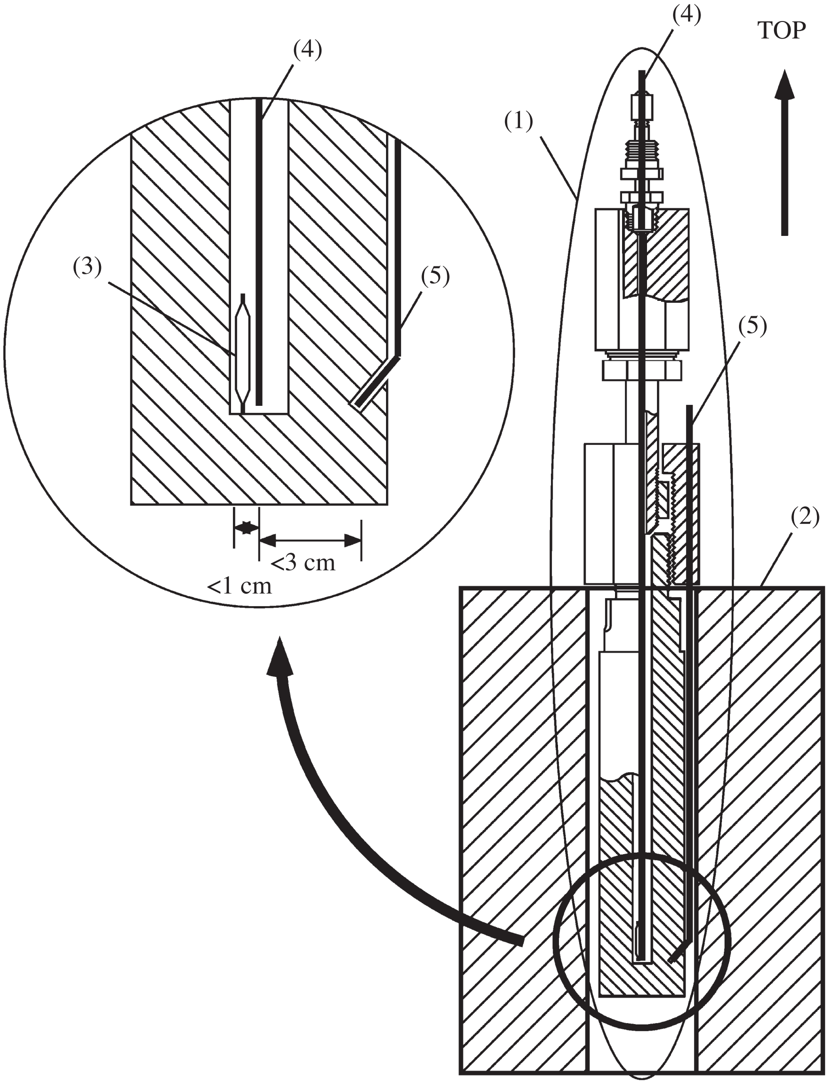

Fig. 1. A hydrothermal synthetic apparatus using vertical externally heated cold seal pressure vessel, with geometries of thermocouples and sample. (1) vessel, connected with pressure gauge, (2) outer electric resistance heater and insulator, (3) sample capsule, (4) Pt/12.7% Rhd thermocouple, (5) chromel–alumel thermocouple.

perature. This temperature gap suggests systematic overestimation of annealing temperature using conventional pressure vessel. Therefore, the early 11-h heating runs are rather erroneous in terms of both heating duration and temperature. We performed six runs at temperatures between 458–548 (for 100-h runs) and 460–610 °C (for 11 h). Temperature was constant within ± 5 °C in the vessel and ± 1 °C at subsurface for 100 h after reaching plateau.

Pressure was kept constant for all runs as 98 MPa, over which the effective pressure is not stable during individual 100-h runs. Zircon grains were heated in supercritical water throughout the study because the $P-T$ condition is over critical point of water (374.15 °C, 16.5 MPa).

3.2. Observation

Experimental and analytical procedures were identical between the original experiments at atmosphere (Yamada et al., 1995a) and the present study, except that a new microscopic system was employed here. Some of the previous samples were reanalyzed using new and old microscopes for cross-calibration. The differences of measured lengths were consistently less than ± 0.2 µm. Unannealed and hydrothermally annealed zircon grains were mounted in PFA Teflon® sheets by handpicking under a binocular so that their prismatic surfaces were ground and polished. Spontaneous FTs used for analysis were etched in NaOH–KOH eutectic etchant (Gleadow et al., 1976; Yamada et al., 1995a,b) at 248 ± 1 °C for 22–26 h until the surface tracks perpendicular to *c*-axes attained the width of 2 µm (Yamada et al., 1995b).

Track lengths were measured for HCTs revealed as track-in-tracks (TINTs; Lal et al., 1969) throughout the study. The lengths of HCTs were measured by taking photographs of tracks using a digital camera Fujifilm HC-300 installed on Nikon FXA microscope with a 100 × dry objective, followed by counting pixel length of the track on an iMac monitor with the help of Canvas® 6 drawing software. Conversion from pixel length to real length was calibrated by measuring a Nikon microscale, 1 µm = 14.85 pixels, that was used by Yamada et al. (1995a) as well. We adopted HCTs for measurement that have orientations >60° to the crystallographic *c*-axis because of enhanced anisotropy in annealing and etching for tracks < 60° (i.e., etched tracks subparallel to *c*-axis appear longer than those sub-perpendicular to *c*-axis) and HCTs for measurement that have widths of 1 ± 0.5 µm in order to minimize the over- or underetching effect on measured HCT length (Yamada et al., 1995b).

4. Results and discussion

Analytical data of the spontaneous FT annealing in zircon are presented in Table 1 and Fig. 2, together with previous annealing data of performed in atmosphere (Yamada et al., 1995a). For 11- and 100-

Table 1
Analytical data of annealing experiments of spontaneous fission tracks in NST zircons

t	T	t_e	N_{all}	N	L_{all}	L	S.D.	S.E.	r
This study—hydrothermal heating									
S_0		22	27	14	11.27	11.26	0.79	0.21	
11 h	460*	22	16	11	10.07	10.07	0.57	0.17	0.89
	500*	22	20	14	9.16	9.46	0.76	0.2	0.84
	610*	22	19	8	7.42	7.43	0.95	0.33	0.66
100 h	458	22	84	52	10.14	10.15	0.71	0.1	0.9
	498	22	48	35	9.28	9.23	0.85	0.14	0.82
	548	22	63	43	7.84	7.67	1.09	0.17	0.68
Yamada et al. (1995a)—atmospheric heating									
S_0		22	107	65	11.14	11.05	0.65	0.08	
11 h	397	22	50	28	10.96	10.89	0.64	0.12	0.99
	499	26	50	30	10.2	10.13	0.6	0.11	0.92
	548	26	50	35	9.42	9.26	0.76	0.13	0.84
	598	26	40	30	7.8	7.74	0.71	0.13	0.7
	649	26	0	0	[~ 6]	–	–	–	–
100 h	350	22	50	35	10.83	10.79	0.7	0.12	0.98
	449	24	51	32	10.23	10.12	0.75	0.13	0.92
	501	26	50	36	9.37	9.2	0.73	0.12	0.83
	549	26	50	27	8.23	8.1	0.68	0.13	0.73
	599	26	6	3	7.24	6.73	1.33	0.77	0.61

t = annealing duration; T = annealing temperature (°C); t_e = etching time (h); N_{all} = number of all measured tracks; N = number of measured tracks >60° to crystallographic *c*-axis; L_{all} = mean length of all measured tracks (µm); L = mean length of tracks >60° (µm); S.D. = standard deviation for the length distribution of tracks >60°; S.E. = standard error of L (µm); r = normalized value of L by L of S_0; In the row of t, S_0 indicates unannealed sample. In the row of L_{all}, the value in brackets is the maximum length of surface tracks, which is roughly estimated using the micrometer attached with an eyepiece of the microscope. In the row of T, values with asterisks are estimated from the subsurface temperature of the vessel and probably overestimated ~ 40 ± 10 °C (see Section 3.1).

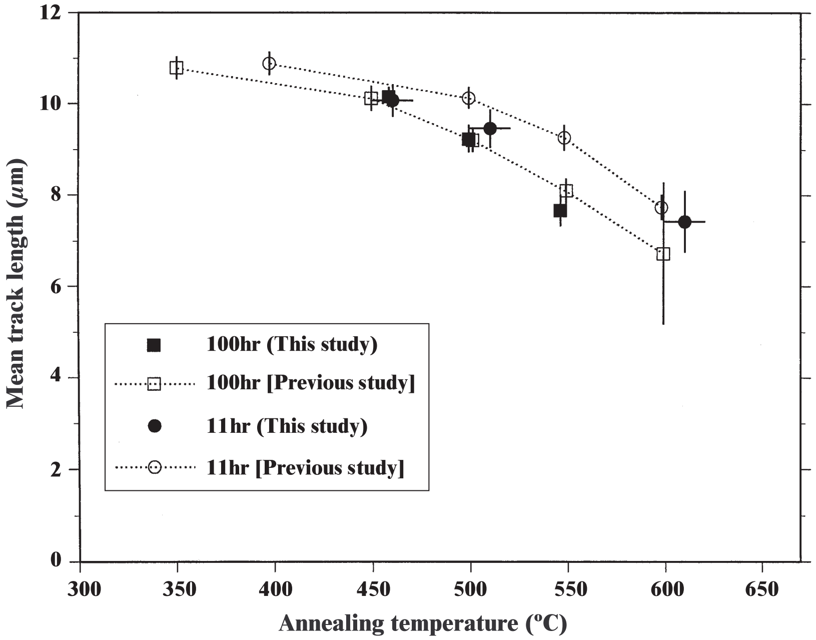

Fig. 2. Mean length of horizontal confined tracks (L in Table 1) against annealing temperature for the present hydrothermal and previous atmospheric heating experiments (Yamada et al., 1995a). Temperatures of 11 h hydrothermal heating are estimated from the subsurface temperature of the vessel (see Section 3.1; Fig. 1) within error ~ 10 °C. Error bars of length represent ± 2 standard errors.

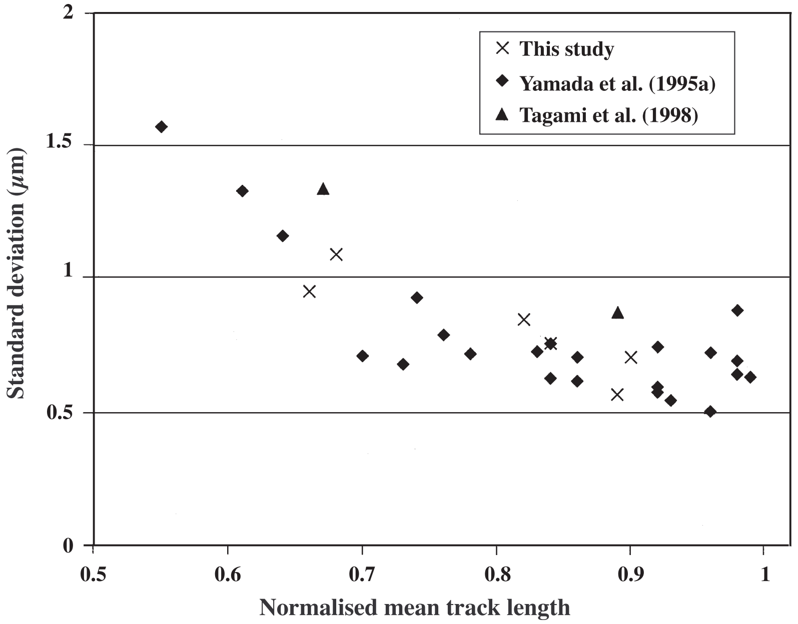

Fig. 3. Standard deviation of HCT length distributions plotted against mean track length normalized to that of unannealed sample (r in Table 1). The present hydrothermal and previous atmospheric heating experiments (Yamada et al., 1995a; Tagami et al., 1998) show a consistent trend of data.

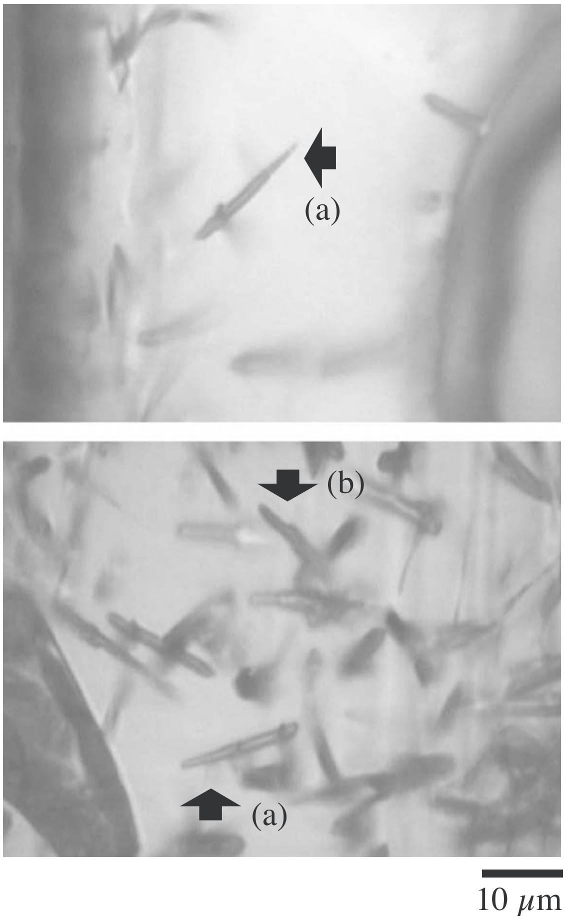

Fig. 4. Photographs of shortened tracks in zircons annealed hydrothermally. (a) are HCTs, and (b) is surface track with a thin part at each end. Tracks, especially both of (a) and (b), look similar to the shortened ones annealed in atmosphere [e.g., figures 8 (c-1, c-2; surface tracks) and 8a (#22, 24 h; HCT) of Yamada et al., 1995b].

h heating, the track length–temperature relationships are indistinguishable within 2 standard errors between the present hydrothermal and previous atmospheric heating. Fig. 3 represents the relationship between the normalized mean track length (r in Table 1) and standard deviation of the track length distribution (S.D. in Table 1), including previous data of heating performed in atmosphere (Yamada et al., 1995a,b; Tagami et al., 1998). The relationship for the present hydrothermal heating is indistinguishable from that for the previous atmospheric heating. In addition, the etched shape of HCTs as well as that of 4η surface tracks appears indistinguishable between the two types of heating experiments (Fig. 4). These lines of evidence suggest that there are no significant differences in track annealing characteristics between the two.

Finally, it is noted that the 98-MPa pressure used in the present experiment is equivalent to ~ 3 km crustal depth. By assuming a geothermal gradient of 30 °C/km, this corresponds to the geoisotherm of ~100 °C. Therefore, the present experiment simulates a natural reheating episode associated with flow or dispersion of anomalously hot fluid, which likely contributes to the modern thermal regime at the toe of accretionary prisms (e.g., geothermal gradients of ~ 100 °C/km observed in ODP Hole 808 near the deformation front of Nankai accretionary prism; Yamano et al., 1992) as well as to ancient thermal regime around a fault (e.g., thermal anomalies deduced around the Nojima fault in two borehole rock sections; Tagami et al., 2001; Murakami et al., 2002).

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