Fission track (FT) thermochronometry using zircon has widely been applied to the resolution of geologic problems, such as to infer the exhumation history of orogenic belts and understand fault-zone processes (e.g., see reviews by Kohn & Gleadow, 2019; Malusà & Fitzgerald, 2019; Tagami, 2005). Thermal annealing characteristics of FTs in zircon have primarily been studied using spontaneous FTs (Murakami, Yamada, & Tagami, 2006; Yamada, Tagami, & Nishimura, 1995; Yamada, Tagami, Nishimura, & Ito, 1995; see also Tagami, Ito, & Nishimura, 1990). This is because (a) in contrast to apatite, the mean length of spontaneous FTs in zircon from rapidly cooled volcanics is indistinguishable from that of induced FTs, suggesting the absence of natural shortening under ambient conditions (Hasebe, Tagami, & Nishimura, 1994), and (b) thermochronologic analyses of geological samples are carried out using spontaneous FTs in natural zircons, for which thermal annealing behaviour should be described.

The thermal annealing kinetics were first determined on spontaneous FTs of the Nisatai Dacite (NST) zircon (Ketcham, 2019; Tagami, Galbraith, Yamada, & Laslett, 1998; Yamada, Murakami, & Tagami, 2007; Yamada et al., 1995). However, it is not well known how the annealing kinetics can vary among natural zircons, for example, as a possible consequence of radiation damage accumulation (Rahn, Brandon, Batt, & Garver, 2004). Kasuya and Naeser (1988) measured confined track length reductions during 1 hr isochronal laboratory annealing for both spontaneous and induced FTs in zircons, and found that (a) induced FTs in pre-annealed zircons are more resistant to thermal annealing than spontaneous FTs, (b) annealing behaviours of spontaneous FTs are indistinguishable between four samples of Palaeogene to Miocene ages showing a range of spontaneous track densities from 0.9 to $10 \times 10^6$ cm$^{-2}$ and (c) induced FTs in non pre-annealed zircons behave like spontaneous tracks. Yamada et al. (1995) also compared confined track length reductions during 1 hr isochronal laboratory annealing between spontaneous and induced FTs in the NST zircon. Experimental procedures were improved by applying analytical criteria and measuring crystallographic orientation of confined FTs to use $L_{60}$ (mean length for tracks with angles
>60° to the crystallographic c-axis) instead of L (mean length for all crystallographic directions) (Yamada et al., 1995). It was found that induced FTs are more resistant to thermal annealing than spontaneous FTs at the advanced stage of annealing, even after correction of track length anisotropy that reflects anisotropic etching and annealing by adopting L (Yamada et al., 1995).

Here, on the basis of those previous results, we perform new laboratory annealing experiments using nine zircon samples of different ages and spontaneous track densities, in order to better reveal the possible variation of annealing behaviours among natural zircons. Confined track lengths were measured on each sample aliquot, after 1 hr isochronal heating at elevated temperatures ranging from 400 to 700°C. Our attention was particularly focused on young natural zircons, in which accumulated radiation damage is minor. This may give a clue to better understand the annealing behaviour of spontaneous and induced FTs and to clarify any difference between them.

### 2 | EXPERIMENTAL PROCEDURE AND SAMPLES

Zircon grains were concentrated and separated from each host rock using conventional mineral separation procedures (Kohn, Chung, & Gleadow, 2019). Nine zircon samples separated from quickly cooled (and not later reheated) volcanics were adopted in this study, including the age standards Fish Canyon, Bishop and Buluk Member Tuffs (Table 1). The zircon samples range in age from 0.6 to 70 Ma and in spontaneous track density from ~0.05 to 7 × 10⁶ cm⁻². For five zircon samples <3 Ma, grains were irradiated by thermal neutrons before thermal annealing at the Thermal Column Pneumatic Tube facility of the Kyoto University Research Reactor. This allowed to form induced FTs from 235U next to spontaneous 238U FTs, so that a sufficient number of confined tracks could be measured in euhedral grains even on these individual sample aliquots. In those five samples, the FTs under consideration for our annealing experiments are thus a mixture of spontaneous and induced FTs.

### TABLE 1  Zircon samples used for annealing experiments

| Sample | Unit name       | T      | ρₛ   | ρₛ + ρᵢ | Hf    | Reference                                      |
|--------|-----------------|--------|------|---------|-------|------------------------------------------------|
| KT06   | Koto Rhyolite   | 69.6 ± 4.6 | 7.16 ± 0.24 | —      | 7.7 ± 1.4* | Tagami et al. (1990) |
| FCT94  | Fish Canyon Tuff| 27.8 ± 1.4 | 5.83 ± 0.53 | —      | 7.7 ± 2.2* | Hasebe et al. (1994), Hurford (1990) |
| NST    | Nisatai Dacite  | 21.0 ± 0.3 | 2.94 ± 0.12 | —      | 8.0 ± 1.4* | Tagami, Uto, Matsuda, Hasebe, and Matsumoto (1995) |
| BM4    | Buluk Member Tuff| 16.3 ± 0.4 | 1.02 ± 0.03 | —      | 5.5 ± 0.4* | Hasebe et al. (1994), McDougall and Watkins (1985) |
| TRG07  | Utaosa Rhyolite | 2.52 ± 0.04 | 0.38 ± 0.06 | 5.54 ± 0.44* | 8.4 ± 1.5* | Uto, Ishizuka, Matsumoto, Kamioka, and Togashi (1997), Uto, Tagami, and Uchiumi (1994) |
| K1018  | Yamakogawa Rhyolite | 1.21 ± 0.04 | ~0.1** | 8.88 ± 0.78* | 8.4 ± 1.1* | Yamada, Tagami, and Kamata (2006) |
| Nz00-22| Potaka Tephra   | 0.97 ± 0.08 | 0.08 ± 0.03 | 5.56 ± 0.45* | 7.4 ± 3.2* | Seward and Kohn (1997) |
| 77G94  | Bishop Tuff     | 0.74 ± 0.07 | 1.22 ± 0.12 | 1.73 ± 0.11* | 8.1 ± 0.5* | Hurford and Green (1983), Izett and Naeser (1976) |
| 75G104 | Verdos ash      | 0.6 ± 0.3  | ~0.05** | 5.85 ± 0.50* | 7.0 ± 1.2* | Naeser, Izett, and Wilcox (1973) |

Note. All errors are quoted at 2σ. All values after references, except for data measured in this study (*) and tentative estimates in this study because the density is too low to be measured accurately (**). Abbreviations: ρₛ = areal density of spontaneous tracks (10⁶ cm⁻²); ρₛ + ρᵢ = areal density of both spontaneous and induced tracks (10⁶ cm⁻²); Hf = Hf content (mg/g); T = age (Ma).
The individual sample aliquots were then mounted into pieces of PFA Teflon sheets, and ground/polished using conventional procedures (Tagami, 2005). FTs were etched by conventional NaOH:KOH eutectic etchant (Gleadow, Hurford, & Quaife, 1976) at $248 \pm 1^\circ C$, until the mean of FT etch pit lengths measured parallel to the crystallographic $c$-axis becomes $2.0 \pm 0.5 \mu m$. Observation and

| Sample    | $T$  | $n$ | $N_{60}$ | $N_{all}$ | $L_{60}$ | $L_{all}$ | $SD_{60}$ | $SE_{60}$ | $t_e$ | $r_{60}$ |
|-----------|------|-----|----------|-----------|----------|-----------|-----------|-----------|------|---------|
| KT06      | —    | 40  | 53       | 30        | 10.94    | 10.79     | 0.78      | 0.14      | 16   | 1       |
|           | 598  | 60  | 51       | 31        | 9.44     | 9.25      | 0.85      | 0.15      | 23   | 0.86    |
|           | 649  | 88  | 53       | 30        | 8.39     | 8.14      | 0.78      | 0.14      | 24   | 0.75    |
|           | 698  | 287 | 32       | 18        | 6.71     | 6.65      | 1.26      | 0.30      | 24   | 0.62    |
| FCT94     | —    | 61  | 57       | 33        | 11.10    | 11.06     | 0.83      | 0.15      | 18   | 1       |
|           | 402  | 16  | 14       | 7         | 10.83    | 10.67     | 0.89      | 0.33      | 18   | 0.96    |
|           | 502  | 33  | 20       | 13        | 10.60    | 10.61     | 0.81      | 0.23      | 18   | 0.96    |
|           | 601  | 86  | 46       | 27        | 9.61     | 9.60      | 0.79      | 0.15      | 25   | 0.87    |
|           | 652  | 77  | 38       | 26        | 8.44     | 8.37      | 0.59      | 0.12      | 25   | 0.76    |
|           | 701  | 108 | 11       | 6         | 6.38     | 5.94      | 0.89      | 0.36      | 25   | 0.54    |
| NST       | —    | 103 | 64       | 35        | 11.31    | 11.21     | 0.68      | 0.12      | 22   | 1       |
|           | 395  | 137 | 50       | 33        | 10.86    | 10.87     | 0.69      | 0.12      | 22   | 0.97    |
|           | 500  | 150 | 50       | 29        | 10.55    | 10.54     | 0.63      | 0.12      | 22   | 0.94    |
|           | 599  | 200 | 50       | 28        | 9.50     | 9.38      | 0.56      | 0.11      | 26   | 0.84    |
|           | 650  | 250 | 50       | 34        | 8.25     | 8.11      | 0.78      | 0.13      | 26   | 0.72    |
|           | 696  | 176 | 14       | 9         | 6.24     | 6.38      | 1.86      | 0.62      | 26   | 0.57    |
| BM4       | —    | 245 | 51       | 24        | 10.53    | 10.47     | 0.92      | 0.19      | 23   | 1       |
|           | 398  | 100 | 21       | 11        | 10.41    | 10.30     | 0.71      | 0.22      | 23   | 0.98    |
|           | 499  | 130 | 26       | 14        | 10.15    | 10.23     | 0.63      | 0.19      | 23   | 0.98    |
|           | 598  | 298 | 42       | 17        | 9.07     | 8.94      | 0.63      | 0.15      | 28   | 0.85    |
|           | 649  | 291 | 35       | 15        | 8.24     | 7.95      | 0.65      | 0.17      | 28   | 0.76    |
|           | 698  | 462 | 22       | 10        | 6.29     | 6.14      | 1.16      | 0.37      | 28   | 0.59    |
|           | 749  | 99  | 1        | 0         | 4.19     | —         | —         | —         | —    | —       |
| TRG07$^a$ | —    | 92  | 52       | 30        | 10.64    | 10.55     | 0.60      | 0.11      | 23   | 1       |
|           | 598  | 77  | 52       | 36        | 9.12     | 9.01      | 0.67      | 0.11      | 28   | 0.85    |
|           | 649  | 110 | 51       | 26        | 8.19     | 8.12      | 0.63      | 0.12      | 28   | 0.77    |
|           | 698  | 120 | 8        | 3         | 5.63     | 5.71      | 0.79      | 0.45      | 28   | 0.54    |
| K1018$^a$ | —    | 83  | 50       | 37        | 10.82    | 10.71     | 0.59      | 0.10      | 28   | 1       |
|           | 599  | 78  | 50       | 38        | 9.37     | 9.34      | 0.72      | 0.12      | 28   | 0.87    |
|           | 651  | 87  | 57       | 40        | 8.57     | 8.41      | 0.71      | 0.11      | 28   | 0.78    |
| Nz00-22$^{a}$ | — | 98  | 52       | 41        | 10.94    | 10.98     | 0.68      | 0.11      | 28   | 1       |
|           | 602  | 63  | 55       | 38        | 9.24     | 9.15      | 0.61      | 0.10      | 28   | 0.83    |
|           | 649  | 75  | 50       | 33        | 8.34     | 8.30      | 0.73      | 0.13      | 28   | 0.76    |
| 77G94$^a$ | —    | 110 | 51       | 32        | 10.94    | 10.95     | 0.58      | 0.10      | 28   | 1       |
|           | 600  | 84  | 48       | 33        | 9.17     | 9.21      | 0.56      | 0.10      | 28   | 0.84    |
|           | 650  | 205 | 48       | 31        | 8.10     | 8.18      | 0.74      | 0.13      | 28   | 0.75    |
| 75G104$^a$ | — | 75  | 52       | 35        | 10.73    | 10.69     | 0.54      | 0.09      | 28   | 1       |
|           | 600  | 62  | 45       | 30        | 9.20     | 9.23      | 0.57      | 0.10      | 28   | 0.86    |
|           | 650  | 61  | 50       | 34        | 8.16     | 8.15      | 0.63      | 0.11      | 28   | 0.76    |

Abbreviations: $L_{60}$ = mean length for >60° to c-axis (µm); $L_{all}$ = mean length for all crystallographic directions (µm); $n$ = number of grains scanned; $N_{60}$ = number of measured tracks for directions >60°to c-axis; $N_{all}$ = number of measured tracks for all crystallographic directions; $r_{60}$ = normalized value of $L_{60}$ by that of unannealed tracks; $SD_{60}$ = standard deviation for the length distribution of tracks for >60° to c-axis (µm); $SE_{60}$ = standard error of $L_{60}$ (µm); $t_e$ = etching duration (hr); $T$ = annealing temperature (°C).  

$^a$Zircon grains of these samples were irradiated with thermal neutrons prior to laboratory annealing.
measurements of etched FTs were carried out under an optical microscope (Nikon Eclipse® E600 microscope with 100× dry objectives and 10× eyepieces) coupled with a digital camera and image analysis system at Kyoto University. Horizontal confined track lengths were measured using tracks-in-track (TINTs; Lal, Rajan, & Tamhane, 1969), with precision of ±0.1 µm (1σ). TINTs were searched and analysed only for those having 1.0 ± 0.5 µm etch width, along with their crystallographic orientations. We only considered horizontal confined tracks lying at an angle >60° relative to the c-axis to calculate the mean value (i.e. $L_{60}$ in Table 2), because the impact of anisotropy is negligible for that azimuth angle interval (Hasebe et al., 1994; Yamada et al., 1995).

The Hf contents of zircon samples were measured with analytical precision of ±0.01% using the LA-ICP-MS system installed at Kanazawa University. Multiple analyses were performed on each sample aliquot using 3–5 grains, following the experimental procedure described by Morishita et al. (2009).

3 | RESULTS

Table 2 shows the analytical results of track length measurements of unannealed and annealed aliquots for the nine zircon samples. Figure 1 presents a plot of mean confined track lengths ($L_{60}$) against annealing temperature. The mean confined track lengths measured on unannealed aliquots range from ~10.5 to ~11.2 µm for the nine zircons, approximately concordant with each other, and also with those of previous studies (Hasebe et al., 1994; Yamada et al., 1995). With increasing annealing temperature, the observed track lengths show consistent and systematic reductions, as observed previously (Kasuya & Naeser, 1988; Yamada et al., 1995). The observed length reductions in spontaneous FTs are approximately concordant, with some outliers, among the four samples of different ages and spontaneous track densities (red in Figure 1). For the five young zircon samples

![FIGURE 1](image1.png)

**FIGURE 1** Variation in mean confined track lengths ($L_{60}$) of individual sample aliquots that were annealed for 1 hr at elevated temperatures. The observed mean lengths of spontaneous (Sp) FTs as well as the mixtures of spontaneous and induced (In) FTs are all approximately concordant among the nine zircons at individual annealing temperatures, whereas mean lengths are significantly shorter than the mean length of induced FTs in pre-annealed zircon at the advanced stage of annealing (i.e. ~650°C and ~700°C). $L_{60}$, mean length of horizontal confined tracks lying at an angle >60° relative to the c-axis; blue symbols, data for induced FTs in pre-annealed NST zircon (Yamada et al., 1995); red symbols, data for spontaneous FTs in four zircons older than 16 Ma; green symbols, data for the mixture of spontaneous and induced FTs in five zircons younger than 3 Ma. All error bars represent ±1 standard error of the mean [Colour figure can be viewed at wileyonlinelibrary.com]

![FIGURE 2](image2.png)

**FIGURE 2** Mean confined track lengths ($L_{60}$) of individual sample aliquots that were annealed for 1 hr at three different temperature conditions (i.e. unannealed, 600 ± 2°C and 650 ± 2°C), plotted versus (a) age and (b) spontaneous track density. At individual temperature conditions, no systematic differences are found for mean track lengths of the nine samples, including young zircons having low spontaneous track densities. Solid circles represent data for spontaneous (Sp) FTs in four zircons older than 16 Ma, whereas open circles represent data for mixtures of spontaneous and induced (In) FTs in five zircons younger than 3 Ma. All error bars represent ±1 standard error of the mean [Colour figure can be viewed at wileyonlinelibrary.com]
containing a mixture of spontaneous and induced FTs (green in Figure 1), the observed length reductions are indistinguishable from each other, and approximately concordant with those of spontaneous FTs. However, mean FT lengths of all those samples are significantly shorter than the induced FT lengths in sample NST (Yamada et al., 1995; blue in Figure 1) at an advanced stage of annealing (i.e. ~650 and ~700°C).

4 | DISCUSSION

4.1 | Comparison among natural zircons

To better compare the results between the nine zircons of different ages, observed mean track lengths were plotted at three different annealing stages (unannealed, 600°C and 650°C) against (a) age and (b) spontaneous track density (Figure 2). At individual temperature conditions, no systematic differences were found for mean track lengths of the nine samples. Even at an advanced stage of track length reduction at 650°C, annealed lengths are indistinguishable at the two standard errors (SE) level from each other among the nine samples, including those young zircons having low spontaneous track densities. Hence, the present data suggest that the nine samples have very similar FT annealing behaviour at the laboratory time-scale of heating, regardless of their age that ranges from ~0.6 to ~70 Ma and of their spontaneous track densities that ranges from ~0.05 to 7 × 10^6 cm^-2. Along with previous results (Kasuya & Naeser, 1988), this implies that identical annealing kinetics may apply for most of the Late Mesozoic and Cenozoic zircons.

Variability in FT annealing behaviour among natural zircons may also be introduced by Hf, which is commonly the most abundant minor element in this mineral. We performed LA-ICP-MS analyses of the nine zircons to determine their Hf content, which ranges from 5.5 ± 0.4 to 8.4 ± 1.1 mg/g (Table 1). We can thus conclude that identical kinetics are applicable to the Late Mesozoic and Cenozoic zircons having Hf composition within the observed range.

4.2 | Comparison between natural zircons versus pre-annealed zircons

Previous studies (Kasuya & Naeser, 1988; Yamada et al., 1995) showed that induced FTs in pre-annealed zircons are more resistant to thermal annealing than spontaneous FTs in natural zircons, which was confirmed by the comparison of the results of this study with the data by Yamada et al. (1995). In addition, we observe that the thermal annealing behaviour of spontaneous FTs is concordant with that of the mixture of spontaneous and induced FTs, regardless of the mixing ratios (Figure 2, Table 1). Specifically, for the three samples with spontaneous track density of ~0.1 or less (i.e. K1018, Nz00-22 and 75G104), about 99% of the mixed populations are induced FTs that underwent laboratory annealing. This suggests that induced FTs in pre-annealed zircons are more resistant to thermal annealing than induced FTs in natural, non pre-annealed zircons.

What is the reason for such a different behaviour? Key factors to be considered are the temperature and time conditions concerned with thermal pre-annealing. The palaeo-temperature of magmatic zircons at the time of their formation is likely in the range of ~550°C to 900°C for geological time durations related to zircon morphology (Pupin, 1980), whereas the laboratory heating conditions to pre-anneal spontaneous FTs in zircons are 800°C for 24 hr (Kasuya & Naeser, 1988) or 1,009 ± 2°C for 2 hr (Yamada et al., 1995). Although zircon formation is kinetically controlled and thus its formation temperature cannot be directly compared with that of laboratory annealing, those rather high temperatures of pre-annealing may have modified some crystallographic properties controlling the FT annealing rate.

Another possible factor is the difference of time periods that elapsed since the last cooling episode to ambient temperature until the time of laboratory annealing of FTs. While the pre-annealed zircons experience merely a few to several months after the last cooling at laboratory (subsequent to the pre-annealing), zircons without pre-annealing were kept unheated for geological time periods (Table 1). During such extended periods of time, some temporal change of zircons, such as the accumulation of radiation damage, may have preceded to change the crystallographic ordering/structure and thus the FT annealing rate. Because of the approximately identical annealing behaviour among zircons that range in age from ~0.6 to ~70 Ma (or in spontaneous track density from ~0.05 to 7 × 10^6 cm^-2), it is implied that the structural change may proceed quickly in zircons after their thermal resetting and is likely to reach saturation within 0.6 m.y. (or with accumulation of spontaneous tracks of ~0.05 × 10^6 cm^-2).

5 | CONCLUSIONS

1. As a result of 1 hr isochronal heating, confined track lengths in nine natural zircons, separated from rapidly cooled volcanic rocks, show consistent and systematic reductions with increasing annealing temperature.

2. No systematic difference was found for FT annealing characteristics among natural zircons of ~0.6 to ~70 Ma age (and spontaneous track density of ~0.05 to 7 × 10^6 cm^-2).

3. The present results, coupled with previous data, imply that identical annealing kinetics can work for many of the Late Mesozoic to Cenozoic zircons, with Hf contents ranging from 5.5 ± 0.4 to 8.4 ± 1.1 mg/g.

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