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研究ノート

Zircon fission-track technique: a laboratory procedure adopted at the Institute of Geology, Academy of Sciences of the Czech Republic, v.v.i.

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Introduction

Along with many other geochronological methods, fission-track analysis is a technique used for the reconstruction of time-temperature evolution of rocks. The fission-track dating method offers easy dating of rocks without using high-priced mass spectrometers and has been applied to many low-temperature geological problems. This paper describes laboratory routine procedures for fission-track analysis (FTA) of zircons including separation, mounting, grinding/polishing, chemical etching, thermal neutron irradiation in nuclear reactor, counting and determination of zeta constant. All procedures of zircon fission-track analysis using an external detector method described here is applied at the Institute of Geology, Academy of Sciences of the Czech Republic, v.v.i. For the details of zircon fission-track method, see Tagami (2005) and references herein.

Samples preparation

A procedure of fission-track sample preparation described in the following chapters (A-D) is adopted from Tagami (1988) and Tagami (2005):

A. Mineral separation

Rock samples collected for mineral separation are in weight from 250 g to 25 kg. The following steps are undertaken to separate the minerals required for analysis: (1) Sample crushing; (2) Wilfley table separation method; (3) Magnetic and heavy liquid separation. Institute of Geology (Academy of Sciences) ho-

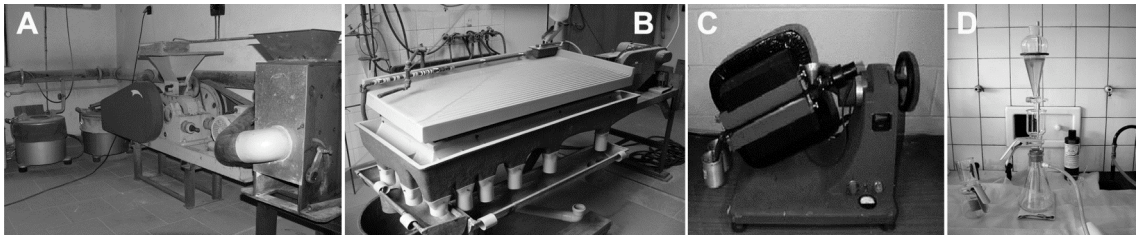

Fig. 1　Equipments at the Institute of Geology for mineral separation: A) mills and jaw crushers; B) Wilfey table; C) Frantz separator for separation of magnetic and non-magnetic minerals; D) separatory funnel for heavy liquid (bromoform and diiodomethane) separation.

mogenization laboratory is equipped with mills and jaw crushers, fume hoods that are used to ground rock samples to fine grained particles (Fig. 1A). Next step is a separation by a shape/density separator known as the Wilfley vibrating table (Fig. 1B), which is used as a quick, reliable and consistent method for de-sliming bulk powder samples. The rock's mineral mixture is placed on the high side of the surface of the Wilfey table and water is flowed over it to move the grains while the surface is vibrated. Heavier grains collect in the grooves while lighter grains are washed down the surface. The third step is a separation by two Franz magnetic separators (Fig. 1C), with variable magnetic field strength, sample feed-through rate, and angle and tilt of sample feed-through. After separation of ferromagnetic minerals (e.g., magnetite), non-ferromagnetic minerals are separated in the isodynamic magnetic separator and composites of magnetic minerals (e.g., biotite, hornblende, pyroxene, ilmenite, epidote) are removed. The non-magnetic fraction that contains datable minerals such as zircons and apatites (plus quartz, fluorite, etc) is then subjected to heavy liquid separations. In next step, a mixture of minerals is placed in the separatory funnel (Fig. 1D) with heavy liquid, the grains with densities lower than that of the liquid will float and grains with densities greater than the liquid will sink. Suitable liquids for density separation include bromoform (density = 2.84 g/cm^3) and diiodomethane (density = 3.32 g/cm^3). Final pure mineral concentrates are achieved through hand-picking individual grains using an optical binocular microscope (Nikon SMZ 800) with fiber-optic lights.

B. Handpicking and mounting

In order to mount the grains into teflon sheets, small fraction of zircon grains is spread on a silica glass slide of size about 5×5 cm^2. Silica glass is preferably used, because the slide is heated during mounting on a hot plate. During handpicking a small quantity of alcohol is put on the glass slide and with a thin paintbrush the zircons are arranged one by one in such a manner that c-axis of all the grains are in one direction (Fig. 2). The zircon grains should be selected under the criterion of large clear crystals (e.g., equant grains, prismatic stubby or prismatic euhedral-needles grains) having well-defined c-axes. After handpicking, all the remaining grains are removed from the glass slide by use of a dry paintbrush and stored in a paper or a plastic pack with a proper label.

The silica glass slide is then shifted onto a hot plate at a temperature of 315°C. A second silica glass plate of size about 3×3 cm^2 is also pre-heated on the same hot plate. A sheet of 0.50 mm thick PFA Teflon® is cut to the size of 1.5×1.5 cm^2. The Teflon sheet is held vertically using tweezers on the silica glass slide and heated for about 30 sec. until its bottom end melts slightly. Then it is tilted to allow falling down on the glass slide. The pre-heated glass plate is put on the Teflon®

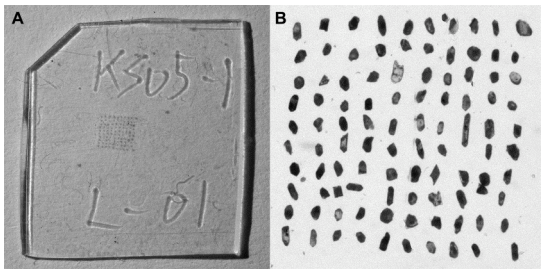

Fig. 2 A) Photo of zircon grains mounted in PFA Teflon (size of sheet is 1.5×1.5 cm^2). B) Detail of previous mount with zircon grains arranged along their c-axis in a square of 10×10.

Fig. 3 Photo of individual steps during mounting procedure for zircon grains into PFA Teflon: A) Silica glass slide with studied zircons shifted on hot plate; B) PFA Teflon sheet falling by gravitation on zircons; C) Tweezers put pressure on small silica glass to remove bubbles from PFA Teflon. Finally, zircon grains are pushed into PFA Teflon and are ready for next procedures.

sheet and pressed gently so that the grains are fixed into the sheet (Fig. 3). The glass slide with the glass plate and Teflon® sheet having zircon grains is removed from the hot plate, and then pressed with a metallic plate to be kept flat during cooling. The sample code is written with a needle pen on the backside of the Teflon® mount after it is taken out from the glass slide and plate. In case of FTA on zircons, it is not preferred to grind zircon crystals along their crystallographic c-axes, because it often produces fractures or damages of the surface, which are enlarged during chemical etching and disturb the track length measurement and counting. Thus zircon grains should be arranged with their c-axes toward the same direction in a mount, and then all grains are ground in one direction (perpendicular to c-axes). In our routine, 64 to 144 mineral grains (within array ranges from 8×8 to 12×12) are arranged and mounted as mentioned above (Fig. 2).

C. Grinding and polishing

As mentioned above, FT length measurement and counting of minerals are carried out on a polished internal surface of mineral grains under an optical microscope. Flat internal surfaces of the mineral grains are exposed by grinding and polishing procedures. In order to stabilize the mount during grinding and polishing, an acrylic block is pasted on grinding and polishing machine and fixed together with zircon mounted grains by using double-coated tape. For using internal surface of zircons, it is essential to remove a certain thickness to expose 4π geometry for FT dating. The thickness corresponds to a half of etchable track length that is ~6 μm for zircon grains (Krishnaswami et al., 1974).

Before 6 μm grinding, the crystal surface of mineral grains must lie flat at the surface of the mount. The mount with zircons is ground gently several times in one direction perpendicular to the c-axis on the wet emery paper of 1500 mesh size, and observed under an optical microscope for the grinding scratches. It is continued until the grinding scratches are observable on the most of grains. The observable area of them will be sufficiently exposed 4π surface after 6 μm grinding, and thus must be described and recorded by convenient photographs or by sketches. For FT length measurement, it is not necessary to describe the surface of mineral grains.

The next step is to grind the mount and remove a certain thickness of grains for exposing 4π geometry. Tagami et al. (1988) measured the depth of the grinding scratch using wetted emery paper of 1500 mesh, which was at least

1 μm depth. Therefore the grain minerals are removed for at least 1 μm in thickness after a series of grinding and polishing processes. Thus the series of the processes is repeated six times, and then the mineral grains will be ground 6 μm at least, that is, exposed their 4π surfaces.

After the grinding, the mount is polished with 15 μm and 3 μm diamond paste (Struers Company). In case of the Institute of Geology, Academy of Sciences, Struers DP-Dac (for 3-6 μm abrasive grain size of diamond paste) and DP-Dur (for 1 μm abrasive grain size of diamond paste) polishing clothes are used for polishing. The grinding and polishing system (MTH Company Kompakt 1031) is equipped with a specimen mover and an extension head system (APX 010). The specimen mover can rotate and weight itself on a polishing cloth. The duration, velocity and weight of polishing depend on the hardness of mineral grains. Fish Canyon Tuff zircon standards are polished for 10 min. to remove the grind scratch with 15 μm paste using 10 N weighted specimen movers (75 rounds/min.), and it also takes 10 min. to remove the 15 μm polishing scratch with 3 μm paste (90 rounds/min.). After finishing grinding and polishing, the mount is cleaned with alcohol in an ultrasonic bath and the mount is ready for etching.

D. Etching of spontaneous fission-tracks in minerals

Fission-tracks in mineral grains can be enlarged and made visible in an optical microscope by chemical etching. Latent fission tracks are only ~1 nm in width, invisible under the microscope. For etching of zircon mounts, we use an electric hot plate with aluminium holes for keeping Teflon® beaker containing the eutectic NaOH:KOH (1:1) etchant. The temperature of the heater in the hot plate is controlled by a precise thermocouple and a temperature controller. This system provides a stable heating up to 500°C with ± 1°C accuracy. The temperature of the etchant is also monitored with a Teflon® coated thermocouple, which is dipped inside the beaker. The required temperature of the etchant is 250° ± 1°C for FT length measurement and 225° ± 1°C for FT dating. Approximately a half-day is necessary time to achieve a proposed temperature of the etchant.

When the etchant is prepared, three or four zircon mounts are put into each beaker for etching. The etching procedure can control the width of measurable tracks between under-etched and over-etched condition. According to etching criteria described by Yamada et al. (1995), the width of the surface tracks is 2 μm for FT length measurement and 1 μm for FT dating. The etching duration depends on a density of tracks in zircon grains (Hasebe et al., 1994). The etching time of Fish Canyon Tuff zircon standard (ρ_s ~5×10^6 cm^{-2}) is about 25 hours and that of Bulk Member zircon standard (ρ_s ~5×10^6 cm^{-2}) is about 50 hours in our system (1 μm criterion at 225° ± 1°C). After taking out the mounts from the etchant, it is cleaned in 5% HCl with ultrasonic cleaner for 10-15 minutes in order to remove the etchant from the grains and from Teflon® sheet. The suitable lifetime of an etchant is one week, and then it must be replaced by a new one.

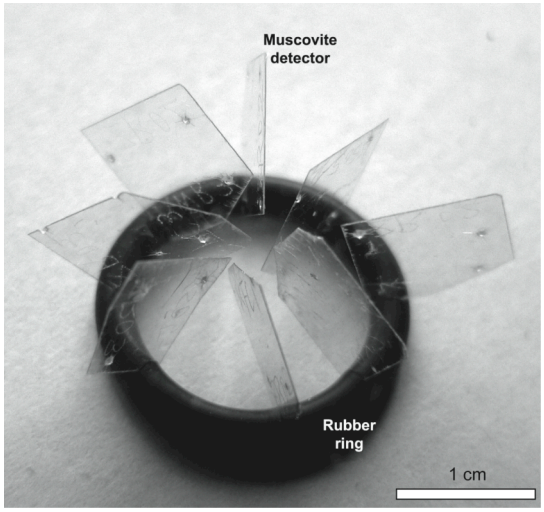

Fig. 4 Photo of muscovite external detectors attached in a rubber ring before etching mica sheets to reveal the induced tracks.

E. Packing and irradiation of samples for irradiation, etching of induced fission-tracks

In the external detector method (Fleischer and Hart, 1972), a mica external detector (ca. 10×10 mm; Fig. 4) is placed over the grain mount during the thermal neutron irradiation. After mica detector is put on the mount (the sample code is written with a needle pen on the back of the mica), the detector is covered with a clean plastic sheet of size slightly larger than the mica sheet, and then they are wrapped up with a tape and the sample code is written on it. The dosimeter glass (IRMM 540R, ~15 ppm U) for monitoring thermal neutron fluence is also attached to the mica sheet as well as the zircon mount.

After micas are attached to mineral mounts and to dosimeter glasses, samples are stacked and packed in a PE-tube for irradiation in a nuclear reactor. The Oregon State University TRIGA Mk. II Research Reactor (OSTR) is used for our purposes. The OSTR operates at maximum steady state power of 1.1 MW and is equipped for thermal neutron irradiations suitable for fission track geochronology.

After thermal neutron irradiation, the mica detectors and plastic sheets are removed from the mounts. During irradiation, the induced fission tracks are recorded in the mica detector. To make the induced tracks visible, the micas are fixed in a rubber ring with several cuts for stabilization of micas (Fig. 4), and then the ring with micas is put into 38% HF at 32.0°C for 8 minutes. After etching, all samples and micas are washed in distilled water with an ultrasonic cleaner for a few minutes. After washing, the samples and corresponding micas are ready for counting of the fission-tracks (Fig. 5). Densities and lengths of spontaneous tracks and densities of induced tracks are measured at the Institute of Geology, Academy of Sciences on Axioplan 2 (Zeiss) microscope equipped with Autoscan™ stage.

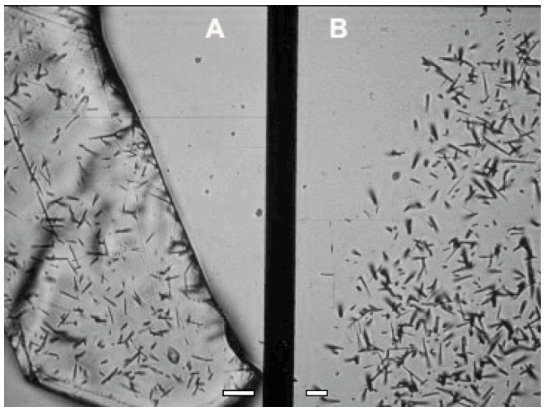

Fig. 5 Spontaneous tracks (A) and (B) induced tracks recorded on mica external detector in zircon Fish Canyon standard. Scale bar is 15 μm.

Concluding remarks

To overcome the uncertainties that are difficult to measure (e.g., the uncertainty of neutron flux, individual counting error), the FT community adopted a system of calibration based on the comparison of sample measurement with the analysis of mineral standards and glass monitors—the zeta calibration ζ (Hurford and Green, 1982). The fission-track

zeta value is an individual constant different between the fission-track geochronology laboratories. The zeta value is calculated from the following equation:

$$\zeta = \frac{\rho_i}{\lambda_D g \rho_s \rho_d} \exp(\lambda_D t_{std} - 1)$$

where λ_D is total decay constant of ^{238}U (1.55125×10^{-10} yr^{-1}; Steiger and Jäger, 1977); ρ_s is the spontaneous fission track density of a standard sample; ρ_i is the induced fission track density of a standard sample measured in a muscovite detector; g is the geometry factor (= 0.5); ρ_d is the induced fission of the dosimeter glass measured in a muscovite detector; t_{std} is the reference age of the standard sample. The statistical error of zeta was calculated by the conventional analysis (Hurford and Green, 1983) adopted in Trackkey program (Dunkl, 2002) that is routinely used for apatite calculation. The results of zeta calibration are in Table 1. Each zeta value was calculated using individual ρ_s, ρ_i and ρ_d values and reference ages of standard zircons, and then six individual zeta values were obtained from two standards (188.3 ± 8.6, 165.7 ± 9.2, 145.2 ± 8.2 for BM, and 209.3 ± 6.1, 181.9 ± 5.2, 212.1 ± 9.6 for FC-3; Table 1). We calculated two sample weighted mean zeta values (SMWZ) using three zeta values in each standard (165.7 ± 5.0 for BM, and 196.1 ± 3.6 for FC-3). We noted that a weighted mean of a discrete set of numbers with weights was given by a sum of multiplications between individual zeta and weight values. Weight zeta value was a reciprocal of squared standard deviation and weighted average FT age for real samples will be calculated as T/(standard deviation)^2. Finally, overall weighted zeta value (OWMZ) of our laboratory was a weighted mean of two zeta values from zircon standards calculated above, and is published as 185.7 ± 2.9 (Table 1). One of the recommendations by Hurford (1990) for the standardization of FT dating states that the standard should be irradiated at least three times. Finally, we consider above published zeta value as a preliminary constant and for next dating will be zeta value re-counted as a personal individual value.

Acknowledgements

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Table 1 Fission-track zircon analytical data for zeta calibration and sample weighted mean zeta values (SWMZ) for fission-track laboratory at the Institute of Geology, Academy of Science, Praha.

Sample code		ρ_s (N_s) [$\times 10^6$ cm^{-2}]	ρ_i (N_i) [$\times 10^6$ cm^{-2}]	ρ_d (N_d) [$\times 10^6$ cm^{-2}]	n	$P(\chi^2)$ (%)	Zeta (± 1σ)
BM	ZR1	1.171 (649)	4.477 (2482)	0.6669 (8011)	32	89	188.3 ± 8.6
	ZR2	0.883 (428)	3.202 (1552)	0.7185 (8631)	31	98	165.8 ± 9.2
	ZR3	1.110 (430)	3.526 (1366)	0.7185 (8631)	24	93	145.2 ± 8.2
						SWMZ	165.7 ± 5.0
FC-3	ZR2	5.763 (1904)	14.39 (4753)	0.6669 (8011)	34	12	209.3 ± 6.1
	ZR3	5.630 (2154)	12.21 (4671)	0.6669 (8011)	39	<1	181.8 ± 5.2
	ZR4	4.206 (725)	10.64 (1834)	0.6669 (8011)	19	19	212.1 ± 9.6
						SWMZ	196.1 ± 3.6
						OWMZ	**185.7 ± 2.9**

ρ_s = spontaneous fission track density; N_s = total number of spontaneous fission tracks counted; ρ_i = induced fission track density; N_i = total number of induced fission tracks counted; ρ_d = induced fission track density in a dosimeter glass; N_d = total track number to determine ρ_d; n = number of grains counted; $P(\chi^2)$ = probability of obtaining the observed value of Galbraith's (1981) χ^2 parameter, for n degrees of freedom, where n = number of crystals -1; SWMZ—sample weight mean zeta; OWMZ—overall mean zeta.

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