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Fission-track analysis using LA-ICP-MS: laboratory procedures adopted at joint Low-temperature Thermochronology laboratory at Sofia University and Geological Institute, BAS

Датиране по метода на следите с LA-ICP-MS: лабораторни процедури, използвани от съвместната нискотемпературна термохроноложка лаборатория на Софийски университет и Геологически институт, БАН

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Introduction

Fission-track (FT) method is a technique used for dating of geological events based on the accumulation of damage trails (fission tracks) left from fragments produced by the spontaneous nuclear fission of ^{238}U in natural minerals and glasses. After the first experiments performed by Price and Walker (1963) the method developed very fast and during the last decades the fission-track analysis became a widespread approach for reconstruction of the thermal history of rocks from the shallow levels of the Earth's crust at temperatures generally between 60 °C and 300 °C. For the details of FT method see Donelick et al. (2005) and Tagami (2005). In the 80s some experiments based on fission-track dating on micas were performed in Bulgaria (Monchev, 1985 and references therein) but the method was not further applied and developed.

Here we present the adopted procedures of preparation and processing of apatite and zircon samples for fission-track dating using LA-ICP-MS for the first time in Bulgaria. Different laboratory procedures and strategies for sample preparation and measurement of the essential track parameters already exist (e.g. Tagami et al., 1988; Kohn et al., 2019). We have tried to adapt them to the conditions available in Bulgaria, whereas some own, specific approaches were also applied. The new joint Low-temperature Thermochronology laboratory was established in a close collaboration with Kyoto University, Japan.

Sample preparation and fission-track etching were processed in the Chemical laboratory of the Geological Institute, Bulgarian Academy of Sciences. Fission track observations and measurements were performed in the Microscope Laboratory in the Department of Geology, Paleontology and Fossil Fuels, Sofia University.

Sample preparation

We have performed the analyses on two fission-track standards – Durango apatite and Fish Canyon Tiff zircon with well-known FT age, as well as on several magmatic (intrusive and effusive) rocks with Cretaceous and Carboniferous ages from the region of Central Sredna Gora Zone.

At least 100 apatite and zircon grains from each sample were separated by standard mineral separation procedures. Further, the most transparent and euhedral crystals, without visible inclusions or fractures and almost of same size were chosen. The apatite grains were arranged in arrays of 7 by 7 or 8 by 8 on a plexiglass plate covered by double sided tape and then mounted in epoxy resin. The mounts were dried for more than 24 hours at room temperature and then detached from the tape. The zircons were arranged in arrays of 5 by 4 grains on a silica glass with their C-axes set in one direction. The silica glass with the arranged zircons and another silica glass were put on a hotplate heated to temperature of 345 °C for 1 min. After heating of the two glasses, a small PFA (tetrafluoroethylene) Teflon sheet (about 1 cm by 1 cm by 0.5 mm) was held vertically with tweezers on the silica glass at about 0.5 cm away from the outer zircon array for at least 10 seconds. In this way its bottom, that touched the glass, was melted and stitched to the glass. Then the whole Teflon sheet was tilted to fall slowly on the zircons. The second heated glass was placed over the Teflon sheet and pressed so that the grains become embedded in the partly molten Teflon. After cooling the glasses are easily detached from the Teflon.

After mounting, pre-grinding, grinding and polishing of the mounts was performed. Each step of these procedures was surveyed by the observation under a microscope. The aim of pre-grinding is maximum possible area of each grain to be exposed. Pre-grinding was performed on 1200 grit silicon carbide paper (wetted with water) in one direction for both apatite and zircon mounts. Depending on the size and specific characteristics of the grains, every mount needed different time for pre-grinding (from 5 to 80 times). The grinding was performed so that at least 8 μm from apatites and 6 μm from zircons to be removed – thickness that corresponds to half of the track length for each mineral. Thereby an internal surface of the crystal was obtained which is intersected by tracks coming from both sides (above and below). This is the so called 4π geometry necessary for correct FT dating. As it is considered that the depth of a grinding scratch is approximately 1 μm, for the apatite grains grinding was done 8 times changing the direction every time by 90° so that we could be sure that the previous scratches were completely removed. Due to the fact that zircons are very easily breakable parallel to C-axis direction, the pre-grinding and grinding of zircons was performed only perpendicular to C-axis. Grinding was done 6 times and every time the scratches were consequently removed by 2500 grit silicon carbide paper applied parallel to C-axis. The apatite and zircon mounts were then polished successively with 6 μm, 3 μm and 1 μm diamond pastes, changing every time the direction of polishing by 90°. The ready mounts were consequently washed in an ultrasonic cleaner.

Fission track etching

In order to reveal the fission tracks and make them visible under an optical microscope, chemical etching of the samples was performed. The apatite

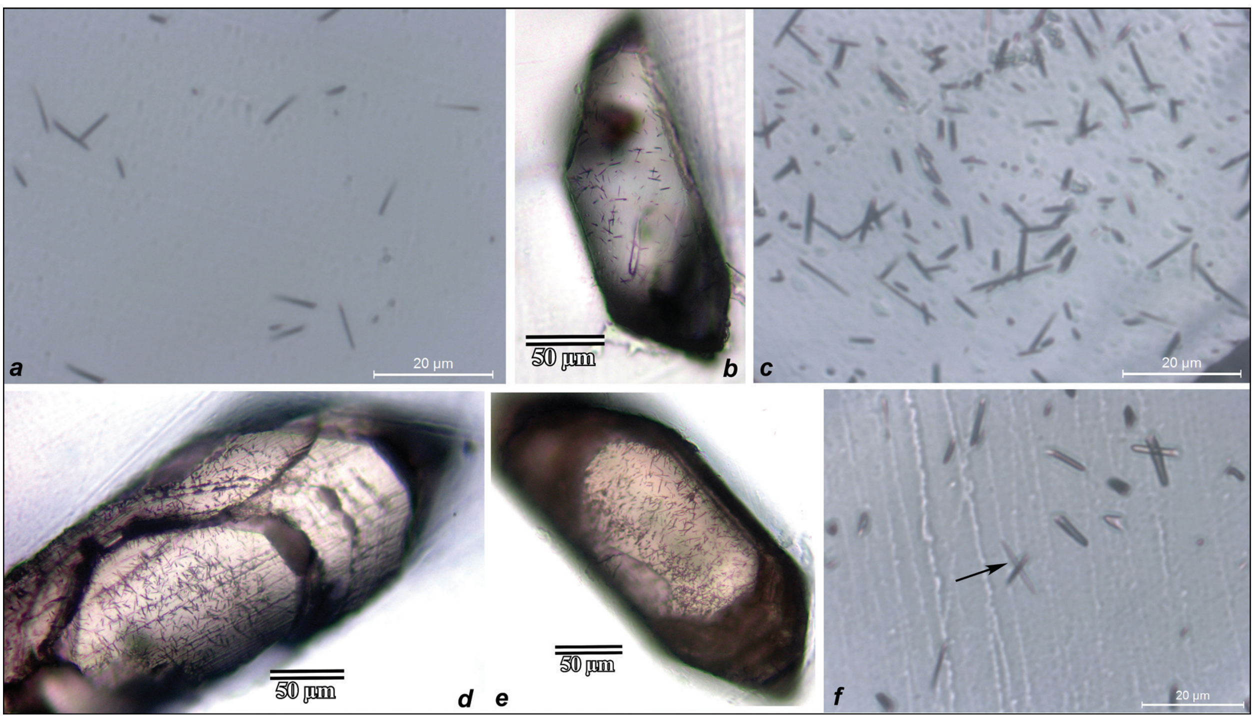

Fig. 1. Fission tracks in etched grains: *a*) Durango apatite; *b*) Fish Canyon Tuff zircon; *c*) high track density in an apatite from Carboniferous granite from Sredna Gora Zone (SGZ); *d*) high track density and zonation in a zircon from Cretaceous monzonite, SGZ; *e*) high track density and zonation in a zircon from Carboniferous granite, SGZ; *f*) a horizontal confined track (the so called TINT pointed by the arrow) intersected by another track in an apatite from Cretaceous subvolcanic body, SGZ.

mounts were etched in 5.5 N HNO_3 for 20 seconds at temperature of 21 °C (Donelick et al., 2005). The zircon mounts were etched in KOH:NaOH eutectic mixture heated in an oven at temperature of 230 °C. The Fish Canyon Tuff zircon standard was etched for 24 hours at one step. The other samples were etched between 14 and 24 hours at two or three steps. As sometimes the zircon grains from one and the same mount need different etching time, it is practically impossible to etch all grains at the same extend. Therefore, we have stopped etching one mount when the tracks in the most suitable for counting grains were well revealed.

Fission track measurements

Fission track counting in apatites and zircons as well as track length and pits diameter measurements in apatites were performed under a Leica DM 2 500 POL optical microscope using x100 dry objective with a total magnification of x1000.

Track counting. Track counting was performed only on surfaces parallel to the crystallographic C-axis of the best polished and free of inclusions and fractures grains. The size of the counted area is determined by a graticule net 10x10 mm fitted in one of the eye-pieces of the microscope. First, we have counted tracks in Durango apatite (Fig. 1a) and Fish Canyon Tuff zircon (Fig. 1b) in order to compare their well-known FT ages with the ones obtained in our lab. For each apatite samples, at least 20 grains were analyzed. Some of the Carboniferous rocks had much higher track density (Fig. 1c) compared to Durango apatites (Fig. 1a). Unfortunately, the quality of zircon grains was often not good enough for track counting due to zonation and the high density of tracks in both Carboniferous and Cretaceous rocks (Fig. 1d, e). This is due to the relatively high U content of the samples and the fact that most of them had not been affected by high-temperature events after their formation. Consequently only 5–10 zircon grains from each sample were counted.

The number of tracks counted over a certain area will be used together with the measured ^{238}U content for calculation of a single grain FT ages.

Track length and track pits diameter measurements. The track lengths were measured of horizontal confined tracks which were revealed by the etchant penetrating to them via other fission tracks of fractures (Fig. 1f). Due to the fact that the track lengths depend on their orientation to the C-axis of the crystal, their angle to this crystallographic axis was also measured. For the grains, where tracks were counted for age calculation and those, where confined horizontal tracks were measured, the average diameter of track pits was also measured and calculated. The measured track lengths and pits will be used further to constrain the thermal histories of the analyzed samples.

The LA-ICP-MS measurement of ^{238}U

In order to calculate the fission-track age, ^{238}U concentration is needed to be directly measured on the same grains and counted areas using LA-ICP-MS. The ablation pit is 35 μm in diameter with beam repetition rate of 7 Hz. ^{238}U concentrations for both apatites and zircons were calibrated against NIST 610 and NIST 612 standard glasses. Ca and Si were used as an internal elemental standard (assuming Ca is essentially stoichiometric in the targeted apatites and Si in zircons). The elements concentrations were calculated using SILLS software.

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