Improvement and development of dating methods using radiation damage for reconstructing Quaternary volcanic history

学　位　論　文　要　旨

博士論文

Improvement and development of dating methods using radiation damage for reconstructing Quaternary volcanic history

第四紀火山活動史解明のための放射線損傷を利用した
年代測定法の改良と開発

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Summary

Widespread tephras especially distributed by explosive eruptions are important time markers to correlate the geology of different regions. Several radiometric dating methods have been applied to Quaternary tephras, however each method has its advantages and disadvantages.

In this doctoral thesis, I focused on the potential of fission track dating method using volcanic glass.

I proposed a new method to estimate the track number per unit volume and corresponding age equation. This new dating protocol has the potential to provide reliable fission-track ages without nuclear irradiation and complicated age correction procedures. I constructed an image processing protocol to reduce time-consuming labor works in counting tracks on many screens. This new counting protocol has the potential to provide reliable fission-track ages easily. I also tried to develop a new age determination method in order to offer additional dates for cross-check. I observed young zircons with the atomic force microscope (AFM), and compared to AFM images from old zircon. The cyclic surface structures observed on old zircons were not observed in young zircon, therefore, they may be the results of alpha - recoil tracks which must be in this particular old zircon in the order of 10^{10}/ cm^2.

Abstract

The "Quaternary", for which the human race becomes adapted to the Earth environment, is the most recent geological period since 2.58 Ma. Among many

environmental factors, a volcanic activity causes damage directly to human beings and volcanic products emitted to the atmosphere would affect the climate (Fig. 1.1). An explosive eruption that largely affects the global environment spreads the volcanic products widely over the world. Widespread tephras especially distributed by explosive eruptions are important time markers to correlate the geology of different regions (e.g. Borchardt et al., 1971; Heiken and Wohletz, 1985; Wicox, 1965). Several radiometric dating methods have been applied to Quaternary tephras. K-Ar method can be applied to several minerals (e.g. feldspars, mica, amphibole or volcanic glass) which are included in tephras (e.g. Bailey et al., 1976; Izett et al., 1970; Izett, 1981; Izett, 1982; Smith, 1979), however, K-rich minerals in tephra sometimes lose radiogenic Ar during secondary hydration and devitrification, which is a specific phenomenon for glass, and hence underestimated ages were reported (Kaneoka, 1972; McDougall and Harrison, 1988). Radiocarbon (^{14}C) method is applicable to inorganic or organic deposits (e.g. Aitken, 1990; Black, 1975; Hildreth, 1981; Miller and Smith, 1987), though materials containing carbon are rarely associated with active lava flows and the technique's dating range is limited to up to around 40,000 years (Fattahi and Stokes, 2003). Luminescence method is also able to apply to several minerals (e.g. feldspar, glass and quartz) in most volcanic products (e.g. Berger, 1992), though problems of the method include such as a type of signal instability called *anomalous fading* of blue luminescence from feldspar (Wintle, 1973; Wintle, 1974); the poor signal-to-noise ratio of quartz and glass thermoluminescence and optically stimulated luminescence analyses (Berger and Huntley, 1994; Miallier et al., 1991). Fission track (FT) dating method has been often applied to zircon or volcanic glass in Quaternary tephra (e.g., Gentner et al., 1969; Ito and Hasebe, 2011; Naeser et al., 1973; Seward, 1979; Stozer and Wagner, 1969; Walter, 1989; Westgate, 1989). For dating zircon by FT method, a zeta calibration (Hurford and Green, 1982; Hurford and Green, 1983), which needs a neutron irradiation in a reactor, is recommended by the Subcommission on Geochronology (Hurford, 1990). Finding zircons in Quaternary tephra is sometimes difficult due to chemical composition or fractionation in mineral assemblages during the eruption-transportation-occurrence processes. On the other hand, FT method using volcanic glass is applicable to many kinds of tephras because volcanic glass is contained in almost all tephras. However the FT method using volcanic glass needs observation of wide surface area to obtain statistically significant number of tracks because glass contains less uranium compared to zircon or apatite, and target ages is Quaternary, which is relatively short time period to produce tracks in most cases (e.g., Moriwaki et al., 2008; Shane et al., 1995; Walter, 1989). Additionally FT in volcanic glass became reduced in size at ambient temperatures, therefore, several correction procedures were proposed (e.g. Gentner et al., 1969; Stozer and Wagner, 1968; Westgate, 1989). The conventional correction procedure takes a long time (30 - 100 days) and also requires the use of radioactive material produced by thermal neutron irradiation at the nuclear reactor. Thus, each method has its advantages and disadvantages. Therefore multiple analyses by several dating methods are needed to know the history of the volcanic activity for further information.

Recently FT method using volcanic glass has rarely applied to the tephra because of complicated and time-consuming corrections in spite of a large amount of volcanic glass being contained in the variety of widespread tephra. If volcanic glass is dated, important and rich information will be provided for reconstructing Quaternary volcanic history. Therefore, in this doctoral thesis, I proposed new protocol for FT method using volcanic glass in order to provide reliable data easily, and also tried to develop new age determination method in order to offer additional date for cross-check.

First in chapter 2, I propose a new method to estimate the track number per unit volume and corresponding age equation. The number of tracks per unit volume must be constant provided that complete track fading does not occur. When the track number per unit volume, instead of the track number per unit area, is used in the age equation, there is no need for age correction. Obsidian shards with known induced FT density were etched in a stepwise manner and all induced FTs were observed through the etching of a significant glass volume. The induced FT number per unit volume is calculated from the number of observed tracks and the glass volume removed by the etching, which is estimated on the basis of a track etching model and measurement of track geometry, and then compared with the expected FT density. As a result, bulk etching rate and the track density are obtained, suggesting that this new dating protocol has the potential to provide reliable fission-track ages without nuclear irradiation and complicated age correction procedures. However this new FT counting strategy in volcanic glass results in a further increase in the number of observed screens. Therefore in the chapter 3, I construct an image processing protocol to reduce time-consuming labor works in counting tracks on many screens. I firstly obtained images of spontaneous tracks in obsidian shards under the microscope. Effects of image capturing conditions on the following image processing are examined. Images must be acquired under some conditions, and the focus must be on the surface within 1 micron above. Image processing detects both of tracks and other structures such as inclusions, dusts, etc. To distinguish ”true” tracks from other structures, circularity and the number of minimum points in the brightness profile along the long axis of ellipse are effective. This new counting protocol has the potential to report more precise fission-track ages without labor works because the image processing can count the number of tracks more than researchers can count. Finally in chapter 4, I also tried to develop a new age dating method in order to cross-check the ages by multiple methods on the same tephra. I observed zircon by the atomic force microscope (AFM) and examined surface regular patterns that were reported by Ohishi and Hasebe (2012). Considering that there might be a possibility that these patterns will be given by alpha recoil tracks, I examined AFM images for these patterns using the zircons from young volcanic rocks in Quaternary (recent rock and 0.32 ± 0.05 Ma) and old zircon (33.0 ± 1.0 Ma) after annealing treatments. The cyclic surface structures with amplitude of 1 - 2 nm was faintly found in the AFM images of young zircons. The small pits, which look like alpha recoil tracks (ARTs) with the depth of 4 - 6 nm, were also observed. Therefore the cyclic surface structures observed on old zircons (waveform with the amplitude of > 4 nm) may be the results of many ARTs formation. The cyclic surface structures are still observed in old zircon after annealing at high temperatures. Hence if these cyclic surface structures are formed by the ARTs, the ARTs might be able to be annealed at more than 1000°C. To apply ART dating, further understanding on how ART looks like on AFM or other nano-scale imaging must be necessary.

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学位論文審査報告書（甲）

1．学位論文題目（外国語の場合は和訳を付けること。）

Improvement and development of dating methods using radiation damage for reconstructing Quaternary volcanic history （第四紀火山活動史解明のための放射線損傷を利用した年代測定法の改良と開発）

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3．審査結果の要旨（600～650字）

本研究は環境研究，災害研究に重要な，第四紀の火山岩の年代決定をめざし，測定法の改良・開発を行ったものである。爆発的な火山噴火時に噴出する広域テフラ中には火山ガラスが普遍的かつ多量に含まれるため，有力な分析対象試料である。火山ガラスの適用できるフィッショントラック年代測定は若返った年代を示すことが多いため，補正のために原子炉での中性子照射が必要など，実験に手間がかかり最近は利用されることがなくなってきている。そのため，段階エッチングを施すことにより単位体積辺りのトラック密度を求める新しい補正方法を提案した。またその解析のために，画像処理を利用してプログラミングを行い，トラックをどのように認識させるかの議論を行い自動計数法を考案した。試料の写真撮影法や画像処理の手順の吟味も行った。また火山ガラスの年代測定結果と相互比較を行なうためにジルコン中のアルファリコイルトラック(ART)を利用した年代測定法の開発に着手した。古い試料と新しい試料，熱処理を施した試料の表面構造を原子間力顕微鏡で精査し，その違いについて報告した。これらの成果の一部は国際誌にて公表されて高く評価されており，本審査委員会は全会一致で学位の申請にふさわしい研究であると判断した。

4．審査結果　(1) 判　定（いずれかに○印）　合　格　・　不合格

(2) 授与学位　博　士（　理学　）