DISCUSSION ON TERMINOLOGY, ANISOTROPY, AND INTERPROCEDURAL CROSS-CHECKS OF FISSION TRACK AGES OF ZIRCON

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Abstract Technical terms should clearly be distinguished for procedural data collection and for statistical data calculation. Available procedural variations are defined and described. According to observation of anisotropic etching and counting especially among lower spontaneous track density zircons, explanatory terms are described; and the relationship between total (mainly on-anisotropic etching-dependent) counting efficiency (η_{EStotal}) and spontaneous track density (ρ_s) is deduced as $\eta_{\text{EStotal}} = 0.123 \log \rho_s + 0.220$. To counter contradictory opinions on possible procedural variations, 1-set-4-aliquot preparation and processing technique is introduced for interprocedural cross-checks of resulting ages, and preliminary results of its application to zircon from the Fish Canyon Tuff (K-Ar age, 27.9 ± 0.7 Ma, and F.T. age, 28.4 ± 0.7 Ma) are reported. Results of 2π Population, $S2\pi\text{ES}-I2\pi\text{ED}$, and $S4\pi\text{ES}-I2\pi\text{ED}$ (External Detector) procedures range from 32.4 to 26.6 Ma. After correcting their intrinsic procedural defects, ages become within the range of 28.2 $\sim$ 27.9 Ma. It is suggested that appropriate alternative(s) among possible procedural variations are selected for the particular geochronological background of zircon specimens, through extensive work on interprocedural cross-checks of resulting ages. In the case of younger or lower spontaneous track density zircon, advantages and dating limits ($\rho_s < 1 \times 10^6 \text{ t}\cdot\text{cm}^{-2}$) of the $S2\pi\text{ES}-I2\pi\text{ES}$ procedure, are especially referred and suggested.

Introduction

Recently in Japan, review papers (TAMANYU, 1981), critical comments on statistical treatment of fission track data (HAYASHI, 1981a, 1982), and proposals for "New Methods" (HAYASHI, 1981b; DANHARA *et al.*, 1982) have been published or orally presented. In most cases, there appeared a confused usage and misconception of technical terms, which resulted in misguided discussions. For example, contrary to its international tacit understanding (e.g. in Japan, SUZUKI, T., 1980), discussions on the Grain by Grain method in Japan are almost exclusively and peculiarly confined to a grain by grain age calculation, and consequently mislead to a

preference for the Population method (HAYASHI, 1981b). As it is an improved procedure to eliminate uncertainties (such as inter-crystal uranium content variation and possible contaminating detrital crystals) involved in the Population method, the Grain by Grain Processing method still retains its own validity. Even in the case of the popular "Subtraction" method (GLEADOW, 1981), details of the procedure become clear after checking its procedural flow diagram, because even those concerned in dating are, more or less, accustomed to a limited number of procedures among divergent, more than 10, possible variations, and a subtractive calculation is encountered in every dating procedure which involves counting induced tracks in non-annealed crystals. Thus, a strict definition of procedural terms is urgently needed for their unanimous understanding, and, of its nature, should be compiled and selec-

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tively recommended by some authorized commission in the near future. Though there may be a feed-back from a statistical data treatment to a procedural data collection (e.g. Fixed-area Population: HAYASHI, 1981b), in general a procedure never designates a particular data calculation, therefore the tentative definition of technical terms is attempted from the viewpoint of data collection procedures.

Concerning an anisotropy of spontaneous fission tracks in zircon, there is neither unanimity nor controversy, but a "black hole" exists. In this connection, the recent publication "A users' guide to fission track dating calibration" (HURFORD & GREEN, 1982), stated in its abstract "...and anisotropic etching (of induced tracks in annealed crystals) ... invalidate all but the population and external detector methods, ...". Such a confusing statement, only understood by inserting the phrase in parentheses, is criticized for missing or underestimating, among four possible kinds of tracks, spontaneous tracks, and induced tracks in an external detector, annealed and non-annealed crystals, the validity and advantages of the last.

Similar citation of reports, with supplementary citations (SUZUKI, T., 1980; GANZAWA, 1982, 1983; SUZUKI, M. *et al.*, in press) and practical data (described elsewhere) concerning an anisotropy of spontaneous tracks in zircon, may lead to a quite different statement. A premature standardization or recommendation for a certain procedure, without detailed and precise experiments on interprocedural cross-checks of resulting ages, and the mere compilation of complicated and frequently contradictory reports, which are mainly concerned with a limited part of a procedure, may misguide consumers of dating results, obstruct developments of new and/or improved procedures, and limit the scope of its application potentiality. It also becomes an urgent need to define explanatory terms for an anisotropic counting, and to develop experimental method for interprocedural cross-checks of resulting ages, preliminary results of which, using the Fish Canyon Tuff zircon, are described elsewhere.

The present paper is concerned with: (1) description of tentative definition of technical terms for data collection procedures and of explanatory terms for anisotropic etching, (2) introduction of 1-set-4-aliquot processing techniques oriented for interprocedural cross-checks of resulting ages (including improved technical/calculative versions), and (3) special reference to advantages of the S2 π ES-I2 π ES (external surface re-etch subtraction) procedure for dating younger or lower spontaneous track density zircon.

Experimental figures were obtained through usage of 8 g NaOH and 11.5 g KOH eutectic zircon etchant at 220°C or 240°C, experimental details of which are almost the same as described elsewhere (SUZUKI, M. *et al.*, in press).

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Recommended Definition of Technical Terms for Data Collection Procedures

The term Grain by Grain was originally biliteral for data collection and calculation (e.g. NAESER, 1969). After the introduction of improved etching techniques of FEP teflon mount and eutectic zircon etchant (GLEADOW *et al.*, 1976), emphasis gradually came to be laid on data collection procedures, the Grain by Grain Processing. Mainly thanks to these techniques, such divergent procedures as External Detector, Re-etch, Re-polish and so on, came into use for zircon dating. Contrary to dissenting opinions and complaints of its laboriousness as against the

Table 1. Correlation of Terminology for Data Collection Procedures

Present Description	Gleadow (1980a, 1981)	Others
S2 π ES-I2 π ED		EED (external surface-external detector: Hayashi, 1982)
S2 π ES-I2 π ES		EID (external surface-internal detector: Hayashi, 1982, citing Nishimura and Ikeda, 1973)
		GBG (biliteral: e.g. Yokoyama et al., 1982)
		GGP (biliteral: Danhara et al., 1982)
S4 π IS-I2 π ED	External Detector	IED (internal surface-external detector: Hayashi, 1982)
S4 π IS-I2 π IS	Re-etch	IID (internal surface-internal detector: Hayashi, 1982)
4 π Population	Subtraction	
4 π Population-A	Population	Fixed-area Population (special case: Hayashi, 1981)
S4 π IS-I4 π IS	Re-polish	
S4 π IS-A-I4 π IS	Re-polish	
(Technical/Calculative Version)		(Derivative Calculative Version)
S4 π IS-I4 π IS conversion		S2 π ES-I2 π ED η_{Estotal} corrected
		2 π Population Cw corrected
		S2 π ES-I2 π ES conversion
		S4 π IS-I2 π ED η_{IStotal} corrected
		4 π Population Cw corrected

Grain by Grain Processing (HAYASHI, 1981a, b, 1982; except for his acceptable claims and proposals for statistical data analyses and test), even in the Population method, the Grain by Grain data collection should be used. And it is possible, with the use of an external detector, to eliminate uncertainties derived from wide inter-crystal uranium content variation and possible contaminating detrital grains involved in the Population method, because the external detector is an alternative to all procedures waiting for its turn.

The following description of terms, mainly from the viewpoint of data collection procedures, is based on the present opinion that technical terms should clearly be distinguished

for procedural data collection and statistical data calculation with the one exception of the biliteral term "Population".

Population: biliteral for procedure and calculation.

S and I: for spontaneous and for induced track number or density data collection.

2 π and 4 π : 2 π and 4 π geometry.

ES, IS, and ED: external and internal surfaces of crystals, and external detectors.

-A: showing an annealing step applied to an aliquot for induced track number or density data collection before irradiation.

Data collection procedures are expressed in terms of a combination of S (geometry) (counting surface) and I (geometry) (counting surface/detector), for example S4 π IS-

I2 π ED (External Detector: GLEADOW, 1980a, 1981, and S4 π IS-A-I4 π IS (Re-polish: GLEADOW, 1980a, 1981). Other (some partially calculative) terms such as Re-etch, Re-polish, Subtraction are implicitly expressed. A correlation of terms is listed in Table 1.

Anisotropy, and Its Explanatory Terms

The first approach to successive etching and observation of spontaneous or induced tracks in crystals, apart from a count of tracks itself, demonstrates an anisotropic etching of zircon (e.g. GLEADOW, 1980b). Removal of the surface (ΔR) by etching, even slight as it is ($\Delta R < 0.4 \mu\text{m}$: SHIRAO, M. and SUZUKI, M., unpublished data), makes an anisotropic etching into a continuous existence, dissenting from the opinion reported by using collimated ^{252}Cf fission fragments from outside the zircon surface (GLEADOW, 1980b). The problems involved are how to discriminate those thin tracks from dislocations similarly running straight, approximately parallel to the c -axis, as in due course both are anisotropically etched thin, and how to count those dot-like shallow tracks of near $0 \mu\text{m}$ in length (in the case of S4 π IS). It seems that different criteria exist for identification of these tracks, mainly due to different circumstances surrounding dating specialists. Those accustomed to dating older zircon using the S4 π IS-I2 π ED procedure, are liable to and indeed must count such tracks, while those, asked to date younger zircon, are hesitant and reluctant to count them. It might be possible to count tracks isotropically even in the case of lower spontaneous track density zircon, as successful reports (GLEADOW, 1980b; OBRADOVICH *et al.*, 1982) suggest, but the present author, as one accustomed to dating younger zircon, assents to opinions for an anisotropic counting of spontaneous tracks (SUZUKI, T., 1980; GANZAWA, 1982, 1983; SUZUKI, M. *et al.*, in press). Given one such track in 10^{-4}cm^2 , one miscount brings a considerable change of density for $\rho_s \sim 10^5 \text{t}\cdot\text{cm}^{-2}$, but a slight and negligible change for $\rho_s \sim 10^6 \text{t}\cdot\text{cm}^{-2}$. Thus, counting lower density spontaneous tracks is

relative, which can be compensated for directly by counting induced tracks in non-annealed zircon at a similar anisotropic counting, or indirectly by correcting the possible absolute count of induced tracks in the external detector with the use of a total counting efficiency.

On the premise of an anisotropic counting, the following explanatory terms are defined and described.

η_{ED} : counting efficiency of mica external detector (=1.00: Datum Point).

η_{A} : on-anisotropic etching-dependent counting efficiency, common to ES and IS.

For practice, η_{A15} is used (see Table 2).

η_{ES} and η_{IS} : on-surface removal-dependent counting efficiency for ES and IS.

η_{EStotal} and η_{IStotal} : total counting efficiency for ES ($\Sigma N_i / \Sigma N_{\text{ED}} = \eta_{\text{ES}} \cdot \eta_{\text{A15}}$) and IS ($\Sigma N_i / \Sigma (2N_{\text{ED}}) = \eta_{\text{IS}} \cdot \eta_{\text{A15}}$).

χ^2_{A} : the exclusive measure for bridging on-anisotropic etching-dependent counting efficiencies of both spontaneous and induced tracks in non-annealed crystals (SUZUKI, M. *et al.*, in press; see Table 2).

Results of counting efficiencies obtained mainly from lower spontaneous track density zircon samples are shown in Table 2 and Figure 1. From these results, the following facts emerge (1) there is no clear difference of on-anisotropic etching-dependent counting efficiencies between different geometries, and between spontaneous and induced tracks (mixed with 22.7% spontaneous tracks) in non-annealed Fish Canyon Tuff zircons, and all of them show a highly anisotropic distribution, and (2) there is a highly significant correlation between a total counting efficiency and spontaneous track density for ES, while no correlation is observed between a total counting efficiency and induced track density.

Thus, a total counting efficiency of induced tracks can directly be determined by $\Sigma N_i / \Sigma N_{\text{ED}}$, while that of spontaneous tracks is indirectly assured to be the same as that of induced tracks through χ^2_{A} test. It is important to point out that η_{EStotal} or η_{IStotal} for both spontaneous and induced tracks is exclusively dependent on ρ_s and remains independent from ρ_i until on irradiation.

Table 2. Results of Counting Efficiencies (with an explanatory figure)

for lower spontaneous track density zircon						Fish Canyon Tuff Zircon (No. 96)				
η_{Estotal}	$\Sigma N_s(t)$	$\Sigma N_z(t)$	$\rho_s(t \cdot \text{cm}^{-2})$	η_{Estotal}	$\rho_z(t \cdot \text{cm}^{-2})$	Datum α -axis	$(N_s' + N_z')_{2\pi}^{**}$	$N_{s2\pi}$	$N_{s4\pi}$	N (total)
1*	963.4	(3285.6)	(2.56×10^6)	(1.05)	(8.73×10^6)	0°-10°	603	601	449	(1653)
2	156	2361	6.34×10^4	0.84	9.60×10^5	10°-20°	621	598	402	(1621)
3	389	4432	1.66×10^5	0.83	1.89×10^6	20°-30°	604	629	428	(1661)
4	199	2280	1.08×10^5	0.82	1.24×10^6	30°-40°	637	618	428	(1683)
5	118	1310	7.97×10^4	0.82	8.85×10^5	40°-50°	642	626	398	(1666)
6	136	1266	5.11×10^4	0.82	5.78×10^5	50°-60°	576	595	411	(1582)
7	49	510	1.03×10^5	0.80	1.07×10^6	60°-70°	519	553	394	(1466)
8	183	2599	9.96×10^4	0.80	1.41×10^6	70°-80°	467	494	326	(1287)
9	108	2169	4.64×10^4	0.79	9.33×10^5	80°-90°	411	438	271	(1120)
10	73	1337	3.73×10^4	0.79	6.84×10^5	(total)	5080	5152	3507	(13739)
11	56	4094	2.18×10^4	0.78	1.59×10^6	χ^2_A ***	3.810	2.135	5.831	Datum
12	189	11119	2.47×10^4	0.77	1.45×10^6	χ^2_R ****	93.49	60.66	64.90	207.21
13	122	2866	3.72×10^4	0.76	8.75×10^5	η_{Al5} *****	0.93	0.94	0.91	0.93
14	116	7738	1.77×10^4	0.75	1.18×10^6					
15	88	6031	1.79×10^4	0.74	1.23×10^6					
16	35	1403	1.19×10^4	0.73	4.77×10^5					
17	116	3347	9.04×10^3	0.72	2.61×10^5					

* Data obtained by conversion calculation of Fish Canyon Tuff zircon; and excluded from the correlation coefficient calculation:

** $\eta_{\text{Estotal}} = 0.123 \log \rho_s + 0.220$ ($r = 0.889 > r(14, 0.01) = 0.6226$)

Mixing ratio of N_s' to N_z' is 1 to 3.41 (77.3% induced tracks).

*** $\chi^2_A = \sum_{j=1}^9 \{ \{ N_{1(10j-5)} - N_{2(10j-5)} \} \cdot \Sigma N_{1'} \cdot (\Sigma N_{2'})^{-1} \}^2 / \{ N_{2(10j-5)} \} \cdot \Sigma N_{1'} \cdot (\Sigma N_{2'})^{-1} \}$

**** $\chi^2_R = \sum_{j=1}^9 \{ \{ N_{1(10j-5)} - \Sigma N_{1'}/9 \}^2 / (\Sigma N_{1'}/9) \}$ (χ^2 test for a random distribution)

***** $\eta_{\text{Al5}} = \Sigma N \cdot \{ 3 \cdot \sum_{j=1}^3 N_{1(10j-5)} \}^{-1}$

tion-dependent change of η_A appears for induced track counting ($\rho_i > 1.57 \times 10^4 \rho_s^{0.47}$; see Figure 1; also see SUZUKI, M. *et al.*, in press, mainly based on results obtained

from experiments on annealed zircon by SUZUKI, T., 1980), thus determines and indicates the degree of metamictization of crystal.

Counting Efficiency

Figure 1. Density dependence of track counting efficiency

Open circle : spontaneous track

Solid circle : induced track in non-annealed zircon

Line : induced track in annealed zircon (see SUZUKI, T., 1980, and SUZUKI, M. *et al.*, in press)

Interprocedural Cross-checks of Resulting Ages

(a) 1-set-4-aliquot Preparation and Processing

The following considerations are exclusively concerned with the handling procedures of zircon dating. Methods in parentheses designate their classification by GLEADOW (1980a, 1981).

Observation of wide inter-crystal uranium content variations in zircon: invalidates 2π Population, 4π Population (Subtraction), and 4π Population-A (Population).

Preference for similar geometry factors for both spontaneous and induced tracks (see NAESER & DODGE, 1969; REIMER *et al.*, 1970; GLEADOW & LOVERING, 1977; GREEN & DURRANI, 1978): invalidates $S4\pi IS-I2\pi IS$ (Re-etch), $S2\pi ES-I4\pi IS$, and $S4\pi IS-I2\pi ED$ (External Detector), ($S4\pi IS-I2\pi ES$ is practically not available).

Observation of different counting efficiencies between zircon surface and external detector (NISHIMURA & IKEDA, 1973): invalidates $S2\pi ES-I2\pi ED$, and $S4\pi IS-I2\pi ED$ (External Detector).

Restoration of original anisotropic characteristics of annealed crystals (SUZUKI, T., 1980; for sphene, see GLEADOW, 1978, 1981): invalidates 2π Population-A, 4π Population-A (Population), and $S4\pi IS-A-I4\pi IS$ (included in Re-polish).

Observation of anisotropic etching of spontaneous tracks in zircon (SUZUKI, T., 1980; GANZAWA, 1982; SUZUKI, M. *et al.*, in press): invalidates $S2\pi ES-I2\pi ED$, and $S4\pi IS-I2\pi ED$ (External Detector).

Probable contamination of external surface of zircon (see HURFORD & GREEN, 1982, notes of Fig. 1; REIMER *et al.*, 1970): invalidates $S2\pi ES-I2\pi ED$, and $S2\pi ES-I2\pi ES$.

Thus, all five alternatives classified by GLEADOW (1980a, 1981) involve to some extent disadvantages against the ideal data collection, and contrary to the dissenting review (HURFORD & GREEN, 1982), even their preferred procedures, the Population (4π Population-A) and the External Detector ($S4\pi IS-I2\pi ED$) methods are never free from their "bonds" of controversy.

Among these procedures, the following are recovered for zircon dating.

$S4\pi$ IS- $I2\pi$ ED (External Detector)*: for its wide applicability for ages and the uranium content of zircon, in conditions where there is no anisotropic etching of spontaneous tracks observed; its improved calculative version is $S4\pi$ IS- $I2\pi$ ED η_{IStotal} corrected* (for Miocene zircon, on-anisotropic etching-dependent counting efficiency ranges from 0.68 to 0.95, and is also dependent on spontaneous track density, GANZAWA, 1982, 1983).

4π Population (Subtraction): on condition that a correction for differences of uranium content between 2 aliquots is applied (4π Population c_w corrected*); another improved technical/calculative version, $S4\pi$ IS- $I4\pi$ IS conversion* (its age calculation equation described elsewhere), is possible through grain by grain data collection with the use of an external detector.

$S2\pi$ ES- $I2\pi$ ED η_{EStotal} corrected* (improved calculative version), $S2\pi$ ES- $I2\pi$ ES*, 2π Population c_w corrected* (improved calculative version), and $S2\pi$ ES- $I2\pi$ ES conversion* (improved calculative version): for further cautious investigation of tracks is merited (GLEADOW, 1980a), especially for dating lower spontaneous track density zircon (SUZUKI, M. *et al.*, in press).

For the purpose of interprocedural cross-checks of resulting ages, only 4 aliquots are required, 2 for 2π , and 2 for 4π geometry. The 25 mm diameter of an irradiation capsule (or tube) for Rotary Specimen Rack of the TRIGA MARK II nuclear reactor, means it is possible to plant zircon crystals in FEP teflon mount in 16×16 matrix, 1 mm interval with 2 mm space for its cutting apart into 4 aliquots, each of which contains up to 64 grains. Gauging wire for polishing out 4π geometry internal surface of crystal, should be mounted with 2 aliquots for 4π geometry. If required to mount more than 64 grains in each aliquot for dating lower spontaneous track density zircon, 2-set-4-aliquot should be prepared. Processing steps and data collection are listed in Table 3, equations for age calculation using procedure with an asterisk (*) mentioned above (including technical/calculative and calculative ver-

sions) are described. These equations, intentionally oriented for younger zircon dating, are described on the premises that, (1) grain by grain data collection of more than 30 grains, even in the case of the Population procedure, and (2) total grain age calculation, are used, and that (3) poisson distribution of both spontaneous and induced track numbers for error calculation (2σ) is assumed (for further detailed discussion, see HAYASHI, 1981a, b, 1982).

(b) Preliminary Results on Zircon from the Fish Canyon Tuff

Results of an interlaboratory comparison of fission track ages of zircon separates from the Fish Canyon Tuff (K-Ar age, 27.9 ± 0.7 Ma) were reported as 28.4 ± 0.7 (2σ) Ma using $\lambda_f \sim 7.0 \times 10^{-17} \text{a}^{-1}$ and the $S4\pi$ IS- $I2\pi$ ED (External Detector) procedure (NAESER *et al.*, 1981).

Preliminary interprocedural cross-checks of resulting ages were achieved on this age standard (without aliquot D in Table 3), and results are shown in Table 4. If errors are taken into consideration, all the resulting ages are consistent with each other except for the 2π Population age. However, precise checks reveal intrinsic defects involved in each procedure. The 2π Population resulted in an older age (32.4 Ma), due to uranium content differences between aliquot A (210 ppm) and aliquot B (180 ppm). The $S2\pi$ ES- $I2\pi$ ED, due to over-balancing ($\eta_{\text{EStotal}} = 1.05$) of on-surface removal-dependent increase in counting ($\eta_{\text{ES}} = 1.13$) against on-anisotropic etching-dependent decrease in counting ($\eta_{\text{AIS}} = 0.93$), gave an older age (29.3 Ma). And the $S4\pi$ IS- $I2\pi$ ED, due to its insufficient offset ($\eta_{\text{IStotal}} \approx 0.95$), resulted in a younger age (26.7 Ma). A calculated η_{EStotal} (1.02) using the equation for η_{EStotal} and ρ_s , is slightly lower than the direct (partially calculative in this case) value (1.05), due to uncertainties involved in this equation. In due course its application for correcting the $S2\pi$ ES- $I2\pi$ ED age resulted in a slightly older age (28.9 Ma). Another possible procedure, the $S2\pi$ ES- $I2\pi$ ES, was cancelled in the present case, because the rough surface and high spontaneous track density ($\rho_{\text{SA}} =$

Table 3. 1-set-4-aliquot Processing and Data Collection

Step	Aliquot A	Aliquot B	Aliquot C	Aliquot D
Irradiation 0				ρ_{D0}
Polish			(4 π Geometry)	(4 π Geometry)
Etch & Count I	ρ_{sA}		ρ_{sC}	$(\rho_s + \rho_i)_D$
Irradiation	ρ_D	ρ_D	ρ_D	ρ_D
Etch & Count II	ρ_{EDA} ρ_{iA}	ρ_{EDB} $(\rho_s + \rho_i)_B$	ρ_{EDC}	ρ_{EDD}

All the densities (ρ) except for ρ_{D0} and ρ_D can be replaced by the number of tracks (N).

S2 π ES-I2 π ED $\eta_{E\text{total}}$ corrected

$$A = \zeta(\rho_{sA} \cdot \rho_{EDA}^{-1} \cdot \eta_{E\text{total}}^{-1}) \rho_D \{1 \pm 2((\Sigma N_{sA})^{-1} + (\Sigma N_{EDA})^{-1} + N_D^{-1})^{0.5}\}$$

S2 π ES-I2 π ES

$$A = \zeta(\rho_{sA} \cdot \rho_{iA}^{-1}) \rho_D \{1 \pm 2((\Sigma N_{sA})^{-1} + (\Sigma N_{iA})^{-1} + N_D^{-1})^{0.5}\}$$

2 π Population Cw corrected

$$A = \zeta\{\rho_{sA} \cdot \rho_{EDB} \cdot ((\rho_s + \rho_i)_B \cdot \rho_{EDA} - \rho_{sA} \cdot \rho_{EDB})^{-1}\} \rho_D \\ \times \{1 \pm 2((\Sigma N_{sA})^{-1} + (\Sigma(N_s + N_i)_B)^{-1} + (\Sigma N_{EDA})^{-1} + (\Sigma N_{EDB})^{-1} + N_D^{-1})^{0.5}\}$$

S2 π ES-I2 π ES conversion

$$A = \zeta((\Sigma N_{sB}) \cdot (\Sigma N_{iB})^{-1}) \rho_D \\ \times \{1 \pm 2((\Sigma N_{sA})^{-1} + (\Sigma N_{sB})^{-1} + (\Sigma N_{iB})^{-1} + (\Sigma N_{EDA})^{-1} + (\Sigma N_{EDB})^{-1} + N_D^{-1})^{0.5}\}$$

(a and b for N_{sB} calculation are determined from the regression equation of

$$N_{sA} = aN_{EDA} + b)$$

S4 π IS-I2 π ED

$$A = \zeta(\rho_{sC} \cdot (2\rho_{EDC})^{-1}) \rho_D \{1 \pm 2((\Sigma N_{sC})^{-1} + (\Sigma N_{EDC})^{-1} + N_D^{-1})^{0.5}\}$$

S4 π IS-I2 π ED $\eta_{I\text{total}}$ corrected

$$A = \zeta(\rho_{sC} \cdot (2\rho_{EDC} \cdot \eta_{I\text{total}})^{-1}) \rho_D \{1 \pm 2((\Sigma N_{sC})^{-1} + (\Sigma N_{EDC})^{-1} + N_D^{-1})^{0.5}\}$$

4 π Population Cw corrected

$$A = \zeta\{\rho_{sC} \cdot \rho_{EDD} \cdot ((\rho_s + \rho_i)_D \cdot \rho_{EDC} - \rho_{sC} \cdot \rho_{EDD})^{-1}\} \rho_{D0} \\ \times \{1 \pm 2((\Sigma N_{sC})^{-1} + (\Sigma(N_s + N_i)_D)^{-1} + (\Sigma N_{EDC})^{-1} + (\Sigma N_{EDD})^{-1} + N_{D0}^{-1})^{0.5}\}$$

S4 π IS-I4 π IS conversion

$$A = \zeta((\Sigma N_{sD}) \cdot (\Sigma N_{iD})^{-1}) \rho_{D0} \\ \times \{1 \pm 2((\Sigma N_{sC})^{-1} + (\Sigma N_{sD})^{-1} + (\Sigma N_{iD})^{-1} + (\Sigma N_{EDC})^{-1} + (\Sigma N_{EDD})^{-1} + N_{D0}^{-1})^{0.5}\}$$

(c and d for N_{sD} calculation are determined from the regression equation of

$$N_{sC} = cN_{EDC} + d)$$

$2.95 \times 10^6 \text{ t} \cdot \text{cm}^{-2}$) after the Etch & Count I step severely disturbed the Etch & Count II step (see Table 3). However, its alternative, S2 π ES-I2 π ES conversion, gave a consistent

age result (28.2 Ma). After correcting their intrinsic defects, three ages come within the range of 28.2~27.9 Ma, (the S4 π IS-I2 π ED $\eta_{I\text{total}}$ corrected age (27.9 Ma) has no

Table 4. A Case Study on Interprocedural Cross-checks of Resulting Ages

Thermal Neutron Dosimetry*: Nuclear Reactor: Lazy Susan (United States Geological Survey), 200 kw - 1h.						
N_D	ρ_D	ϕ	λ_f	I	B_{C-2**}	σ
t	$t \cdot \text{cm}^{-2}$	$n \cdot \text{cm}^{-2}$	a^{-1}		$t \cdot n^{-1}$	$a \cdot \text{cm}^2 \cdot t^{-1}$
19311	7.01×10^5	1.60×10^{15}	$(7.03 \pm 0.11) \times 10^{-17}$	137.88	4.38×10^{-10}	1.37×10^2

* Cross-checked using SRM 962, Corning Glass 1 and 2 (see Suzuki, M. et al., in press)

** Corning Glass 2

Track Density

Aliquot	N_G	ρ_s	(ΣN_s)	ρ_i	(ΣN_i)	ρ_{ED}	(ΣN_{ED})
(Geometry)		$t \cdot \text{cm}^{-2}$	(t)	$t \cdot \text{cm}^{-2}$	(t)	$t \cdot \text{cm}^{-2}$	(t)
A(2 π)	37	2.95×10^6	(2742)			9.68×10^6	(8981)
B(2 π)	15	2.56×10^6	(963.4)	8.73×10^6	(3285.6)	8.32×10^6	(3131)
C(4 π)	89	5.18×10^6	(2313)			9.33×10^6	(4166)

Ages

Procedure	Age(Ma) $\pm 2\sigma$
2 π Population	32.4 ± 1.74
S2 π ES-I2 π ED	29.3 ± 1.34
S2 π ES-I2 π ED η_{Estotal} corrected (Calculative)	28.9 ± 1.34
S2 π ES-I2 π ES conversion: $N_{sA} = 0.294N_{\text{EDA}} + 2.779$ ($r = 0.970$)	28.2 ± 2.58
S2 π ES-I2 π ED η_{Estotal} corrected (Direct/Calculative)	27.9 ± 1.89
S4 π IS-I2 π ED η_{Estotal} corrected (Calculative)	27.9 ± 2.79
2 π Population C_w corrected	27.9 ± 1.83
S4 π IS-I2 π ED (External Detector)	26.7 ± 1.43

Counting Efficiencies

$\eta_{\text{Estotal}} = 0.123 \log \rho_s + 0.220 = 1.02$ (Calculative)	$\eta_{\text{EDpolycarbonate}} = 1.004 \pm 0.010$
$\eta_{\text{Estotal}} = (\Sigma N_{iB}) / (\Sigma N_{\text{EDB}}) = 1.05$ (Direct/Calculative)	$\eta_{\text{Istotal}} = \{ \Sigma N_{sC} \cdot \Sigma N_{\text{EDA}} \cdot (\Sigma N_{iB}) \} / (2 \Sigma N_{\text{EDC}} \cdot \Sigma N_{sA} \cdot \Sigma N_{\text{EDB}}) = 0.95$
$\eta_{\text{ES}} = \eta_{\text{Estotal}} / \eta_{\text{A15}} = 1.05 / 0.93 = 1.13 \pm 0.03$	$\eta_{\text{IS}} = \eta_{\text{Istotal}} / \eta_{\text{A15}} = 0.95 / 0.93 = 1.03 \pm 0.05$

validity, due to its circular reasoning, see $\eta_{\text{IS total}}$ calculation in Table 4). Concerning anisotropic counting (see Table 2), there may arise some criticism for its under-etching, therefore it would be better to point out in advance that ρ_{sc} ($=5.18 \times 10^6 \text{ t} \cdot \text{cm}^{-2}$; $N_{\text{G}}=89$) is well within the range ($\rho_{\text{S4}\pi}$: $2.60 \sim 4.99$ (mean) $\sim 6.36 \times 10^6 \text{ t} \cdot \text{cm}^{-2}$) of results of an interlaboratory comparison (NAESER *et al.*, 1981).

From these results, preliminary comments are made for procedural aspects of the fission track dating method: (1) there is no cure-all procedure applicable to the whole range of ρ_{s} (or AC in FLEISCHER *et al.*, 1975, p. 171–172), because (i) if the relationship between on-anisotropic etching-dependent counting efficiency particularly and spontaneous track density (ρ_{s}) is taken for granted, the age gap between the “true” age and the age obtained by the procedure which involves the use of an external detector becomes wider as ρ_{s} decreases, and (ii) the S2 π ES–I2 π ES procedure could not be applied to the present case; (2) the effect on age of so called “contamination” of an external surface (HURFORD & GREEN, 1982) was not observed (see also SUZUKI, M. *et al.*, in press); and (3) the proposed “age standard” method for the sake of by-passing the controversy on decay constant, and/or neutron dosimetry, requires caution, because intrinsic procedural defects do not always affect to a similar degree the ages of unknown-age and age-standard samples. Therefore a suitable correction should be applied to each particular procedure. Thus through extensive work on interprocedural cross-checks of resulting ages, an appropriate alternative among possible procedural variations can be selected for a particular geochronological background of zircon samples.

Comparison of S2 π ES–I2 π ES·ED and S4 π IS–I2 π ED for Younger Zircon Dating

As already discussed, various problems such as geometry, counting efficiency, “contamination” of the external surface of crystals and so on, are involved in possible procedural

variations. Concerning dating younger or lower spontaneous track density zircon, the highest priority should be given to the positive identification of tracks. It is suggested that the observation of tracks on an external surface of crystals may merit further cautious investigation in the case of a considerably higher uranium content material such as zircon, because effects derived from “contamination” become negligible (GLEADOW, 1980a; SUZUKI, M. *et al.*, in press). Careful identification of tracks requires up-and-down focusing to check the running directions of tracks, resulting in a preference for a longer (not projected) track length of the S2 π ES subprocedure. A comparison between the S2 π ES–I2 π ES·ED and S4 π IS–I2 π ED procedures is listed in Table 5. The present choice is the S2 π ES–I2 π ES·ED procedure, for its cautious identification, double checks and compensation capability of relative counts, of both spontaneous and induced tracks.

In this connection, dating limits of the S2 π ES–I2 π ES procedure should be described. A slight removal of surface (ΔR) occurs during etching ($\Delta R < 0.4 \mu\text{m}$: SHIRAO, M. & SUZUKI, M., unpublished data), and increment ratio (ΔN_{s}) of N_{s} at the Etch & Count II step is given by $\Delta N_{\text{s}} = \int_0^{2\Delta R} (R/2 - x) dx / \left(\int_0^{R/2} x \cdot dx + \int_0^{\Delta R} (R/2 - x) dx \right)$. The production ($N_{\text{s}} \cdot \Delta N_{\text{s}}$) of N_{s} and ΔN_{s} is included in an induced track number (N_{i}') at the Etch & Count II step, therefore the ratio of the age (A') obtained using N_{i}' to the “true” age (A) is given by $A'/A = (N_{\text{i}}' - N_{\text{s}} \cdot \Delta N_{\text{s}}) / N_{\text{i}}'$. Given ΔR equal $0.4 \mu\text{m}$, then the age ratio becomes 0.99 in the case where N_{s} to N_{i} is 1 to 10. Therefore, the ratio of N_{s} to N_{i} is preferably less than 1 to 10, and practically 1 to 3 ($A'/A = 0.96$) at the most. Another factor, the observational limit of the total track density ($\rho \sim 5 \times 10^6 \text{ t} \cdot \text{cm}^{-2}$ for 2 π geometry re-etch subtraction), puts the practical limit for the spontaneous track density at $\rho_{\text{s}} \sim 1 \times 10^6 \text{ t} \cdot \text{cm}^{-2}$, (at this density, the advantages of S2 π ES–I2 π ES are already negated). A case study of the application

Table 5. Comparison of S2 π ES-I2 π ES•ED and S4 π IS-I2 π ED

	S2 π ES-I2 π ES•ED	S4 π IS-I2 π ED
Polish	No need.	Need to polish at least 5.7 μ m off.
Etching	Longer etching durable for lower spontaneous track density zircon.	Crystals easily fall down from FEP teflon (this disadvantage may be recovered using gauging wire).
Actual Length of Spontaneous Tracks	$R/2 \sim R$ ($= 11.4 \mu$ m).	$0 \sim R$; even if optimally etched, shorter tracks ($\sim 2 \mu$ m) are difficult to identify with certainty on a rough surface after longer etching for lower spontaneous track density zircon.
ρ_s	Double checks at Etch & Count I and II.	Once at Etch & Count I ($\rho_{s4\pi} \approx 2 \times \rho_{s2\pi}$).
ρ_i	Double checks at Etch & Count II in crystal surface and external detector ($\eta_{Estotal} = 0.123 \log \rho_s + 0.220$).	Once at Etch & Count II (for ρ_{ED}), but absolute determination (only) is possible.
η_A	Possible to check through χ^2_A .	No way to check an anisotropic counting efficiency of induced tracks in internal surface through external detector counting.
EE (External Effect)	Might be probable in the case of zircon, generally negligible: Suzuki <i>et al.</i> , 1983).	No.
Others		Possible to keep processed specimens.

of the S2 π ES-I2 π ES procedure to a higher spontaneous track density specimen, contrary to the theoretical expectation of its younger age, resulted in an older age, mainly due to the difficulties of optimal etching (etching was suspended at the underetching condition) at the Etch & Count II step. On the other hand, on-irradiation-dependent change of counting efficiency appears as induced track density (ρ_i) approaches to the ρ_s -dependent density threshold given by $\rho_i = 1.57 \times 10^4 \cdot \rho_s^{0.47}$, therefore the suitable range of ρ_i for the typical spontaneous track density of $3 \times 10^4 \text{ t} \cdot \text{cm}^{-2}$ for the Quaternary zircon specimens, is 1×10^5 to $2 \times 10^6 \text{ t} \cdot \text{cm}^{-2}$.

References

- DANHARA, T., NISHIMURA, S., and TOKUHASHI, S., 1982 : On the new "GGP method" of fission-track age dating with special reference to some late Quaternary volcanic materials with poor uranium content (abstract). *Proceedings of Workshop on Fission-Track Dating*, Nikko, Japan, June 29, 1982, 10-12.
- FLEISCHER, R. L., PRICE, P. B., and WALKER, R. M., 1975 : *Nuclear Tracks in Solids: Principles and Applications*. 605 p. University of California Press, Berkeley.
- GANZAWA, Y., 1982 : Fission track dating of Tertiary volcanic rocks in northern Japan (abstract). *Proceedings of Workshop on Fission-Track Dating*, Nikko, Japan, June 29, 1982, 17-18.
- GANZAWA, Y., 1983 : Geometry factor in fission track dating. *Jour. Fac. Sci., Hokkaido Univ.*, Ser. IV, **20**, 195-292.
- GLEADOW, A. J. W., 1978 : Anisotropic and variable track etching characteristics in natural sphene. *Nuclear Track Detection*, **2**, 105-117.
- GLEADOW, A. J. W., 1980a : Fission track dating: what are the real alternatives? (abstract).

- Proceedings of Fission Track Dating Workshop*, Pisa, Italy, September 1980.
- GLEADOW, A. J. W., 1980b : Fission track age of the KBS tuff and associated hominid remains in northern Kenya. *Nature*, **284**, 225—230.
- GLEADOW, A. J. W., 1981 : Fission track dating : what are the real alternatives ? *Nuclear Tracks*, **5**, 3—14.
- GLEADOW, A. J. W., HURFORD, A. J., and QUAIFFE, R. D., 1976 : Fission track dating of zircon : improved etching techniques. *Earth and Planetary Science Letters*, **33**, 273—276.
- GLEADOW, A. J. W. and LOVERING, J. F., 1977 : Geometry factors in fission track dating. *Nuclear Track Detection*, **1**, 99—106.
- GREEN, P. F. and DURRANI, S. A., 1978 : A quantitative assessment of geometry factors for use in fission track studies. *Nuclear Tracks*, **2**, 207—213.
- HAYASHI, M., 1981a : Problems of the grain by grain method in fission track dating.** *Jour. Japan. Assoc. Min. Pet. Econ. Geol.*, **76**, 233—238.
- HAYASHI, M., 1981b : Fission track dating by a newly developed "Fixed-area population method".** *Jour. Geol. Soc. Japan*, **87**, 307—313.
- HAYASHI, M., 1982 : Statistical tests of the fission track dating data by the grain by grain method.** *Jour. Geol. Soc. Japan*, **88**, 691—697.
- HURFORD, A. J. and GREEN, P. F., 1982 : A users' guide to fission track dating calibration. *Earth and Planetary Science Letters*, **59**, 343—354.
- NAESER, C. W., 1969 : Etching tracks in zircon. *Science*, **165**, 388.
- NAESER, C. W. and DODGE, F. C. W., 1969 : Fission-track ages of accessory minerals from granitic rocks of the Sierra Nevada Batholith, California. *Bulletin of Geological Society of America*, **80**, 2201—2212.
- NAESER, C. W., ZIMMERMANN, R. A., and CEBULA, G. T., 1981 : Fission track dating of apatite and zircon : an interlaboratory comparison. *Nuclear Tracks*, **5**, 65—72.
- NISHIMURA, S. and IKEDA, T., 1973 : Application of the fission track dating method to geosciences.* *Tsukumo Chigaku*, **8**, 1—14.
- OBRADOVICH, J. D., NAESER, C. W., IZETT, G. A., PASINI, G., and BIGGAZZI, G., 1982 : Age constraints on the proposed Plio-Pleistocene boundary stratotype at Vrica, Italy. *Nature*, **298**, 55—59.
- REIMER, G. M., STORZER, D., and WAGNER, G. A., 1970 : Geometry factors in fission track counting. *Earth and Planetary Science Letters*, **9**, 401—404.
- SUZUKI, M., WIKARNO, BUDISANTOSO, SAEFUDIN, I. and ITIHARA, M. : Fission track ages of pumice tuff, tuff layers, and javites of hominid fossil-bearing formations in Sangiran, Central Java. (in press) *Special Publication of the Geological Research and Development Centre, Bandung, Indonesia*.
- SUZUKI, T., 1980 : Fission track ages of the Tertiary volcanic rocks in the Oga peninsula, northern Japan.** *Jour. Geol. Soc. Japan*, **86**, 441—451.
- TAMANYU, S., 1981 : Fission track dating method—its principles and laboratory procedures—.** *J. Geogr.*, **90**, 22—33.
- YOKOYAMA, T., MATSUDA T., and NISHIMURA, S., 1982 : Some special problems about "GBG method" of fission-track age dating on the volcanic materials (abstract). *Proceedings of Workshop on Fission-Track Dating*, Nikko, June 29, 1982, 47—48.
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* in Japanese (tentative translation by the present author).

** in Japanese with English abstract.

ジルコン結晶のフィッショントラック年代測定における 用語法, 異方性, 及び手法間比較について

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(要 旨)

フィッショントラック年代測定法の手法を表示する用語は, 実験的側面とデータ処理の側面とを明確に区別して, 使用すべきである. 種々の可能な方法を, 実験的側面から定義し記載した. 特に自発トラック密度の低いジルコンに観察される異方性エッチングと計数を表現する用語を記載した. 外部表面の総計数効率(η_{EStotal})と自発トラック密度(ρ_s)との間には, $\eta_{\text{EStotal}} = 0.123 \log \rho_s + 0.220$ で示される関係がある. それぞれの手法の評価をめぐる対立する見解を検討するために, 1-set-4-aliquot 試料処理法を紹介した. 標準年代試料として一般に利用される Fish Canyon Tuff (K-Ar 年代 27.9

± 0.7 Ma, フィッショントラック年代 28.4 ± 0.7 Ma) のジルコンに適用した予察の結果では, 2π Population 法, $S2\pi\text{ES}-I2\pi\text{ED}$ 法, 及び $S4\pi\text{IS}-I2\pi\text{ED}$ (External Detector) 法によって得られた年代は, $32.4-26.6$ Ma にわたる. それぞれの手法のもつ欠陥を補正すると, $28.2-27.9$ Ma になる. このような手法間比較を推しすすめることによって, 試料の地質年代学的背景に最適な手法が選択されるであろうと考えられる. 新しい, 自発トラック密度の低いジルコンの場合には, $S2\pi\text{ES}-I2\pi\text{ES}\cdot\text{ED}$ 法の有利なこと (ただし, $\rho_s < 1 \times 10^6 \text{ t}\cdot\text{cm}^{-2}$ の条件下で) をとくに言及した.