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DATING OF MINERALS**

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ABSTRACT

The effects of muon induced fission on geological dating of samples by the fission track method are evaluated for the case of muscovite minerals. It is found a small but significant effect, greater for the longer ages. Since calculations are developed under the hypothesis of constant atmosphere and primary cosmic ray flux it is suggested that any discrepancy found in ages of very old material that cannot be accounted for by well known environmental influences, be taken as an indication of variation on either the atmospheric stopping power or the intensity of cosmic radiation along the ages.

Key-words: Dating; Fission-track; Muons; Cosmic-rays.

INTRODUCTION

Geochronology studies received a powerful aid after the introduction of the fission track dating method, following the discovery of solid state track detectors among uranium containing minerals (Fleischer et al 1975). Although age evaluations are strongly affected by environmental influences such as thermal events, high pressure, leakage of uranium in or out the sample, etc, all situations are fairly well studied allowing precise age determinations in many cases.

Bombardment with particles of cosmic radiation may also lead to great nuisance since it may bring into the sample an additional source of tracks indistinguishable from the spontaneous fission tracks that constitute the basic component of the dating method. However, thanks to magnetic and atmospheric protection, strongly interacting particles of the primary cosmic radiation seldom come down to sea level or even to mountain altitudes thus keeping those effects well below safe limits. Unfortunately the same is not true of the secondaries generated in the atmosphere by the primary beam; among them neutrons and muons are the candidates potentially more effective to induce fission in uranium atoms of the sample.

Fast neutrons may fission ^{238}U nuclei and slow neutrons may fission ^{235}U . Most knowledge about atmospheric neutrons at sea level or mountain altitudes come from Simpson's monitors operating since the International Geophysical Year (Hatton 1971); they, however, do not teach enough about energy spectrum and absolute counting rates so that estimates are rather crude. Nevertheless neutrons are highly reactive particles and are strongly absorbed by large

amounts in nearly any surrounding material, so that a number presumably negligible is left to produce fission in the rare uranium atoms of the sample.

Muons, on the contrary, are weakly active nuclear interacting particles. Unstable, they are doomed to disintegrate if they are not captured before by a nucleus in an electron-like orbit and then get absorbed by the atomic nucleus. Nuclear absorption of muons has received the attention of many authors; nuclear fission in uranium isotopes is treated in more detail in (Ahmad et al 1986). Finally muons are charged particles so, contrary to the case of neutrons, the flux of arrival and energy spectrum are well known both at sea level and deep underground.

The purpose of this communication is to provide an estimate of the influence of muon induced fission on the ages determined by the fission track dating method, using the best data available.

FISSION TRACKS: SPONTANEOUS AND MUON INDUCED

In what follows the sample will be assumed to be a muscovite mineral; this will not imply restrictions other than those related to particular values of parameters that are required to reach quantitative results.

Uranium content of muscovite runs from the part per billion to the part per million level, per weight of mineral (Fleischer et al 1975). The two Uranium isotopes occurring more frequently in natural Uranium, ^{235}U , ^{238}U , are both unstable nuclei, decaying by alpha ray emission or by spontaneous fission with disintegration constants as shown in TABLE I. Depletion of the uranium content

in any sample occurs significantly only by alpha ray emission, as one can see by comparing the values of the disintegration constants in TABLE I. Also the number of tracks from spontaneous fission of ^{238}U will be always larger than those from spontaneous fission of ^{235}U in such a proportion that these can be neglected. Muscovite mica as a solid state track detector is insensitive to alpha rays; only fission tracks will be recorded and those will come from ^{238}U spontaneous fission as we have seen above.

The intercept of the fission tracks with the mica surface will be distributed uniformly with a density

$$\rho_{sf} = \lambda_{sf} T N_v q R_{238} \eta \quad (1)$$

where λ_{sf} is the disintegration constant for spontaneous fission of ^{238}U ; T is the age of the sample; N_v is the number of U-atoms per unit volume in the sample and q the fraction of them that are ^{238}U ; R_{238} is the range of the tracks left by the fission fragments and finally, η is the probability for a track to be seen after etching (Fleischer et al 1975).

As to the tracks from muon induced fission, they may originate in muon capture both in ^{235}U and ^{238}U . The absolute yields for both processes were measured by Ahmad et al 1986 and are reproduced in TABLE II, where yields are given in number of fissions per muon stopping in ^{235}U or ^{238}U (enriched targets). In a minor proportion direct electromagnetic excitation of fission modes is also capable to induce fission but photofission cross sections are small (O'Connell & Schima 1988) and that process will not contribute significantly.

Therefore, in order to use those results one needs the number of muons stopping in the sample per unit time and unit area that are captured by uranium nuclei. The simplest procedure is to use a relationship between the vertical muon intensity and the atmospheric depth (Miyake 1963; Menon & Ramanamurthy 1967); it has recently been revised by Barbouti & Rastin 1983, who improved its application to shallow atmospheric depths. Since that relation was obtained empirically it includes all relevant effects governing the diffusion of muons through atmosphere down to deep underground. In order to write an universal relation, depth is measured in units of hg/cm^2 of "standard rock": density $2.65 \text{ g}/\text{cm}^3$; average $Z/A = 0.5$; average $Z^2/A = 5.5$. The depth in such a unit can be converted to standard units by means of a simple formula that requires only knowledge of the muon stopping power of the medium (Menon & Ramanamurthy 1967). We will assume furthermore that our sample is a muscovite mica inset in standard rock.

The number of muons removed from the vertical beam per second, per cm^2 and per sr, as one goes through an extra step dh in depth is $S dh$, where:

$$S = dI_v/dh \quad (2)$$

where $I_v(h)$ is the vertical muon intensity as function of depth h as given by Miyake's formula. New muons are removed from the vertical beam because: 1) they decay in flight; 2) they are scattered off the vertical in dh ; 3) they stop in dh . The number of events belonging to cases 1) and 2) is negligibly small when compared to 3); therefore formula (2) will give also the number of muons stopping

in dh per unit time, per unit area and per sr.

Now it is required to know how many of those stopping muons are captured by uranium isotopes in the sample. This probability is a function of time, since the uranium content in the sample decreases continuously following alpha decay of ^{235}U and ^{238}U . Let $p(t)$, $q(t)$ be the relative concentrations of ^{235}U and ^{238}U in natural uranium as function of time; then it is easy to show that the probability for muon capture in uranium with fission of either ^{235}U or ^{238}U is:

$$W(t) = C(t) A_{\text{Mica}} [Y^{235} p(t) + Y^{238} q(t)] / A(t) \quad (3)$$

where $C(t)$ is the number of ppm of natural Uranium per weight of the sample, A_{Mica} is the molecular weight of muscovite mica, Y^{235} and Y^{238} are the yields given in TABLE II, $A(t)$ the atomic weight of natural uranium:

$$A(t) = 235 p(t) + 238 q(t) \quad (4)$$

The concentrations $p(t)$ and $q(t)$ are easily obtained from the relations governing disintegration rates and the values of the disintegration constants in TABLE I.

It is also easy to show that:

$$C(t) = (A(t)/A(T))C(T) \quad (5)$$

where T stands for present day values.

Before writing an expression for the surface density of tracks

from muon induced fissions two points have yet to be discussed: 1) the linear dimension, dh , where muon stop and produce etchable tracks; 2) muons also come from directions at an angle with the vertical, which were not accounted for in our evaluation.

As to the first point, it is well known that fission fragments have to intersect the surface of the track detector so as to be preferentially etched and become visible at the microscope; that means that only muons stopping at distances not greater than R_{238} (or R_{235}) at each side of a detector surface will be able to give origin to etchable tracks. We then take:

$$dH = 2(pR_{235} + qR_{238}) \approx 2R_{238} \quad (6)$$

As to the angular distribution of the muon beam it obeys the general shape:

$$I(\theta) = I_v \cos^m(\theta) \quad (7)$$

where m depends on atmospheric depth. We extrapolated to sea level Miyake's data measured deep underground (Menon & Ramanamurthy 1967) to obtain $m=1.15$. Therefore, in place of I_v one has to use the integration over angles of $I(\theta)$ in (7), giving the factor $4\pi I_v/2.15$.

Finally one obtains for the surface density of tracks from muon induced fissions:

$$\rho_{\mu f} = (4\pi/2.15) S 2R_{238} C(T) (A_{Mica}/A(T)) \eta J(T)$$

$$J(T) = \int_0^T (Y^{235} p(t) + Y^{238} q(t)) dt \quad (8)$$

For ρ_{sf} we transform equ. (1) by using

$$N_v = C(T) N_A \rho_{Mica} / A(T) ,$$

where N_A is Avogadro's number, to obtain

$$\rho_{sf} = \lambda_{sf} T C(T) N_A (\rho_{Mica} / A(T)) q R_{238}^n \quad (9)$$

The ratio $r = \rho_{\mu f} / \rho_{sf}$ for $h=10$ and $h=5$ is shown in TABLE III; $h=10$ is an atmospheric depth close to sea level and $h=5$ corresponds to an altitude a little higher than that of the Bolivian altiplane in Andes. At greater depth underground the effect becomes vanishingly small.

DISCUSSION AND CONCLUSIONS

The estimated errors in figures shown in TABLE III are less than 20%; in that sense they represent a small but significant contribution to the amount of tracks left by spontaneous fission of ^{238}U . How significant they are it depends however upon thinking over the following points:

1) We have used the disintegration constants for alpha decay recommended by the International Subcommittee on Geochronology, as quoted by Gale 1982, and their values are indeed accurately known but the same is not true of the disintegration constant for spontaneous fission of ^{238}U . We have used the value that has been adjusted by geochronologists by applying the fission track method to ages of samples known by other methods (Fleischer et al 1975) but

the fact is that the nearly 30 measurements of that quantity have produced a rather diffuse set of values between 10^{-16} and 10^{-17} year⁻¹.

2) this evaluation takes for granted that the atmosphere of Earth as well as the intensity of the cosmic radiation have always been as we know them nowadays. However any long lasting fluctuation in Earth's atmosphere, changing its density or composition in such a way as to alter significantly its stopping power for muons could change significantly our results, since both the angular distribution of the incident muons and their diffusion in the atmosphere down to deep underground could be affected. Unfortunately our calculation, because it uses the empirical formula of Miyake 1963, do not allow extrapolations to other atmospheric models.

As to the constance of cosmic ray intensity all that one can say is that for the last million years it has not shifted more than a factor two off the present day value (Honda 1967); for larger times, indications are uncertain but it is fair to expect significant time variations in cosmic ray primary intensity owing both to changes in solar modulation and to the wandering of the whole Solar System along and across the galactic arm throughout regions where the arrival of cosmic rays differs from the present one. If we take a factor four to account for those variations we see that figures in TABLE III would change proportionally and the effect of muon induced fission would become enormous at least for the most aged samples. Discrepancies with ages determined by other methods could then be used to study the history of our atmosphere and of the primary cosmic radiation.

TABLE I

Disintegration Constants for Alpha Ray Emission¹ and
Spontaneous Fission² for U-Isotopes (in yr⁻¹)

Isotope	$\lambda_{\alpha} \cdot 10^{10}$	$\lambda_{sf} \cdot 10^{17}$
²³⁵ U	9.8485 ± 0.0007	0.198 ± 0.18
²³⁸ U	1.5512 ± 0.0067	6.85 ± 0.20 ³

1-From Gale 1982; 2-From Lederer & Shirley 1978; 3-From Fleischer et al 1975

TABLE II

Absolute Fission Yields per Muon Stop
in ^{235}U and ^{238}U ¹

Isotope	Yield
^{235}U	0.142 ± 0.023
^{238}U	0.068 ± 0.013

1- From Ahmad et al 1986

TABLE III

Ratio of Surface Densities for Tracks from Muon Induced
Fission to Tracks from Spontaneous Fission

8

h (hg/cm ²)	T (years)				
	10 ⁵	10 ⁷	10 ⁸	10 ⁹	4 · 10 ⁹
10	3.65	3.65	3.70	3.90	4.80
5	5.85	5.85	5.95	6.30	7.70

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