Ber. nat.-med. Ver. Innsbruck	Band 64	S. 7-11	Innsbruck, Okt. 1977

Fission track measurement of the decay constant for spontaneous fission of ^{238}U

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Bestimmung der Zerfallskonstanten für den spontanen Zerfall von ^{238}U mit der Spaltspurenmethode

Synopsis: Unter Verwendung von Plastikdetektoren und einem neuen empfindlichen Ätzkanalnachweisverfahren wurde die Zerfallskonstante für den spontanen Zerfall von ^{238}U mit Hilfe der inversen Spaltspurenmethode neu bestimmt und ein Wert von $\lambda_f = (10{,}1 \pm 1{,}2) \cdot 10^{-17}\ a^{-1}$ erhalten.

Dieser Wert ist um ca. 30% größer als andere mit der inversen Spaltspurenmethode bestimmte Werte, stimmt jedoch innerhalb der angegebenen Fehlergrenzen überein mit den sehr verläßlichen Werten der Spaltspurenmessungen von datierten Uranglaswaren.

I. Introduction:

In view of the general geophysical significance of the spontaneous fission of ^{238}U, precise measurement of this decay constant λ_f is highly desirable. In recent studies of mineral ages by the fission track technique (FLEISCHER et alii, 1975), all of the necessary parameters except λ_f, were either known or measured to within 10% accuracy. In order to make these fission track mineral age determination on an absolute basis, λ_f, however, must be known to at least 10% accuracy.

Previously determined values of λ_f which appear to be somewhat reliable, range from 6,6 to $11{,}9 \cdot 10^{-17} yr^{-1}$ (THIEL, 1973). These values have been obtained mainly by three different methods: direct measurement of the spontaneous fission by means

Anschriften der Verfasser:

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of counters, chemical separation of radioactive fission products and measurement of their activity, and finally as in the present study, by means of a »reversed« fission track method where a solid state track recorder SSTR is brought into contact with natural uranium for several months. The latter method has two advantages: lower background than the first method and greater simplicity than the second method. In contrast to similar experiments in which plastic detectors are utilized as SSTR, in the present experiment a new technique to distinguish very easily real fission tracks from background effects (such as impurities or inhomogenities in the plastic detector) has been employed.

II. Experimental procedure:

Sheets of polycarbonate-foils (Macrofol from Bayer), initially containing no tracks were placed next to discs of natural metallic uranium (∅ = 10 mm, d = 1 mm). At first these sandwiches were stored for about 8 months ($3{,}67 \cdot 10^5$min) to collect spontaneous fission tracks. The sandwiches were stored at 4°C (refrigerator) to collect spontaneous fission tracks because it was suspected that at room temperatures track fading might occur in the polycarbonate detector due to annealing. An experiment carried out during this storage time showed, however, storage at room temperature not to reduce the track density during this period. At the end of the above storage time these sandwiches were provided with new polycarbonate-foils and irradiated*) with thermal neutrons to a dose of $(1{,}35 \pm 0{,}03)$ 10^9 neutrons/cm^2 in the ASTRA-reactor Seibersdorf, to collect induced fission events.

Special care was taken in the exact neutron dose determination, because a reliable absolute number of the integrated flux is essential to the present method. Since large flux gradients may be present in the thermal column of this reactor (see Fig. 3 in Märk et alii, 1974), the sandwiches were individually in close contact with two monitors suitable for low neutron dose determination ($< 1 \cdot 10^{10}$n/cm^2). In the present study ^{55}Mn neutron dosimeters were used. They have been checked for purity and homogeneity, and have yielded results consistent with those obtained by calibrated Au and Co monitors at various integrated neutron doses. A multichannel-gammacounter, whose efficiency was determined with a calibrated ^{54}Mn source, was used for absolute counting. Typical counting results of a monitor and a reference source were 15700 and 16500 counts respectively in 100s. Results obtained for the two monitors differed only within the error limits.

Fission tracks produced by the spontaneous fission of ^{238}U in the natural uranium disc and stored in the solid state foil detector for several months, have only a density of several 100/cm^2. This constitutes a serious challenge for accurate counting of

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*) In order to irradiate with this low neutron dose the samples had to stay in the thermal column for only ~ 1,5s. Thus the detectors were not subject to any annealing due to the temperature of ~ 80°C present in the thermal column.

tracks in the optical microscope, i.e. overlooking some of the spontaneous tracks would lead to a smaller value of λ_f and would be one explanation that all values determined by this method are rather small e.g. $(6{,}6 \pm 0{,}8) \cdot 10^{-17} yr^{-1}$ (FLEISCHER and PRICE, 1964), $(7{,}03 \pm 0{,}11) \cdot 10^{-17} yr^{-1}$ (*ROBERTS* et alii, 1968), $(6{,}8 \pm 0{,}6) \cdot 10^{-17} yr^{-1}$ (KLEEMAN and LOVERING, 1971), $(7{,}3 \pm 0{,}16) \cdot 10^{-17} yr^{-1}$ (LEME et alii, 1971) and $(6{,}82 \pm 0{,}55) \cdot 10^{-17} yr^{-1}$ (KHAN and DURRANI, 1973). As mentioned below, in the present study we have employed a new method to observe fission tracks in plastic foils with very high sensitivity.

Detectors containing spontaneous tracks and those containing induced tracks were etched together after the end of the storage period in 6n NaOH at 110°C for 5 minutes. After etching, the samples were glued to a glass-slide with glycerin and the etched fission tracks were observed in a phase contrast microscope by means of an immersion objective at an overall magnification of 800 and 1250, respectively. In addition, polarized condenser light was used and by optimizing with a polarization analyzer, the contrast (colour) between a real track and other track-like impurities could be considerably improved.

III. Results and Discussion:

Track densities counted independently by two of us (T.M., E.B.) are given in Table 1. The weighted average of the ratio p_s/p_i is 0,0123 ± 0,0010. In order to calculate λ_f from this measured ratio the formula for the fission track age determination for ages $< 10^8$ yr:

$$t = \frac{p_s}{p_i} \quad \frac{n \cdot \sigma_f \cdot I}{\lambda_f}$$

can be used, where p_s and p_i are the spontaneous and induced track densities, n the thermal neutron dose, σ the cross section for thermal fission of ^{235}U and I is the ratio of the abundance of ^{235}U and ^{238}U. The values of σ and I were taken to be $\sigma = 580{,}2 \pm 1{,}8$ barn and $I = (7{,}259 \pm 0{,}104) \cdot 10^3$ (THIEL, 1973). With these values follows from the above equation:

$$\lambda_f = (10{,}1 \pm 1{,}2) \cdot 10^{-17} yr^{-1}.$$

This result ist somewhat higher when compared with other values obtained with the same method, it corresponds however within the experimental error with the values $(8{,}57 \pm 0{,}42) \cdot 10^{-17} yr^{-1}$ (THIEL, 1973), $(8{,}49 \pm 0{,}76) \cdot 10^{-17} yr^{-1}$ (STORZER, 1970) and $8{,}4 \cdot 10^{-17} yr^{-1}$ (GENTER et alii, 1972). The latter values have been derived from fission track measurements with dated uranium-glassware where the spontaneous fission tracks are also detectable with high sensitivity and reliability (THIEL, 1973). Track fading during the storage time of the spontaneous fission tracks in the glass detector at room temperature is probably negligible. In both cases, the glassware determination and the present determination, alterations of the spontaneous fission track density by fission tracks induced by the cosmic rays is negligible according to an estimate made by THIEL (1973).

Table 1

Experimentally obtained spontaneous and induced track densities p_s and p_i, i.e. sample MMP1 - 4 induced, and MMP6 - 8 spontaneous fission tracks, respectively. Tracks were counted in the microscope by T.M. with an overall magnification of 800 and by E.B., of 1250. This might account partly for the different absolute track densities counted by T.M. and E.B.

	determination by T.M.		determination by E.B.	
	absolute number of counted tracks	track density in cm^{-2}	absolute number of counted tracks	track density in cm^{-2}
MMP 1	481	$(4{,}84 \pm 0{,}22) \cdot 10^4$	158	$(5{,}85 \pm 0{,}46) \cdot 10^4$
MMP 2	439	$(4{,}48 \pm 0{,}22) \cdot 10^4$	142	$(5{,}56 \pm 0{,}47) \cdot 10^4$
MMP 3	459	$(4{,}29 \pm 0{,}21) \cdot 10^4$	131	$(5{,}78 \pm 0{,}50) \cdot 10^4$
MMP 4	508	$(4{,}84 \pm 0{,}22) \cdot 10^4$	142	$(6{,}16 \pm 0{,}52) \cdot 10^4$
$\bar{p}_i$		$(4{,}62 \pm 0{,}13) \cdot 10^4$		$(5{,}84 \pm 0{,}28) \cdot 10^4$
MMP 6	100	543 ± 54	50	738 ± 104
MMP 7	100	590 ± 59	50	804 ± 114
MMP 8	100	560 ± 56	50	665 ± 93
$\bar{p}_s$		564 ± 39		736 ± 74
$\bar{p}_s / \bar{p}_i$		$0{,}0122 \pm 0{,}0012$		$0{,}0126 \pm 0{,}0019$

V. Acknowledgements:

The authors want to express their gratitude to the Österreichischer Fonds zur Förderung der Wissenschaftlichen Forschung for financial assistance under Project Nr. 839 and 1932. The authors also wish to thank the Studiengesellschaft für Atomenergie in Seibersdorf for irradiating the samples.

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