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# Standard Test Method for Application and Analysis of Solid State Track Recorder (SSTR) Monitors for Reactor Surveillance, E706(IIIB)<sup>1</sup>

This standard is issued under the fixed designation E 854; the number immediately following the designation indicates the year of original adoption or, in the case of revision, the year of last revision. A number in parentheses indicates the year of last reapproval. A superscript epsilon ( $\epsilon$ ) indicates an editorial change since the last revision or reapproval.

### 1. Scope

1.1 This test method describes the use of solid-state track recorders (SSTRs) for neutron dosimetry in light-water reactor (LWR) applications. These applications extend from low neutron fluence to high neutron fluence, including high power pressure vessel surveillance and test reactor irradiations as well as low power benchmark field measurement. (1) This test method replaces Method E 418. This test method is more detailed and special attention is given to the use of state-of-the-art manual and automated track counting methods to attain high absolute accuracies. In-situ dosimetry in actual high fluence-high temperature LWR applications is emphasized.

1.2 This test method includes SSTR analysis by both manual and automated methods. To attain a desired accuracy, the track scanning method selected places limits on the allowable track density. Typically good results are obtained in the range of 5 to 800 000 tracks/cm<sup>2</sup> and accurate results at higher track densities have been demonstrated for some cases. (2) Track density and other factors place limits on the applicability of the SSTR method at high fluences. Special care must be exerted when measuring neutron fluences (E>1MeV) above  $10^{16}$  n/cm<sup>2</sup>. (3)

1.3 High fluence limitations exist. These limitations are discussed in detail in Section 13 and in references (3-5).

1.4 SSTR observations provide time-integrated reaction rates. Therefore, SSTR are truly passive-fluence detectors. They provide permanent records of dosimetry experiments without the need for time-dependent corrections, such as decay factors that arise with radiometric monitors.

1.5 Since SSTR provide a spatial record of the timeintegrated reaction rate at a microscopic level, they can be used for "fine-structure" measurements. For example, spatial distributions of isotopic fission rates can be obtained at very high resolution with SSTR.

1.6 This standard does not purport to address the safety problems associated with its use. It is the responsibility of the user of this standard to establish appropriate safety and health

practices and determine the applicability of regulatory limitations prior to use.

#### 2. Referenced Documents

- 2.1 ASTM Standards:
- E 418 Method for Fast-Neutron Measurements by Track-Etch Techniques<sup>2</sup>
- E 844 Guide for Sensor Set Design and Irradiation for Reactor Surveillance, E706 (IIC)<sup>3</sup>

#### 3. Summary of Test Method

3.1 SSTR are usually placed in firm surface contact with a fissionable nuclide that has been deposited on a pure nonfissionable metal substrate (backing). This typical SSTR geometry is depicted in Fig. 1. Neutron-induced fission produces latent fission-fragment tracks in the SSTR. These tracks may be developed by chemical etching to a size that is observable with an optical microscope. Microphotographs of etched fission tracks in mica, quartz glass, and natural quartz crystals can be seen in Fig. 2.

3.1.1 While the conventional SSTR geometry depicted in Fig. 1 is not mandatory, it does possess distinct advantages for dosimetry applications. In particular, it provides the highest efficiency and sensitivity while maintaining a fixed and easily reproducible geometry.

3.1.2 The track density (that is, the number of tracks per unit area) is proportional to the fission density (that is, the number of fissions per unit area). The fission density is, in turn, proportional to the exposure fluence experienced by the SSTR. The existence of nonuniformity in the fission deposit or the presence of neutron flux gradients can produce non-uniform track density. Conversely, with fission deposits of proven uniformity, gradients of the neutron field can be investigated with very high spatial resolution.

3.2 The total uncertainty of SSTR fission rates is comprised of two independent sources. These two error components arise from track counting uncertainties and fission-deposit mass uncertainties. For work at the highest accuracy levels, fissiondeposit mass assay should be performed both before and after the SSTR irradiation. In this way, it can be ascertained that no significant removal of fission deposit material arose in the

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<sup>&</sup>lt;sup>2</sup> Discontinued; see 1983 Annual Book of ASTM Standards, Vol 12.02.

<sup>&</sup>lt;sup>3</sup> Annual Book of ASTM Standards, Vol 12.02.

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course of the experiment.

## 4. Significance and Use

4.1 The SSTR method provides for the measurement of absolute-fission density per unit mass. Absolute-neutron fluence can then be inferred from these SSTR-based absolute fission rate observations if an appropriate neutron spectrum average fission cross section is known. This method is highly discriminatory against other components of the in-core radiation field. Gamma rays, beta rays, and other lightly ionizing particles do not produce observable tracks in appropriate LWR SSTR candidate materials. However, photofission can contribute to the observed fission track density and should therefore be accounted for when nonnegligible. For a more detailed discussion of photofission effects, see 13.4.

4.2 In this test method, SSTR are placed in surface contact with fissionable deposits and record neutron-induced fission fragments. By variation of the surface mass density  $(\mu g/cm^2)$  of the fissionable deposit as well as employing the allowable range of track densities (from roughly 1 event/cm<sup>2</sup> up to  $10^{5}$ events/cm<sup>2</sup> for manual scanning), a range of total fluence sensitivity covering at least 16 orders of magnitude is possible, from roughly  $10^2$  n/cm<sup>2</sup> up to 5 ×  $10^{18}$  n/cm<sup>2</sup>. The allowable range of fission track densities is broader than the track density range for high accuracy manual scanning work with optical microscopy cited in 1.2. In particular, automated and semiautomated methods exist that broaden the customary track density range available with manual optical microscopy. In this broader track density region, effects of reduced counting statistics at very low track densities and track pile-up corrections at very high track densities can present inherent limitations for work of high accuracy. Automated scanning techniques are described in Section 11.

4.3 For dosimetry applications, different energy regions of the neutron spectrum can be selectively emphasized by changing the nuclide used for the fission deposit.

4.4 It is possible to use SSTR directly for neutron dosimetry as described in 4.1 or to obtain a composite neutron detection efficiency by exposure in a benchmark neutron field. The fluence and spectrum-averaged cross section in this benchmark field must be known. Furthermore, application in other neutron fields may require adjustments due to spectral deviation from the benchmark field spectrum used for calibration. In any event, it must be stressed that the SSTR-fission density measurements can be carried out completely independent of any cross-section standards (6). Therefore, for certain applications, the independent nature of this test method should not be compromised. On the other hand, many practical applications exist wherein this factor is of no consequence so that benchmark field calibration would be entirely appropriate.

## 5. Apparatus

5.1 *Optical Microscopes*, with a magnification of  $200 \times \text{or}$  higher, employing a graduated mechanical stage with position readout to the nearest 1  $\mu$ m and similar repositioning accuracy. A calibrated stage micrometer and eyepiece scanning grids are also required.

5.2 *Constant-Temperature Baths*, for etching, with temperature control to 0.1°C.

5.3 Analytical Weighing Balance, for preparation of etching bath solutions, with a capacity of at least 1000 g and an accuracy of at least 1 mg.

## 6. Reagents and Materials

6.1 *Purity of Reagents*—Distilled or demineralized water and analytical grade reagents should be used at all times. For high fluence measurements, quartz-distilled water and ultrapure reagents are necessary in order to reduce background fission tracks from natural uranium and thorium impurities. This is particularly important if any pre-irradiation etching is performed (see 8.2).

6.2 Reagents:

- 6.2.1 Hydrofluoric Acid (HF), weight 49 %.
- 6.2.2 Sodium Hydroxide Solution (NaOH), 6.2 N.
- 6.2.3 Distilled or Demineralized Water.
- 6.2.4 Potassium Hydroxide Solution (KOH), 6.2 N.
- 6.2.5 Sodium Hydroxide Solution (NaOH), weight 65 %.
- 6.3 Materials:
- 6.3.1 Glass Microscope Slides.
- 6.3.2 Slide Cover Glasses.

## 7. SSTR Materials for Reactor Applications

7.1 Required Properties—SSTR materials for reactor applications should be transparent dielectrics with a relatively high ionization threshold, so as to discriminate against lightly ionizing particles. The materials that meet these prerequisites most closely are the minerals mica, quartz glass, and quartz crystals. Selected characteristics for these SSTR are summarized in Table 1. Other minerals such as apatite, sphene, and zircon are also suitable, but are not used due to inferior etching properties compared to mica and quartz. These alternative SSTR candidates often possess either higher imperfection density or poorer contrast and clarity for scanning by optical microscopy. Mica and particularly quartz can be found with the additional advantageous property of low natural uranium and thorium content. These heavy elements are undesirable in neutron-dosimetry work, since such impurities lead to background track densities when SSTR are exposed to high neutron fluence. In the case of older mineral samples, a background of fossil fission track arises due mainly to the spontaneous fission decay of <sup>238</sup>U. Glasses (and particularly phosphate glasses) are NOTICE: This standard has either been superceded and replaced by a new version or discontinued. Contact ASTM International (www.astm.org) for the latest information.

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NOTE 1—The track designated by the arrow in the mica SSTR is a fossil fission track that has been enlarged by suitable preirradiation etching. FIG. 2 Microphotograph of Fission Fragment Tracks in Mica

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less suitable than mica and quartz due to higher uranium and thorium content. Also, the track-etching characteristics of many glasses are inferior, in that these glasses possess higher bulk etch rate and lower registration efficiency. Other SSTR materials, such as Lexan<sup>4</sup> and Makrofol<sup>5</sup> are also used, but are less convenient in many reactor applications due to the presence of neutron-induced recoil tracks from elements such as carbon and oxygen present in the SSTR. These detectors are also more sensitive (in the form of increased bulk etch rate) to the  $\beta$  and  $\gamma$  components of the reactor radiation field (13). Also, they are more sensitive to high temperatures, since the onset of track annealing occurs at a much lower temperature for plastic SSTR materials.

#### 7.2 Limitations of SSTR in LWR Environments:

7.2.1 *Thermal Annealing*—High temperatures result in the erasure of tracks due to thermal annealing. Natural quartz crystal is least affected by high temperatures, followed by mica. Lexan and Makrofol are subject to annealing at much lower temperatures. An example of the use of natural quartz

crystal SSTRs for high-temperature neutron dosimetry measurements is the work described in reference (14).

7.2.2 *Radiation Damage*—Lexan and Makrofol are highly sensitive to other components of the radiation field. As mentioned in 7.1, the bulk-etch rates of plastic SSTR are increased by exposure to  $\beta$  and  $\gamma$  radiation. Quartz has been observed to have a higher bulk etch rate after irradiation with a fluence of  $4 \times 10^{21}$  neutrons/cm<sup>2</sup>, but both quartz and mica are very insensitive to radiation damage at lower fluences (<10<sup>21</sup> neutrons/cm<sup>2</sup>).

7.2.3 *Background Tracks*—Plastic track detectors will register recoil carbon and oxygen ions resulting from neutron scattering on carbon and oxygen atoms in the plastic. These fast neutron-induced recoils can produce a background of short tracks. Quartz and mica will not register such light ions and are not subject to such background tracks.

#### 8. SSTR Pre- and Post-Irradiation Processing

## 8.1 Pre-Irradiation Annealing:

8.1.1 In the case of mica SSTR, a pre-annealing procedure designed to remove fossil track damage is advisable for work at low neutron fluences. The standard procedure is annealing

<sup>&</sup>lt;sup>4</sup> Lexan is a registered trademark of the General Electric Co., Pittsfield, MA. <sup>5</sup> Makrofol is a registered trademark of Farbenfabriken Bayer AG, U. S.

<sup>&</sup>lt;sup>2</sup> Makrotol is a registered trademark of Farbenfabriken Bayer AG, U. 3 representative Naftone, Inc., New York, NY.

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FIG. 2 Quartz Glass (continued)

FIG. 2 Quartz Crystal (001 Plane) (continued)

for 6 h at 600°C (longer time periods may result in dehydration). Fossil track densities are so low in good Brazilian quartz crystals that pre-annealing is not generally necessary. Annealing is not advised for plastic SSTR because of the possibility of thermal degradation of the polymer or altered composition, both of which could effect track registration properties of the plastic.

### 8.2 Pre-Irradiation Etching:

8.2.1 *Mica*—Unannealed fossil tracks in mica are easily distinguished from induced tracks by pre-etching for a time that is long compared to the post-etching conditions. In the case of mica, a 6-h etch in 48 % HF at room temperature results in large diamond-shaped tracks that are easily distinguished from the much smaller induced tracks revealed by a 90-min post-etch (see Fig. 2)).

8.2.2 *Quartz Crystals*—Pre-etching is needed to chemically polish the surface. Polish a crystal mechanically on the 001 or 100 plane so that it appears smooth under microscopical examination, etch for 10 min in 49 % HF at room temperature, then boil in 65 % NaOH solution for 25 min. Examine the crystal surface microscopically. If it is sufficiently free of pits, select it for use as an SSTR.

8.2.3 *Quartz Glass*—If the glass has been polished mechanically, or has a smooth surface, then pre-etch in 49 % HF for 5 min at room temperature. Upon microscopical examination a few etch pits may be present even in good-quality quartz glass. If so, they will be larger than tracks due to fission fragments revealed in the post-etch, and readily distinguished from them.

8.2.4 *Plastic-Track Recorders*—If handled properly, background from natural sources, such as radon, will be negligible. Consequently, both preannealing and pre-etching should be unnecessary.

8.3 Post-Irradiation Etching:

8.3.1 *Mica*—Customary etching is for 90 min in 49 % HF at room temperature. Both the etch time and temperature may be varied to give optimum track sizes for the particular type of mica used. Except for work at the highest accuracy levels, precise control of the temperature is not necessary due to the zero bulk etch rate of the mica perpendicular to the cleavage planes. In the event that precise etching control is necessary, a technique has been demonstrated for mica that permits highly reproducible and standardized track size distributions (10).

8.3.2 *Quartz Crystals*—Etch for 25 min in boiling 65 % NaOH solution. Minimize evaporation by covering the nickel or platinum crucible in which the solution is heated. If left open, condense evaporated water and return to the solution. The value of the optical efficiency is dependent on the etching conditions (since the bulk etch rate is not zero), so both the concentration of the NaOH solution and the etching temperature must be controlled.

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TABLE 1	Characteristics of	SSTR	Candidates f	or LWR	Reactor	Applications
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SSTR	Optical Efficiency, %	Asymptotic <sup>A</sup> Sensitivity	Conditions Under Which Accurate An- nealing Corrections Can Be Made	Track Reduction, %
Muscovite mica	$0.9875 \pm 0.0085^B$	$(1.114 \pm 0.018)  imes 10^{19}$ $^{238}$ U atoms/cm $^{2B}$	501°C, 146.5 h <sup>C</sup>	0 <sup><i>c</i></sup>
Makrofol N Quartz glass Natural quartz	$95.2 \pm 0.53^{D}$ $\sim 70^{E}$ $\sim 80^{E}$	  	 402°C, 8 h <sup>C</sup> 857°C, 1 h <sup>F</sup>	73 <sup>C</sup> 20 <sup>F</sup>
Crystal				

<sup>A</sup> Needs to be known only if used with asymptotically thick sources.

<sup>B</sup> Etched 90 min in 49 % HF (6, 7, 8).

<sup>C</sup> Data from Ref (9).

<sup>D</sup> Etched ~20 h in 6.2 N KOH solution at room temperature (6).

<sup>E</sup> Quartz glass etched 5 min in 48 % HF at room temperature. Quartz crystal etched in boiling 65 % NaOH solution for 25 min (10, 11).

<sup>F</sup> Data from Ref (12).

8.3.3 *Quartz Glass*—Etch for 5 min in 48 % HF at room temperature. Temperature control is essential because of the high bulk etch rate.

8.3.4 Lexan,<sup>4</sup> or Makrofol<sup>5</sup>, N—Various time temperature combinations in 6.2 N NaOH or KOH solution have proved satisfactory, depending upon the desired purpose. Examples of appropriate conditions are: (1) 50 h in 6.2 N NaOH solution at 20°C, (2) 24 h in 6.2 N KOH solution at 20°C, and (3) 30 min in 6.2 N KOH solution at 50°C.

#### 9. SSTR Fissionable Deposits

#### 9.1 Properties:

9.1.1 *Fission Deposit Characteristics*— Perhaps the most critical factor in attaining high accuracy in SSTR neutron dosimetry is the quality of the fission deposit. High quality SSTR fission deposits possess the following characteristics: (6-17)

9.1.1.1 Accurately known total mass and mass density. The overall accuracy of the mass calibration must be consistent with the desired overall accuracy of the measurement.

9.1.1.2 Accurately known isotopic composition. Possible interfering isotopes must be minimized and the overall fission rate must be corrected for contributions from interfering isotopes.

9.1.1.3 *Negligible Impurities*—Impurities that contribute to the measured fission rate must be minimized (<1 % contribution) and the overall fission rate must be corrected for contributions from impurities.

9.1.1.4 High uniformity is recommended. An independent measurement is required which verifies the uniformity of the deposit to an uncertainty commensurate with the desired accuracy of subsequent measurements using the deposit. Conversely, use of nonuniform deposits entails scanning of the entire SSTR surface to attain accurate results.

9.1.2 As has already been stated in 3.2, the accuracy of fission deposit characterization provides a fundamental limitation for the accuracy of the SSTR method. Fission-deposit mass assay as well as uniformity are important. Dosimetry goal accuracies provide bounds for the acceptable quality of SSTR fission deposits. For work at the highest accuracy levels, fission deposits can be prepared at close to or better than 1 % mass assay. Less accurate SSTR dosimetry can, however, be performed at a lower cost with less stringent requirements for fission deposit characterization. The deposit backing should

contribute negligible background and the deposit should be flat, rigid, and capable of maintaining good contact with the SSTR. The deposit should be firmly adherent to the backing. The appropriate mass density for a particular LWR application may be calculated from:

$$\phi t \times W = \frac{\rho M}{\eta N_o \bar{\sigma} I} \tag{1}$$

where:

ρ

Ι

σ

 $\phi t$  = the expected fluence,

W = the mass density of the deposit, g/cm<sup>2</sup>,

- = the track density (the optimum track density for most manual scanning is about  $5 \times 10^4$  tracks/cm<sup>2</sup>),
- = the isotopic abundance (atomic fraction),

 $\eta$  = the optical efficiency of the SSTR,

- = the spectral average fission cross section,
- M = the average atomic weight of the isotopic mixture used, and

 $N_o =$ Avogadro's number (6.022 × 10<sup>23</sup>).

9.1.3 In Eq 1, the assumption is made that the thickness (mass density) of the deposit is much less than the range of a fission fragment in the deposit material. Under these conditions, self-absorption is negligible and sensitivity depends linearly on *W*. For deposit thicknesses greater than about 100  $\mu$ /cm<sup>2</sup>, self-absorption of fission fragments by the deposit becomes increasingly important. For deposit thicknesses greater than twice the range of a fission fragment in the deposit material, the effective thickness may be represented by a constant value. This constant value is referred to as the asymptotic sensitivity,  $s_{\infty}$ . It can be analytically shown (**6**) for a uniform deposit with no flux depression that the asymptotic sensitivity is approximately given by:

$$s_{\infty} \simeq \eta \, \frac{(R)}{2} \tag{2}$$

where:

 $\langle R \rangle$  = the mean fission fragment range in the deposit.

In the case of uranium metal, an asymptotic sensitivity of  $4.522 \pm 0.070 \text{ mg/cm}^2$  has been measured (6,8). Thicknesses in the approximate range from 0.1 to 30 mg/cm<sup>2</sup>should be avoided due to problems arising from self-absorption of fission fragments in the source. While it is possible to work in this range, additional error will be incurred due to the need to correct for self-absorption. In the region beyond 30 mg/cm<sup>2</sup>,

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one should use the asymptotic sensitivity.

9.2 *Isotypes Required*—In general, when performing reaction rate measurements for a particular isotope, contributions to the fission rate from other isotopes must be either negligible or corrected with sufficient accuracy. For example, use of the threshold reaction  $^{238}$ U (n,f) in a neutron field where the thermal flux is appreciable requires highly depleted uranium in order to minimize contributions from  $^{235}$ U (n,f). Similarly chemical purity must be taken into account. When measuring the reaction rate for an even-even nuclide such as  $^{240}$ Pu, the abundance of the fissionable even-odd isotopic neighbors  $^{239}$ Pu and  $^{241}$ Pu must be minimized. For low-flux measurements, contributions from spontaneously fissioning nuclides must be minimized and if necessary spontaneous fission track contributions must be subtracted.

## 9.3 Source Preparation:

9.3.1 Electrodeposition and vacuum deposition are the most frequently used and the most effective techniques. The latter method normally results in more uniform deposits, but economy of material and convenience may favor the former. In both cases, actinide deposits are produced more easily in the oxide than in the metallic form. Adherence of the deposit to the backing material can often be accomplished by heating the deposit to red heat in an inert atmosphere. Uniformity can be demonstrated by  $\alpha$ -autoradiography using an  $\alpha$ -sensitive SSTR such as cellulose nitrate or by fission track radiography with uniform neutron field irradiations.

9.3.2 Metallic backing for the fission deposit should be chosen to meet a number of requirements. For dosimetry purposes the backing should only be thick enough to ensure firm contact between the track recorder and the deposit (see Fig. 1). Furthermore, since it is preferable that no foreign elements be introduced into the radiation environment, backing materials should be chosen wherever possible from constituent elements that already exist in the radiation environment. Neutron field perturbations due to the backing are considered in Section 12. For high-fluence measurements, extremely pure-backing materials are required in order to reduce background fission tracks from natural uranium and thorium impurities. The surface of the backing material must be smooth and preferably possess a mirror finish.

#### 9.4 Mass Assay:

9.4.1 Absolute Disintegration Rate—Mass assay may be accomplished by absolute  $\alpha$ -counting using a low geometry  $\alpha$ -counter (6). In many cases, the alpha decay constant is known to an accuracy of better than 1 %. In fact, the uncertainty of the alpha decay constant provides a fundamental limitation in this mass-assay method. Relative masses of several sources of the same isotope may be established to better than 1 % by  $\alpha$ -counting in a  $2\pi$  proportional counter. (See Table 2 for a summary of alpha decay constants of the actinide elements (15).)

9.4.2 *Mass Spectrometry*—Mass spectrometry combined with isotopic dilution techniques is a potentially useful method for mass assay of deposits. Mass spectrometry is particularly useful for low specific activity isotopes or isotopes with decay constants that have not been measured to an accuracy of 1 %. While mass spectrometry can provide accuracies of better than 1 %, it suffers from an inherent disadvantage, namely the need for destructive analysis.

9.4.3 *Isotopic Spikes*—High specific activity isotopes may be used as a tracer to indicate target mass. Alpha active isotopes such as <sup>230</sup>Th, <sup>236</sup>Pu, and <sup>238</sup>Pu as well as  $\gamma$ -emitting isotopes such as <sup>237</sup>U and <sup>239</sup>Np are useful for relative mass determinations. When using isotopic spikes, care must be taken to ensure that the source isotope and the spike are chemically equivalent. Also, the fission rate of the isotopic spike and its daughter products should be kept negligible compared to the fission rate of the isotope of interest. The use of isotopic spikes that feed complex decay chains (such as <sup>228</sup>Th and <sup>232</sup>U) should be avoided.

9.4.4 Less Frequently Used Methods—Ion, X-ray, and Auger microprobe analysis, X-ray fluorescence, neutron activation analysis, and wet chemical analysis methods may be useful for specific applications, but rarely attain an accuracy comparable to previously mentioned methods.

9.5 Ultra Low-mass Deposits—Methods for producing and calibrating ultra low-mass fissionable deposits are described in reference (3). Because of the low masses involved, typically  $10^{-14}$  to  $10^{-9}$  grams, care must be taken to avoid contamination of the deposits. Therefore, the deposits must be made under

TABLE 2	Decay	Constants a	and Associated	Uncertainties	Used in	Actinide Mass	Quantification
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Nuclide	$t_{1/2}$ (years)	$\lambda(s^{-1})$	Uncertainty, %	Reference (15)		
				Vol	No.	Date
<sup>230</sup> Th	$(7.538 \pm 0.030) \times 10^4$	$2.914  imes 10^{-13}$	0.40	40	3	1983
<sup>232</sup> Th	$(1.405\pm0.006) \times 10^{10}$	$1.563  imes 10^{-18}$	0.43	36	3	1982
<sup>233</sup> U	$(1.592 \pm 0.020) \times 10^{5}$	$1.380  imes 10^{-13}$	1.26	24	2	1978
<sup>234</sup> U	$(2.45 \pm 0.02) \times 10^{5}$	$8.965  imes 10^{-14}$	1.38	40	4	1983
<sup>235</sup> U	$(7.038 \pm 0.005) \times 10^8$	$3.121  imes 10^{-17}$	0.071	40	1	1983
<sup>237</sup> U <sup>A</sup>	6.75 ± 0.01 days	$1.189 imes10^{-6}$	0.15	23	1	1978
<sup>238</sup> U	$(4.468 \pm 0.003) \times 10^9$	$4.916  imes 10^{-18}$	0.067	38	2	1983
<sup>237</sup> Np	$(2.14 \pm 0.01) \times 10^{6}$	$1.026  imes 10^{-14}$	0.47	23	1	1978
<sup>239</sup> Np <sup>A</sup>	2.355 ± 0.004 days	$3.407 imes10^{-6}$	0.17	40	1	1983
<sup>236</sup> Pu <sup>A</sup>	2.851 ± 0.008	$7.704 imes10^{-9}$	0.28	36	3	1982
<sup>238</sup> Pu	$(8.774 \pm 0.004) \times 10^{1}$	$2.503  imes 10^{-10}$	0.046	38	2	1983
<sup>239</sup> Pu	$(2.4119 \pm 0.026) \times 10^4$	$9.107  imes 10^{-13}$	0.11	40	1	1983
<sup>240</sup> Pu	$(6.569 \pm 0.006) \times 10^3$	$3.344  imes 10^{-12}$	0.091	43	2	1984
<sup>241</sup> Pu <sup>B</sup>	$(1.4355 \pm 0.0007) \times 10^{1}$	$1.530  imes 10^{-9}$	0.049	44	2	1984
<sup>242</sup> Pu	$(3.733 \pm 0.012) \times 10^{5}$	$5.884 \times 10^{-14}$	0.32	45	3	1985

<sup>A</sup> Tracer materials used for quantification of low mass primary deposits (may be  $\alpha$  or  $\beta/\gamma$  emitters, or both).

<sup>*B*</sup> The branching ratio for alpha emission is  $(2.46 \pm 0.01) \times 10^{-3}$  %. The partial half-life for alpha decay is  $5.79 \times 10^{5}$  years  $(\pm 3.2 \text{ \%})$ .