

Standard Test Method for Measuring Neutron Fluence Rates by Radioactivation of Cobalt and Silver¹

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1. Scope

1.1 This test method covers a suitable means of obtaining the thermal neutron fluence rate, or fluence, in well moderated nuclear reactor environments where the use of cadmium, as a thermal neutron shield as described in Test Method E262, is undesirable because of potential spectrum perturbations or of temperatures above the melting point of cadmium.

1.2 This test method describes a means of measuring a Westcott neutron fluence rate (Note 1) by activation of cobaltand silver-foil monitors (See Terminology E170). The reaction 59 Co $(n,\gamma)^{60}$ Co results in a well-defined gamma emitter having a half-life of 1925.28 days (1).² The reaction 109 Ag(n, γ) 110m Ag results in a nuclide with a complex decay scheme which is well known and having a half-life of 249.76 days (1). Both cobalt and silver are available either in very pure form or alloyed with other metals such as aluminum. A reference source of cobalt in aluminum alloy to serve as a neutron fluence rate monitor wire standard is available from the National Institute of Standards and Technology (NIST) as Standard Reference Material 953.³ The competing activities from neutron activation of other isotopes are eliminated, for the most part, by waiting for the short-lived products to die out before counting. With suitable techniques, thermal neutron fluence rate in the range from 10⁹ cm⁻² · s⁻¹ to 3×10^{15} cm⁻² · s⁻¹ can be measured. For this method to be applicable, the reactor must be well moderated and be well represented by a Maxwellian low-energy distribution and an (1/E) epithermal distribution. These conditions are usually met in positions surrounded by hydrogenous moderator without nearby strongly absorbing materials. Otherwise, the true spectrum must be calculated to obtain effective activation cross sections over all energies.

Note 1—Westcott fluence rate = $v_0 \int_0^\infty n(v) dv$.

- 1.3 The values stated in SI units are to be regarded as the tandard.
- 1.4 This standard does not purport to address all of the safety concerns, if any, 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:⁴

E170 Terminology Relating to Radiation Measurements and Dosimetry

E177 Practice for Use of the Terms Precision and Bias in ASTM Test Methods

E181 Test Methods for Detector Calibration and Analysis of Radionuclides

E261 Practice for Determining Neutron Fluence, Fluence Rate, and Spectra by Radioactivation Techniques

E262 Test Method for Determining Thermal Neutron Reaction Rates and Thermal Neutron Fluence Rates by Radioactivation Techniques

3. Significance and Use

3.1 This test method uses one monitor (cobalt) with a nearly 1/v absorption cross-section curve and a second monitor (silver) with a large resonance peak so that its resonance integral is large compared to the thermal cross section. The pertinent data for these two reactions are given in Table 1. The equations are based on the Westcott formalism ((2, 3) and Practice E261) and determine a Westcott 2200 m/s neutron fluence rate nv_0 and the Westcott epithermal index parameter $r\sqrt{T/T_0}$. References (4, 5, and 6) contain a general discussion of the two-reaction test method. In this test method, the absolute activities of both cobalt and silver monitors are determined. This differs from the test method in the references wherein only one absolute activity is determined.

3.2 The advantages of this test method are the elimination of three difficulties associated with the use of cadmium: (1) the

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 $^{^{2}\,\}mathrm{The}$ boldface numbers in parentheses refer to references listed at the end of this test method.

³ Standard Reference Material 953 is available from National Institute of Standards and Technology, U.S. Dept. of Commerce, Washington, DC 20234.

⁴ For referenced ASTM standards, visit the ASTM website, www.astm.org, or contact ASTM Customer Service at service@astm.org. For *Annual Book of ASTM Standards* volume information, refer to the standard's Document Summary page on the ASTM website.

TABLE 1 Recommended Constants

Cumphal	ol Parameter —	Cobalt (60Co)		Silver (110mAg)	
Symbol		Value ^A	Reference	Value ^A	Reference
t _{1/2}	Half-life	1925.28 (14) days	(1)	249.76 (4) days	(1)
A	Abundance of parent isotope	100 % (⁵⁹ Co)	(1)	48.161 (8) % (¹⁰⁹ Ag)	(<mark>1</mark>)
σ_a	Absorption 2200 m/s cross section for target ⁵⁹ Co and ¹⁰⁹ Ag	37.233 b ± 0.16 %	B,C	91.0 b ± 1 %	(7)
σ_0	2200 m/s cross section for formation of ⁶⁰ Co and ^{110m} Ag	37.233 b ± 0.16 %	B,C	4.12 (10)	(8)
S_0	Correction factor which describes the departure of the cross section from the 1/v law in the epithermal region	1.80 [⁵⁹ Co(n,γ) ⁶⁰ Co]	D	$18.1(7)$ [109 Ag(n,γ) 110m Ag]	(8)
I ₀	Resonance Integral	75.421 b ± 0.77 % [59 Co(n, γ) 60 Co]	(9) ^E	67.9 (31) b [¹⁰⁹ Ag(n,γ) ^{110m} Ag]	(8)
σ_2	Effective absorption cross section for product nuclide (reactor spectrum)	2 b	(10)	82 b	(11)
$G_{\rm th}$	Thermal neutron self-shielding factor	Table 3	(12)	≅ 1 – 4/3 R∑ _a	(4)
$G'_{\rm res}$	Resonance neutron self-shielding factor	Table 3	(12)	Fig. 1 ^F	
g	Correction factor which describes the departure of the cross section from 1/v law in thermal region	1.0	(2)	See Table 4	(2)

A The numbers in parenthesis following given values are the uncertainty in the last digit(s) of the value; 0.729 (8) means 0.729 ± 0.008, 70.8(1) means 70.8 ± 0.1

perturbation of the field by the cadmium; (2) the inexact cadmium cut-off energy; (3) the low melting temperature of cadmium. In addition, the reactivity changes accompanying the rapid insertion and removal of cadmium may prohibit the use of the cadmium-ratio method. However, the self-shielding corrections remain important unless the concentrations of cobalt and silver are small. Studies indicate that the accuracy of the two-reaction method for determination of thermal neutron fluence is comparable to the cadmium-ratio method (14).

3.3 The long half-lives of the two monitors permit the determination of fluence for long-term monitoring.

4. Apparatus

- 4.1 *NaI(Tl) or Germanium Gamma-Ray Spectrometer* (using a multichannel analyzer)—For the NaI(Tl) technique and the germanium technique, see Test Methods E181.
 - 4.2 Precision Balance.
 - 4.3 Digital Computer.

5. Materials and Manufacture

- 5.1 The two monitors required for this test method are cobalt and silver. Although these two materials are available commercially in very pure form, they have been used (15) alloyed with aluminum (\leq 1 % cobalt and \leq 1 % silver) to minimize the self-shielding effect and to permit insertion into a high thermal-neutron fluence rate ($>10^{15}$ cm⁻²s⁻¹) facility (6, 16). Typical alloys contain 0.1 % silver or cobalt in aluminum) see 6.1 and 8.1).
- 5.2 The uncertainties and nonuniformity of alloy concentrations must be established by one or more different test methods. These might include chemical and activation analysis, or spectrometry. The purity of the aluminum matrix should also be established.
- 5.3 Whenever possible, the alloys should be tested for interfering impurities by neutron activation.

5.4 The method of encapsulating the monitors for irradiation depends upon the characteristics of the facility in which the measurements are to be made. The monitors have essentially the same chemical characteristics as pure aluminum; therefore, an environment in which aluminum would not be adversely affected would be generally satisfactory for the alloys. However, the low mechanical strength of the monitors requires in many instances that it be encapsulated or shielded from physical disturbances by some type of container. Aluminum cans or tubing are satisfactory for many cases of interest, but for hostile environments, stainless steel or vanadium may be preferable. Perturbation due to the presence of the container must be accounted for, especially in the case of stainless steel. The container should be constructed in such a manner that it will not create a significant flux perturbation and that it may be opened easily, especially if the monitors must be removed remotely.

6. Westcott Neutron Fluence Convention

6.1 The Westcott neutron fluence convention is designed primarily for calculations involving reactions rather than those involving scattering or diffusion. It states that the reaction rate per atom present, R, is equal to the product of an effective cross section, $\hat{\sigma}$, with the Westcott neutron fluence $\phi_w = nv_0$, where n = the neutron density, including both thermal and epithermal neutrons, cm⁻³, and $v_0 = 2200$ m/s.

Thus:

$$R = \varphi_w \hat{\sigma} = n v_0 \hat{\sigma} \tag{1}$$

The true equation for reaction rate is given by the equation:

$$R = \int_0^\infty n(v) v \sigma(v) dv \tag{2}$$

where:

n(v) = neutron density per unit velocity,

v = neutron velocity, and

 $\sigma(v)$ = cross section for neutrons of velocity v.

 $^{^{}B}$ A 2200 m/s cross section (E = 0.0253 eV, T = 20°C) was taken from the sources indicated in Ref (9).

^C Cross section uncertainty data is taken from Ref (7), the cross section comes from the other reference.

^D Calculated using Eq 10.

E Cross section uncertainty comes from covariance data provided in the cross section source. The other reference indicates the source of the cross section.

F In Fig. 1, $\Theta = 4E_r kT/A\Gamma^2 = 0.2$ corresponds to the value for ¹⁰⁹Ag for T = 293 K, $\Sigma_r = N_0 \sigma_{r, max} = 29999$ barn at 5.19 eV (13).

Therefore, the effective cross section is defined by the equation:

$$\hat{\sigma} = \int_0^\infty n(v) v \sigma(v) dv / n v_0 \tag{3}$$

The neutron spectrum assumed by Westcott has the form: $n(v) = n(1 - f)P_{\rm m}(v) + nfP_{\rm e}(v)$, where $P_{\rm m}$ and $P_{\rm e}$ are the Maxwellian and epithermal density distribution functions normalized so that: $\int_0^\infty P_m(v) dv = \int_0^\infty P_c(v) dv = 1$. The quantity f is the fraction of the total density, n, in the epithermal distribution. The epithermal distribution is assumed proportional to 1/E per unit energy interval. This distribution is terminated by a cut-off function at a suitable lower limit of energy. Based on the above spectrum, one obtains the following relation for the effective cross section:

$$\hat{\sigma} = \sigma_0(g + rs) \tag{4}$$

where:

= cross section of 2200 m/s neutrons,

= a measure of the departure of the cross section from 1/vdependence in the thermal region,

= $S_0 \sqrt{T/T_0}$, a factor which describes the departure of the cross section from the 1/v law in the epithermal region, including resonance effects, and

= a measure of the proportion of epithermal neutrons in the reactor spectrum.

More specifically:

$$r = f\sqrt{\pi\mu_{\rm n}}/4\tag{5}$$

where:

= fraction of the total density in the epithermal distribution, and

= a factor chosen to give the proper normalization to the epithermal density distribution. A suitable factor for water moderated systems is 5 (2).

6.2 Limitation of the Westcott Convention-Sufficient conditions for the applications of the Westcott convention are that:

$$\sum_{a}/\xi \sum_{s} < 0.1 \tag{6}$$

and:

$$T/T_{\rm m} < 1.07$$
 (7)

where:

 $\Sigma_{\rm a}$ = macroscopic absorption cross section averaged over all materials affecting spectrum,

= average logarithmic energy decrement per collision,

= macroscopic scattering cross section averaged over all materials affecting spectrum,

= neutron temperature, K, and

 $T_{\rm m}$ = temperature of the moderator, K.

If as a result of neutron captures (for example, in the fuel) the quantity $\sum_a/\xi\sum_s$ becomes too great or if the neutron temperature T is too great relative to the moderator temperature $T_{\rm m}$, the Maxwell spectrum hypothesis fails and the true spectrum must be calculated and the effective cross section determined with this spectrum.

6.3 The conventional 2200 m/s thermal neutron-fluence rate, φ_0 , and the epithermal fluence-rate parameter, φ_e , as defined in Test Method E262, can be obtained from the Westcott neutron-fluence rate, φ_w , and the Westcott epithermal index, $r\sqrt{T/T_0}$, by means of equations Eq 8 and Eq 9:

$$\varphi_0 = \left(1 - \frac{4 \, r}{\sqrt{\pi \mu_n}}\right) \varphi_w \tag{8}$$

$$\varphi_e = \frac{2}{\sqrt{\pi}} r \sqrt{\frac{T}{T_0}} \varphi_w \tag{9}$$

6.4 In Eq 8, it is necessary to estimate the neutron temperature, T, in order to obtain the value of r from the index $r\sqrt{T/T_0}$. Provided inequality (Eq 7) is satisfied, only slight error is introduced by assuming $T = T_{\rm m}$, the moderator temperature.

6.5 Although the ${\rm Ag}^{109}({\rm n},\!\lambda){\rm Ag}^{110{\rm m}}$ S_0 value in Table 1 is a measured value, S_0 can be calculated by the following equa-

$$S_0 = \frac{2}{\sqrt{\pi}} \frac{I''_0}{\sigma_0} = \frac{2}{\sqrt{\pi}} \left(\frac{I_0}{\sigma_0} - 2g \sqrt{\frac{E_0}{E_{\rm Cd}}} \right)$$
 (10)

where:

= resonance integral excess over the 1/v cross section value, cm²,

= 2200 m/s cross-section value, cm²,

= resonance integral, $\int_{E_{cd}}^{\infty} \frac{\sigma(E)}{E} dE$ = 0.0253 eV, and

7. Procedure

7.1 Decide on the size and shape of the monitors to be irradiated, taking into consideration the size and shape of the irradiation space. The mass and exposure time are parameters which can be varied to obtain a desired disintegration rate for a given neutron fluence rate level. To facilitate the convergence of the two activity equations for the fluence rate and the epithermal index, the concentration of the alloys should be chosen so that the ratio of the disintegration rates is on the order of one.

7.2 Weigh the samples to a precision of $\pm 1.0 \%$ (1S %) as defined in Practice E177.

7.3 Irradiate the samples for the predetermined time period. Record the power level and any changes in power during the irradiation, the time at the beginning and end of the irradiation, and the relative position of the monitors in the irradiation facility.

7.4 A waiting period is necessary between termination of the exposure and start of counting when using Co-Al and Ag-Al monitors. This allows the 0.62356 days (17) half-life ²⁴Na which is formed by fast-neutron reactions on ²⁷Al or by thermal-neutron captures by ²³Na impurities to decay below levels at which its radiations may cause interferences. It is sometimes advisable to count the samples periodically and follow the decay of the portions of the activities due to the ²⁴Na. The length of the waiting period can be reduced by the use of a germanium detector.

7.5 With the gamma-ray spectrometer, analyze the silver sample for $^{110\rm m}{\rm Ag}$ and the cobalt sample for $^{60}{\rm Co}$. Obtain the net count rate in each full-energy gamma-ray peak of interest, that is, 657.7623 keV or 884.684 keV for $^{110\rm m}{\rm Ag}$, 1332.501 keV for $^{60}{\rm Co}$ (see Test Methods E181). See Table 2 for gamma radiations of $^{110\rm m}{\rm Ag}$.

8. Calculation

- 8.1 Calculate the activities of ^{110m}Ag and ⁶⁰Co in disintegrations per second.
- 8.2 A Westcott 2200 m/s neutron fluence rate, nv_0 , or φ_w and the Westcott epithermal index parameter, $r\sqrt{T/T_0}$ are related to the measured activities of the silver and cobalt monitors by the following equation:

$$A = N_0 \lambda BFG \hat{\sigma}_1 \varphi_w t_i \tag{11}$$

where:

A = measured activity at the end of the exposure time, disintegrations/s,

 N_0 = number of target atoms of ⁵⁹Co or ¹⁰⁹Ag at start of irradiation,

 λ = disintegration constant of product nuclide, s⁻¹,

B = Self-absorption factor of the decay gamma ray in the monitor material,

F = burnup and decay correction factor,

G = self-shielding factor (see Eq 15, Table 3 and Fig. 1).

 $\hat{\sigma}_1$ = Westcott's effective absorption cross section for production of the product nuclide, cm²,

 φ_w = a 2200 m/s neutron fluence rate in which *n* is the neutron density (including both thermal and epithermal neutrons) and v_0 is 2200 m/s, and

 t_i = exposure time.

8.3 The self-absorption factor, if not known for the gamma rays being measured, can be approximated by the following equation (20):

$$B \approx 1 - (4/3)(\mu_{\circ}R)$$
 (12)

where:

 $\mu_{\rm a} = \text{linear absorption coefficient in monitor, cm}^{-1}(21), and$ R = radius of monitor wire, cm.

8.4 The burnup and decay correction factor is given by:

TABLE 2 Gamma Radiations of ^{110m}Ag (18)^A

		3(-)	
Energy of Gamma ^B (keV)		Intensity ^B (%)	
1.	657.7600 (11)	95.61	
2.	884.6781 (13)	75.00 (11)	
3.	937.485 (3)	35.0 (3)	
4.	1384.2931 (20)	25.1 (5)	
5.	763.9424 (17)	22.60 (7)	
6.	706.6760 (15)	16.69 (7)	
7.	1505.0280 (20)	13.33 (15)	
8.	677.6217 (12)	10.70 (5)	
9.	818.0244 (18)	7.43 (4)	
10.	687.0091 (18)	6.53 (3)	
11.	744.2753 (18)	4.77 (3)	
12.	1562.294 (18)	1.22 (3)	

^A The number of parentheses following some given values is the uncertainty in the last digit(s) of the value: 0.729 (8) means 0.729 ± 0.008 , 80.8 (1) means 70.8 ± 0.1 . ^BSee Ref (17) for an alternate source of data.

TABLE 3 Self-Shielding Factors for Cobalt Wires (12)

		-		. ,
	Wire Diameter in. (mm)	Cobalt Content, (mass %)	<i>G</i> ' _{res} (132 eV)	G_{th}
_	0.050 (1.27)	0.104	1.00	1.00
	0.050 (1.27)	0.976	0.95 ± 0.04	0.99 ± 0.01
	0.001 (0.03)	100	0.81 ± 0.03	0.99 ± 0.02
	0.005 (0.13)	100	0.52 ± 0.02	0.97 ± 0.01
	0.010 (0.25)	100	0.42 ± 0.02	0.94 ± 0.01
	0.015 (0.38)	100	0.38 ± 0.01	0.92 ± 0.02
	0.020 (0.51)	100	0.34 ± 0.01	0.90 ± 0.02
	0.025 (0.64)	100	0.32 ± 0.01	0.88 ± 0.03

$$F = \frac{exp(-\hat{\sigma}_a \varphi_w t_i) - exp(-(\lambda + \hat{\sigma}_2 \varphi_w) t_i)}{(\lambda + \hat{\sigma}_2 \varphi_w - \hat{\sigma}_a \varphi_w) t_i}$$
(13)

where:

 $\hat{\sigma}_a$ = Westcott's effective absorption cross section for target nuclide, cm², and

 $\hat{\sigma}_2$ = Westcott's effective absorption cross section for the product nuclide, cm².

8.5 The self-shielding factor is given by:

$$G = \frac{gG_{\text{th}} + \left(r\sqrt{T/T_0}\right)S_0G'_{\text{res}}}{g + \left(r\sqrt{T/T_0}\right)S_0}$$
(14)

where:

g = correction factor which describes the departure of the cross section from the 1/v law in the thermal region (see Table 4 for silver "g" factors),

 G_{th} = thermal neutron self-shielding factor, G'_{res} = resonance neutron self-shielding factor,

r = a measure of the proportion of epithermal neutrons in the reactor spectrum,

T = neutron temperature, K,

 $T_0 = 293.6 \text{ K}, \text{ and}$

 S_0 = correction factor which describes the departure of the cross section from the 1/v law in the epithermal region.

- 8.6 Substituting the measured activities of the cobalt and the silver monitors into Eq 12 yields two nonlinear equations in the two unknown parameters $r\sqrt{T/T_0}$ and ϕ_w .
- 8.7 PC software products such as *MathCad* and *Mathematica* can be programmed to solve these two nonlinear equations with a variety of iterative solvers. A FORTRAN IV computer program, COAG2 (22), was written to solve these equations. The program iterates until the epithermal index and the fluence values give calculated activities that are within 0.1 % of their measured values. The constants, cross sections, and other measured values used in the program should be set equal to those listed in Table 1.
- 8.8 If the burnup corrections are negligible, that is, if the factor F, (Eq 14) is equal to $(1-exp(-\lambda t_i))/\lambda t_i$, the equations can be solved in closed form. If the subscripts 1 and 2 refer to the two reactions (in either order) the parameters are calculated as follows:

$$\varphi_{w} = \left(\frac{R_{2}}{S_{02}G'_{rec2}\sigma_{02}} - \frac{R_{1}}{S_{01}G'_{rec1}\sigma_{01}}\right) / \left(\frac{g_{2}G_{th2}}{S_{02}G'_{rec2}} - \frac{g_{1}G_{th1}}{S_{01}G'_{rec1}}\right) (15)$$

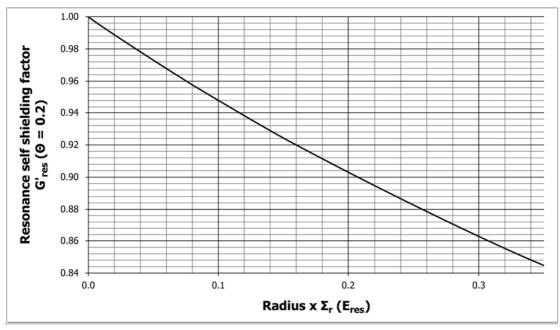


FIG. 1 Resonance Self-Shielding Factor for Cylinders (θ = 0.2) as a Function of Radius × Macroscopic Absorption Cross Section at the Peak of the Resonance, for ¹⁰⁹Ag at 5.19 eV, $\sigma_{\gamma,max}$ = 29999 barn (13). Calculations used shielding curve for cylinders from (19). To convert σ_{r} to Σ_{r} , use Σ_{r} = $N_{0}\sigma_{r}$, where N_{0} is the number density of ¹⁰⁹Ag atoms in the wire.

TABLE 4 "g" Factor for Silver

TABLE 4 "g" Factor for Silver		
T (°C)	g (Ag)	
20	1.0044	
40	1.0053	
60	1.0062	
80	1.0071	
100	1.0080	
120	1.0090	
140	1.0099	
160	1.0108	
180	1.0117	
200	1.0126	
220	1.0136	
240	1.0145	
260	1.0154	
280	1.0164	
300	1.0173	
330	1.0187	
360	1.0201	
390	1.0215	
420	1.0230	
450	1.0244	
480	1.0258	
510	1.0273	
540	1.0287	
570	1.0302	
600	1.0316	
640	1.0336	
680	1.0356	
720	1.0376	
760	1.0395	
800	1.0416	
840	1.0436	
880	1.0456	
920	1.0476	
960	1.0497	
1000	1.0517	
1060	1.0549	
1120	1.0580	
1180	1.0612	
1240	1.0644	
1300	1.0676	

$$r\sqrt{\frac{T}{T_0}} = \frac{g_1 G_{th1}}{S_{01} G'_{res1}} \frac{g_2 G_{th2}}{S_{02} G'_{res2}} \left(\frac{R_1}{g_1 G_{th1} \sigma_{01}} - \frac{R_2}{g_2 G_{th2} \sigma_{02}}\right) / \left(\frac{R_2}{S_{02} G'_{res2} \sigma_{02}} - \frac{R_1}{S_{01} G'_{res1} \sigma_{01}}\right)$$

$$(16)$$

where:

 R_1 and R_2 = the measured reaction rates per target atom = $A/N_0\lambda BFt_i$

9. Precision and Bias

Note 2—Measurement uncertainty is described by a precision and bias statement in this standard. Another acceptable approach is to use Type A and B uncertainty components (23, 24). This Type A/B uncertainty specification is now used in International Organization for Standardization (ISO) standards and this approach can be expected to play a more prominent role in future uncertainty analyses.

- 9.1 There are several sources of errors that affect the precision of this test method. Random errors appear in the determination of the weight percent and homogeneity of low concentrations of cobalt and silver alloyed with some other material such as aluminum, the determination of the cobalt and silver activities after irradiation, and the measurement of cross sections and other related data. These errors should not exceed $\pm 3\%$ (1 $\sigma\%$), $\pm 3\%$ (1 $\sigma\%$), and $\pm 5\%$ (1 $\sigma\%$) as defined in Practice E177.
- 9.2 The bias due to trace impurities was calculated to be negligible. Self-shielding and the burnup of the product nuclide are accounted for in the mathematical treatment. Self-shielding may become appreciable unless care is taken to minimize this effect by the use of low concentrations of cobalt and silver in the alloy used, as very thin foils. The burnout of the product nuclide is a very small effect unless the cross section is greatly in error.

10. Keywords

10.1 activation; cobalt; dosimetry; foil; silver; thermal neutron

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