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DIRECT MEASUREMENT OF k_o FOR MONITOR NEUTRON ACTIVATION ANALYSIS

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This paper introduces the thermal column or the cold neutron guide beam of the 20 MW NBSR at the National Institute of Standards and Technology as a direct k_o measurement facility. Measurement of k_o at this facility not only produces accurate values, but avoids the additional correction factors needed in other measurement methods. The k_o of Sb, Ag and Cr with respect to Sc as monitor have been measured and their values are comparable to values based on tabulated nuclear constants and to those measured by other researchers.

Central to the application of monitor neutron activation analysis are accurate values for the quantity k_0. By definition, k_0 is a compound constant whose composite nuclear constants can be obtained from nuclear data tables. Unfortunately, the k_0's calculated from the tabulated values of the component constants do not always agree with the measured values of k_0 within their uncertainty. What is more, the measured values sometimes disagree from one measurement facility to another. The problems posed by this situation can be seen by comparing the compilation of k_0 by DeCorte et al[1], Kafala[2] and earlier k_0 values of a few elements by Ahmad[3]. We feel that the problem is fundamental and the solution should not be too complicated, otherwise the trust in k_0 values and their future uses will continue to be questioned[4].

The k_0 of a nuclide x with respect to a monitor C is given by:

$$k_{0,C}(x) = \frac{M_C \theta_x \sigma_{0,x} \gamma_x}{M_x \theta_C \sigma_{0,C} \gamma_C} \tag{1}$$

where M is the atomic weight, θ is the isotopic abundance, σ_0 is the 2200 m/s activation cross section, and γ is the gamma abundance. When a measurement is carried out in a neutron field, $\Phi(E)$,

$$k_C(x) = \frac{M_C \theta_x \gamma_x \int_0^\infty \sigma_x(E)\Phi(E)dE}{M_x \theta_C \gamma_C \int_0^\infty \sigma_C(E)\Phi(E)dE} \tag{2}$$

is obtained. Experimentally we have:

$$k_C(x) = \frac{(A_0 / m\varepsilon_p)_x}{(A_0 / m\varepsilon_p)_C} \tag{3}$$

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Table 1: Comparison of methods in k_0 measurement.

Method	Equation	Remark
Based on Tabulated Nuclear Consts.	$k_{0,c}(x) = \frac{(\theta\gamma\sigma_0/M)_x}{(\theta\gamma\sigma_0/M)_c}$	Readily Available But Uncertainty May be > 5%
Normalized for Experimental Conditions	$k_{0,c}(x) = \frac{(A_{sp}/\varepsilon_p)_x / [G_{th}f + G_e Q_0(\alpha)]_x}{(A_{sp}/\varepsilon_p)_c / [G_{th}f + G_e Q_0(\alpha)]_c}$	Both of these Techniques are Laborious and Measurement Errors Accumulate.
Cd-Subtraction Technique	$k_{0,c}(x) = \frac{\{[A_{sp} - (A_{sp,cd})/F_{cd}]/(\varepsilon_p G_{th})\}_x}{\{[A_{sp} - (A_{sp,cd})/F_{cd}]/(\varepsilon_p G_{th})\}_c}$	
Direct Method	$k_{0,c}(x) = \frac{(A_{sp}/\varepsilon_p)_x}{(A_{sp}/\varepsilon_p)_c}$ In a pure thermal neutron spectrum.	Very Easy, Reliable and Uncertainty Could be <1%

A_{sp} = A_0/m
m = Elemental mass.

$$A_0 = \frac{\lambda C e^{\lambda t_1}}{(1 - e^{-\lambda\Delta})(1 - e^{-\lambda\tau})}$$

λ = Decay constant.
τ = Irradiation time.
t_1 = Time from end of irradiation to start of count.
Δ = Live time of counting.
C = Net counts in photopeak.
f = Subcadmium (thermal)-to-epithermal neutron flux ratio ($f = \Phi_s/\Phi_e$).
Φ_s = conventional subcadmium (thermal) neutron flux.
Φ_e = conventional epithermal neutron flux.
F_{Cd} = Correction factor for Cd-transmission of epithermal neutrons.
G_e = Correction factor for epithermal neutron self shielding.
G_{th} = Correction factor for thermal neutron self shielding.
M = Atomic weight.
Q_0 = Resonance integral (1/E) to 2200 m/s cross-secction ratio. ($Q_0 = I_0/\sigma_0$)
α = Parameter describing the $\Phi_e(E)$ as $1/E^{1+\alpha}$ neutron flux distribution.
ε_p = Full energy peak detection efficiency.
Cd = Subscript, related to irradiation under Cd-cover.
γ = Gamma abundance.
θ = Isotopic abundance.
σ_0 = 2200 m/s cross-section

where m is the elemental mass, ε_p is the full energy peak detector efficiency and A_0 is as defined below.

This measured $k_C(x)$ will be equal to $k_{0,C}(x)$ under two conditions: 1) whenever $\sigma_x(E)$ and $\sigma_C(E)$ have the same shape, e.g. 1/v, or 2) whenever the measurement is made in a spectrum with no significant epithermal neutrons. For nuclides that satisfy the first

condition, selecting nuclear constants from tables would have been enough except for the problems caused by compounding of errors. Therefore, the k_0 of all nuclides must be measured.

Two methods have been used in the compilation of k_0 by DeCorte[1]. In the first method, normalization for experimental conditions is done by correcting for epithermal reactions and for non-1/v behavior. In the second method, the Cd-subtraction technique, the epithermal reaction is accounted for by irradiating monitors with and without cadmium cover. The complications in these two methods would be eliminated if the measurement were done in a strongly thermalized neutron spectrum which satisfy the second condition stated above. A comparison of the methods in k_0 measurements is shown in table 1. Recommendation of a special facility for the direct measurement of k_0 is the object of this paper.

The Direct Method

In our direct method, we have used the thermal column of the 20 MW NBSR at the National Institute of Standards and Technology which satisfies the second condition stated above, since the cadmium ratio for gold[8] exceeds 1000. Therefore the correction factors, namely, f, Q, and F_{cd} in the methods listed in table 1 become unnecessary. This is true because as $\Phi(E)$ approaches totally thermalized flux, $\Phi_{th}(E,T)$, equation (2) becomes:

$$k_C(x) = \frac{M_C \theta_x \gamma_x \sigma_{0,x} g_x(T)}{M_x \theta_C \gamma_C \sigma_{0,C} g_C(T)} \tag{4}$$

where g(T) is the non-1/v factor at neutron temperature T. In the limit that the neutron energy approaches zero, equation (4) becomes:

$k_C(x) = \frac{M_C \theta_x \gamma_x \sigma_{0,x}}{M_x \theta_C \gamma_C \sigma_{0,C}}$ which is the desired $k_{0,C}(x)$. This statement is valid, in general, since,

$\sigma(E) \to \sigma_0 \sqrt{\frac{E_0}{E}}$ and $g(T) \to 1$, for all nuclides in a field where $E \to 0$

Experimentally, therefore, $k_{0,C}(x) = \frac{(A_0 / m\varepsilon_p)_x}{(A_0 / m\varepsilon_p)_C}$ in a pure thermal neutron spectrum.

Unlike other k_0 compilations in which gold is chosen as the monitor, we have chosen scandium as the monitor. Scandium was chosen as the monitor because it is readily activated by thermal neutrons and its activation product has a relatively long half life and radiates high energy gamma rays. But most importantly, the ratio of

Table 2: Nuclear data of interest in monitor neutron activation analysis of some selected elements. (Cross sections are taken from ref. 5, half life, gamma energy and abundances from ref. 7)

Element	Atomic mass	Target isotope	Isotopic abundance % (% error)	σ_0, b (% error)	I, b (% error)	$Q_0 = I/\sigma_0$	Isotope formed	E_r, keV (% error)	Half life (% error)	Main gamma keV	Gamma abundance% (% error)
Ytterbium	173.04	^{174}Yb	31.83 (0.01)	69.4 (7.2)	27.1 (1.0)	0.39	^{175}Yb	602 (8.0)	4.19d (0.2)	396.3	6.5 (12)
Scandium	44.96	^{45}Sc	100	27.2 (0.7)	12 (4.2)	0.44	^{46}Sc	5130 (17.0)	83.82d (0.02)	889	99.98 (0.02)
Chromium	52	^{50}Cr	4.35 (0.2)	15.9 (1.3)	7.8 (5.1)	0.49	^{51}Cr	7530 (11.0)	27.69d (0.04)	320.1	9.85 (0.9)
Gold	196.97	^{197}Au	100	98.65 (0.09)	1550 (1.8)	15.71	^{198}Au	5.65 (7.1)	2.695d (0.1)	411.8	95.56 (0.1)
Silver	107.87	^{109}Ag	48.17 (0.01)	4.7 (4.3)	72.3 (5.5)	15.38	^{110m}Ag	6.08 (1.0)	249.76d (0.01)	657.8	94.51 (0.1)
Antimony	121.75	^{121}Sb	57.3 (1.6)	5.9 (3.4)	200 (10.0)	33.90	^{122}Sb	13.1 (3.8)	2.7d (1.1)	564.1	70.55 (0.5)
Antimony	121.75	^{123}Sb	42.7 (2.1)	4.14 (2.4)	125 (16)	30.19	^{124}Sb	28.2 (6.4)	60.2d (0.05)	602.7	97.89 (0.05)
Uranium	238.03	^{238}U	99.27 (0.02)	2.68 (0.7)	277 (1.1)	103.36	^{239}Np	16.9 (1.2)	2.355d (0.2)	277.6	14.2 (1.4)

its resonance integral to thermal cross section, as shown in table 2, is approximately equal to what one expects of a 1/v cross section as illustrated with equation (5).
For a 1/v cross section,

$$\frac{I_0}{\sigma_0} = 2\sqrt{\frac{E_0}{E_{Cd}}} = 0.45 \qquad (5)$$

where

$$I_0 = \int_{E_{Cd}}^{\infty} \frac{\sigma(E)dE}{E}, \quad \sigma(E) = \sigma_0\sqrt{\frac{E_0}{E}}, \quad E_0 = 0.0253\,eV \quad \text{and} \quad E_{Cd} = 0.5\,eV.$$

Three binary solutions Sb-Sc, Ag-Sc and Cr-Sc were made starting with high purity materials (>99.99%). It must be mentioned that the stability of the mass ratios in these binary solutions are crucial. Certifying those masses and monitoring their stability over a long period of time poses the same problem as encountered with multi-element standards. The goal of monitor neutron activation analysis is the reduction of the number of such standards necessary in a single irradiation. In our experience, it is difficult to prepare primary solution standards of some of these elements starting with their pure metals.

Binary solutions were pipetted onto 0.25 inch diameter discs of ashless filter paper. Each filter paper has approximately 3μg of Sc along with an amount of the second element to give equivalent count rates. These filter papers were dried and sealed in 1 mil polyethylene. Duplicate packages placed in each rabbit, one at the

bottom and the other at the top, were separated by foam. The rabbits were irradiated in the thermal column, RT5 of NBSR. Following a reasonable cooling time the monitors were counted at a distance of seven inches from a high purity germanium detector.

The net areas under the peaks of the major gamma lines of the nuclides, ^{46}Sc, ^{122}Sb, ^{124}Sb, ^{51}Cr and ^{110m}Ag were determined using the SUM program[6]. The SUM program treats the background problems encountered with net area evaluation and its subsequent error analysis more consistently than other net area determination programs.

Table 3: Comparison of k_0 values of some nuclides.

Product Nuclide	E_γ (KeV)	$k_{0,Sc}$ (error %)			
		This Work	DeCorte[1]	Kafala[2]	From Table[5]
^{124}Sb	602.7	0.0225 (1.71)	0.0243 (0.6)	0.0238 (2.1)	0.0236
^{122}Sb	564.1	0.0332 (1.57)	0.0359 (1.5)	0.0392 (4.3)	0.0326
^{110m}Ag	657.8	0.0287 (1.39)	0.0282 (0.6)	No Data	0.0329
^{51}Cr	320.1	0.00216 (3.69)	0.00215 (0.5)	0.00212 (1.3)	0.00216

Results

The results are shown in table 3 along with values from other k_0 compilations. It must be mentioned that since the other k_0's have been measured with respect to gold as monitor, we have converted these values to those of scandium as monitor to facilitate easy comparison.

Discussion of Results

Our interests in the direct method are two-fold. Firstly, it will yield accurate values for k_0 without additional corrections. Secondly, once such simple k_0 measurements are made, they can be used in other facilities along with the shape independent model, introduced below, to calibrate the facility.

As shown in figure 1, the $k_{0,C}$ is measured at the specialized facility. k_C's are measured at the user's irradiation facility at several positions where epithermal reactions are not negligible. Therefore, k_C's are much higher than k_0 especially for Sb which has a higher ratio of resonance to thermal cross section than Sc, a 1/v monitor. In the shape-independent method two monitors are used, Sc to monitor the thermal spectrum and Sb to monitor the epithermal spectrum. A plot of the k_{Sc}'s of an unknown nuclide x, e.g. silver as in figure 1,

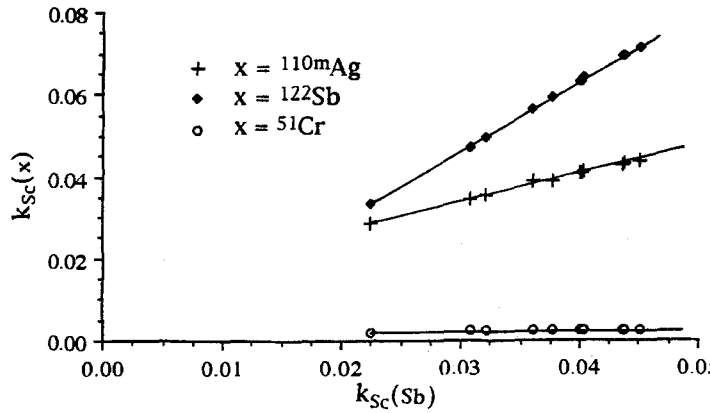

Figure 1: A plot of $k_{Sc}(x)$ versus $k_{Sc}(Sb)$ of the measurements made at Ford Nuclear Reactor. The lowest k_{Sc} in this figure is $k_{0,Sc}$ and its measurement was made at NBSR.

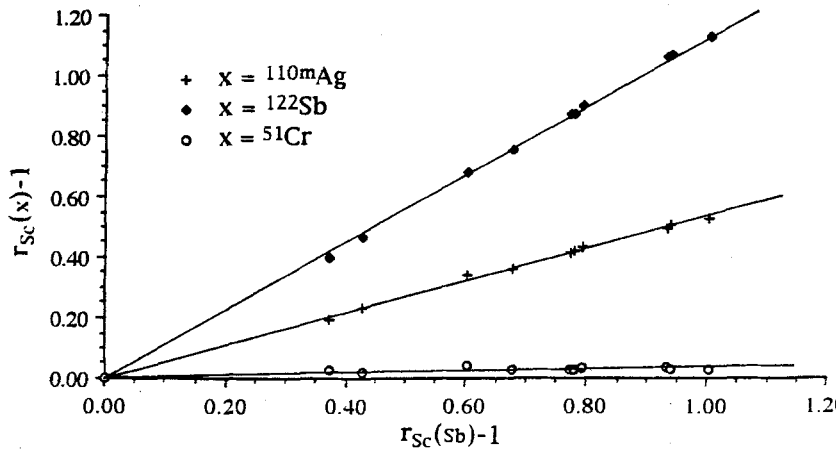

Figure 2: Normalized $k_{Sc}(x)$ versus $k_{Sc}(Sb)$ of the data plotted in figure 1.

versus the k_{Sc}'s of Sb, results in a straight line. To deduce a unique parameter, B(x), from this line, that characterizes the irradiation facility and its counting system, the ratios are normalized with respect to k_0 as shown in figure 2. The slope of the line in figure 2, B(x), is given by equation (6):

$$B(x) = \frac{r_c(x) - 1}{r_c(Sb) - 1} \tag{6}$$

where

$$r_c(x) = \frac{k_c(x)}{k_{0,c}(x)} = \frac{A_0(x)/A_0(c)}{[A_0(x)/A_0(c)]_0}$$

The values of B(x) obtained from the normalized data are free of two important systematic errors. 1) errors due to efficiency of detectors cancel out and 2) errors due to monitor masses are eliminated, if the same binary solution is used to measure the k_0's as

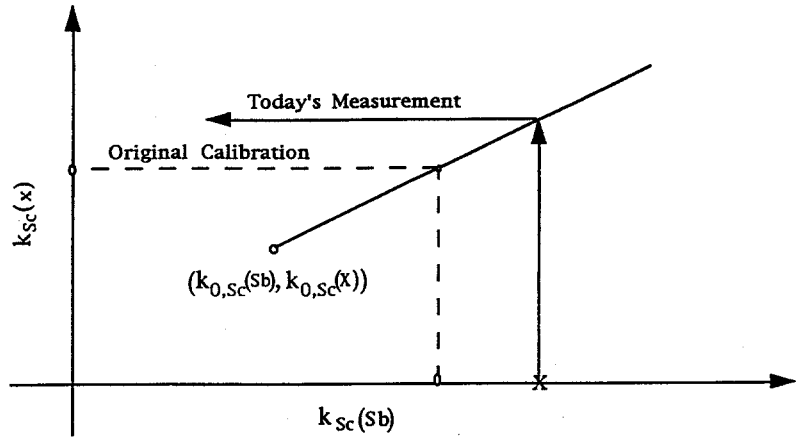

Figure 3: Illustration on How to Use the Shape-Independent Method.

was used to measure k_C's. One could obtain a preliminary estimate of the value of B(x) using the following expression which is based on an idealized neutron spectrum shape.

$$B(x) \approx \frac{Q_0(x) - Q_0(c)}{Q_0(sb) - Q_0(c)} \tag{7}$$

Once B(x) has been properly measured in the user's facility, then that facility is calibrated and ready for monitor neutron activation analysis, since we can write:

$$\frac{k_C(x)}{k_{0,C}(x)} - 1 = B(x)\left\{\frac{k_C(Sb)}{k_{0,C}(Sb)} - 1\right\} \tag{8}$$

and use Sc and Sb as the only standards in future activation analysis of nuclide x. Figure 3 illustrates how one obtains a new value of $k_{Sc}(x)$ based on a new measurement of $k_{Sc}(Sb)$.

*

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