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NUCLEAR DATA AND THE EFFECTS OF ITS INCONSISTENCY ON INSTRUMENTAL NEUTRON ACTIVATION ANALYSIS

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Abstract

In this study, we examine the role of nuclear data in Instrumental Neutron Activation Analysis (INAA) particularly as it affects determination of reactor flux parameters and application of comparator methods. The work reviewed the available sources of nuclear data, the variations that exist from one source to the other and the effects of such variations on INAA. Measurement of Neutron flux parameters in inner and outer irradiation channel of a miniature neutron source reactor was carried out using two independent nuclear data sources to investigate the effects of inconsistency of nuclear data on the precision of analytical result. The result obtained shows a slight variation of flux parameters with nuclear data source. It was also observed that modification of the earlier compiled basic nuclear data lead to inconsistencies in the secondary data that applies it.

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Introduction

The beginning of nuclear data compilation and evaluation dates back to 1935 when an Italian, Giorgio Fea (G. Fea 1935) published the first table of isotopes. Thereafter, information on nuclear data started appearing from all over the world and hence the formation of groups to analyze, organizes, and disseminates nuclear data information. Nuclear data sheets (Oak Ridge), Table of Isotopes and Isotope project (Berkeley), National Nuclear Data Center (Brookhaven) and many others are some of the fruits of these efforts (Firestone et. al., 1996, 1998, 1999). The exertion was later united into the international network of Nuclear Structure and Decay Data (NSDD) Evaluators under the auspices of the Nuclear Data Section of the International Atomic Energy Agency, (IAEA) Vienna.

The emergence of Internet has brought a wide range of nuclear data sources to the doorsteps of every interested party; however, the limited access to the facility and low level of computer literacy among developing countries made printed data sources still relevant and convenient. The consequence of these numerous data sources is that, the experimentalist was left with a larger option of the basic nuclear data from which to choose composite nuclear constants. In practice, they are usually determined by direct measurements (De Corte and Simonits 2003), partly because equivalent constants derived from the basic data are often discrepant. The objective of this work therefore, is to point out the gray areas to the users of nuclear data, particularly for those involved in Instrumental Neutron Activation Analysis (INAA).

The main fields of INAA application are analytical chemistry, geology, biology and the life and environmental science. Its accuracy, the virtual absence of matrix effects and the completely different physical basis when compared to other analytical techniques, make it particularly suitable for the certification of reference materials. Over the years, several efforts were made to perform standardization of the frequently used relative method of NAA (equation 1) to the absolute or single comparator method (Girardi et. al., 1965) in order to achieve an increased applicability to multi-elemental analysis and minimize human errors during experimental work. However, these efforts were hampered by the need for accurate and reliable data sets such as atomic mass, isotropic abundance, (n,γ) capture cross-section, the absolute γ-ray abundance and absolute neutron flux (equation 7).

To overcome this problem, the k_0 standardization method was launched in the 70's (Simonits et. al., 1975) with the aim of replacing the absolute nuclear data by *k*-factors, which are experimentally determinable nuclear constants (De Corte et. al., 1987). Firestone (2003), gave an overview of nuclear data and its various sources. Recently, De Corte and Simonits (2003) assembled compilations of recommended nuclear data for use in k_0-NAA into comprehensive tabulations.

Theoretical Consideration

For the determination of concentration of elements in a sample, either absolute (parametric), relative (using primary matrix reference material or synthetic element standard) or single-comparator (k_0-standardization) method is used. In the relative standardization method, a chemical standard (index 's') with a known mass 'w' of the element is co-irradiated with the sample of known mass 'W' and both are counted in the same geometrical arrangements with respect to the detector. The equation for the concentration of element in a sample using relative method is therefore given as:

$$C = \frac{\left(\frac{Np}{DCWt_m}\right)_a}{\left(\frac{N_p}{DCwt_m}\right)_s} \qquad (1)$$

where $D = \exp(-\lambda t_d)$ and $C = 1 - \exp(-\lambda t_m)$

N_p = net number of counts in the full-energy peak corrected for pulse losses.

Equation (1) is valid provided that the neutron flux gradient between the sample and standard position in the irradiation container is negligible.

The k_0-standardization method is based on co-irradiation of sample and a neutron fluence rate monitor like gold and on using an experimentally determined composite nuclear constant $k_{0,Au}$. The concentration of an element in a sample using k_0-method is given by:

$$C(ppm) = \left(\frac{I_a}{I_s} \cdot \frac{1}{k_{0,Au(a)}} \cdot \frac{f + Q_{0,s}(\alpha)}{f + Q_{0,a}(\alpha)} \cdot \frac{\varepsilon_{p,s}}{\varepsilon_{p,a}}\right) \qquad (2)$$

$$\text{where} \quad I = \frac{N_p}{t_m SDC} \qquad (3)$$

'a' = analyte; 's' = standard e.g Gold (Au)

$$S = 1 - \exp(-\lambda t_{irr})$$

$$\text{with} \quad \lambda = \frac{LN2}{T_{1/2}} \tag{4}$$

$$Q_0(\alpha) = \frac{Q_0 - 0.429}{(E_r)^{\alpha}} + \frac{0.429}{(2\alpha + 1)(E_{Cd})^{\alpha}} \tag{5}$$

$$\text{with} \quad Q_0 = \frac{I_0}{\sigma_0} \tag{6}$$

$$k_{0,Au(a)} = \frac{M_{Au}\theta_a\sigma_{0,a}\gamma_a}{M_a\theta_{Au}\sigma_{0,Au}\gamma_{Au}} \tag{7}$$

To determine flux parameters in an irradiation channel, there is need to measure cadmium ratio of the monitor nuclide (Ahmed 2004). The equation that relates the two parameters is given by:

$$(R_{Cd} - 1)Q_0(\alpha) = f \tag{8}$$

also $$f = \frac{\phi_{th}}{\phi_{epi}} \tag{9}$$

with ϕ_{th} = thermal flux

ϕ_{epi} = epithermal flux

M = molar mass, W = sample mass

θ = relative isotopic abundance of the target isotope

γ = absolute gamma-intensity (emission probability) of the particular radioisotope.

t_c = counting time, tm = measuring time, td = decay time

tirr = irradiation time

α = measure for the deviation of the epithermal neutron fluence rate distribution from the 1/E shape.

ε_p = full-energy peak detector efficiency

E_{cd} = cadmium cut-off energy =0.55 eV

f = the thermal to epithermal neutron fluence rate ratio, I_0 = resonance integral

σ_0 = activation cross-section for neutrons with energy of 0.025 eV

Sources of Nuclear Data

It is not possible to list in this paper all the sources of nuclear data that are in existence as many laboratories and authors have published and are still publishing and updating nuclear information. However, effort is made to highlight the popular ones among them, which could be classified into Internet and Printed types.

Internet Nuclear Data

1. IAEA Nuclear Data Services (Vienna) http://www-nds.iaea.org/
2. IAEA Technical and Scientific Reports http://www.iaea.org.at/progammes/ripc/nd/publications.htm
3. The k_0-Website, Technical University of Delft. http://iri.tudelf.nl/~rc/fmr/k0www3/mainframes3.htm
4. LBNL Isotope project nuclear dissemination home page (Berkeley) http://ie.lbl.gov/toi.html
5. Gamma-ray Spectroscopy Centre (Idaho falls) http://id.inel.gov/gamma/
6. National Nuclear Data Center (Brookhaven) http://www.nndc.bnl.gov/
7. National Institute for Standards and Technology (NIST) http://physics.nist.gov/physRefData/contents.html
8. Nuclear Data for Charged Particle Activation (Belgium) http://allserv.rug.ac.be/kstrykmn/nuclear/#Q-value
9. T2 Nuclear Information Service (Los Alamos) http://t2.lanl.gov/data/data.html
10. TUNL Nuclear Data Evaluation Group (North Carolina) http://www.tunl.duke.edu/nucldata

Printed Nuclear Data

1. IAEA Technical and Scientific Documents (IAEA Vienna)
2. The gamma-rays of the radionuclides (Erdtmann and Soyka, 1979)
3. Table of Isotopes (Firestone et.al., 1996)
4. Table of Radioactive Isotopes (Browne et.al., 1986)
5. Nuclear Data sheets (ENSDF) NDS Journal
6. Energy levels of light Nuclei (Nuclear Physics A Journal)
7. Nuclear Wallet cards (Tuli 2000)
8. Charts of the nuclides

9. The k_0-consistent IRI Gamma-ray catalogue for INAA (Blauuw, 1998)
10. Recommended nuclear data for use in the k0-standardization of NAA (De Corte and Simonits, 2003).

Variations in Nuclear Data

The primary source of inconsistency is the thermal cross-section, which is usually determined from the activation method with the introduction of data for the half-life, isotopic abundance and the gamma-ray intensity of the capturing isotope. However, the literature values of these nuclear data are not always the same (Ahmed 2005). The following are examples of typical variations usually found in literature:

Decay Schemes

Comparison of ^{75}Se data from the k_0-Website with that of Decay Data Evaluation Project (Browne and Schonfeld 2004) reveals that the former has no uncertainties and the gamma transition probabilities often disagree (Table 1). However, it contains cross-section information not included in the latter.

Cross-section

A publication by F. Decorte in: Nuclear Data in Science and Technology, Tsukuba, 2001 (http://wwwndc.tokai.jaeri.go.jp/nd2001/) reveals a remarkable case of inconsistency in the thermal cross-section value of Yb-175 (m+g) as reported by different authors between 1981 and 2003 (Figure 1). Similarly, a review of the Fe-59 cross-section showed that the abundance of this nuclide used to be 0.32% with a cross-section of 1.15 barns (Wagemans et. al., 2002). But after 1970 the abundance was adjusted to 0.282% without corresponding increase in cross-section values. This discrepancy let to thermal fluxes measured with Fe-59 being inconsistent with those derived from other reactions (De Corte and De Wispelaere, 2003).

Isotopic Abundance

The recommended k_0 tables are usually obtain from the analysis of only one isotope of each element thereby making isotopic abundance an implicit term. It is therefore expected of users to account for the uncertainty in the natural abundance of the material analyzed. Examples of some visible problems could be seen in:

* ^{36}Se- the reported abundance has 50% uncertainty (Firestone, 2003) because the natural abundance varies widely.

* ^{234}U- its actual abundance in any sample can vary widely because of the mobility of ^{234}Th after it is produced by alpha decay of ^{238}U.

* ^{235}U-substantial depletion of its abundance has been observed in the Oklo natural reactor and other cases like reprocessed sources (Firestone, 2003).

Energy

Taking gold as a case study, it was found out (Firestone 2005) that changes in the calibration energy of ^{198}Au have contributed to significant variations in different measurements of standard calibration sources. The variation of the gold energy over time is shown in Table 2.

Half-life, Effective Resonance Energy and Q_0-Value

The gamma-rays of the radio nuclides by Erdtmann and Soyka, 1979; the k_0-consistent IRI Gamma-ray Catalogue for INAA by Blaauw, 1996 and the Recommended nuclear data for use in the k_0 standardization of neutron activation analysis by De Corte and Simonits, 2003 were use to demonstrate some of the inconsistencies found in the values of half-life, effective resonance energy and the ratio of thermal cross-section to resonance integral (Table 3).

Results and Discussion

Table 1: Comparison of decay data from k_0-Website and the DDEP

Energy	Transition Prob. (k_0)%	Transition Prob. (DDEP)%
66.0518	---	1.112 (12)
96.7340	3.481	3.42 (3)
121.115	17.32	17.2 (3)
136.0001	58.98	58.2 (7)
198.6060	1.472	1.48 (4)
264.6576	59.1	58.9 (3)
279.5422	25.18	24.99 (13)
303.9326	1.342	1.316 (8)
400.6572	11.56	11.47 (9)

Table 2: Variation of ^{198}Au gamma-ray energy over time (Source: Firestone, 2005)

Year	Energy (keV)
1964	411.795 ± 0.009
1977	411.80441 ± 0.00015
2004	411.80205 ± 0.00017

A case of remarkable inconsistency in nuclear data compilations is illustrated in Figure 1 (De Corte 2002). The thermal cross-section of Yb-175 as reported by Mughabghab (Mughabghab 1984) was 69 barns and in 1987 IAEA gives a value of 63 barns (Figure 1). While compiling k_0-values, De Corte and Simonits (1988) reported to have obtained a high value of 128 barns which made other workers like CHNUCL 1996 and HOLD 1996 to adjust their initial values to 120 barns. However, by introducing double gamma-intensity values in experimental k_0-factors, the Miyahara group realized that their initial value was wrong (Miyahara et.al., 1994).

From Table 3, it will be observed that the major problem with nuclear data is the effective resonance energy, half-life and the ratio of resonance integral to thermal

cross-section values. The largest variation of Q_0-value among the nuclides selected for this work is the case of Fluorine (F-19) and Argon (Ar-40).

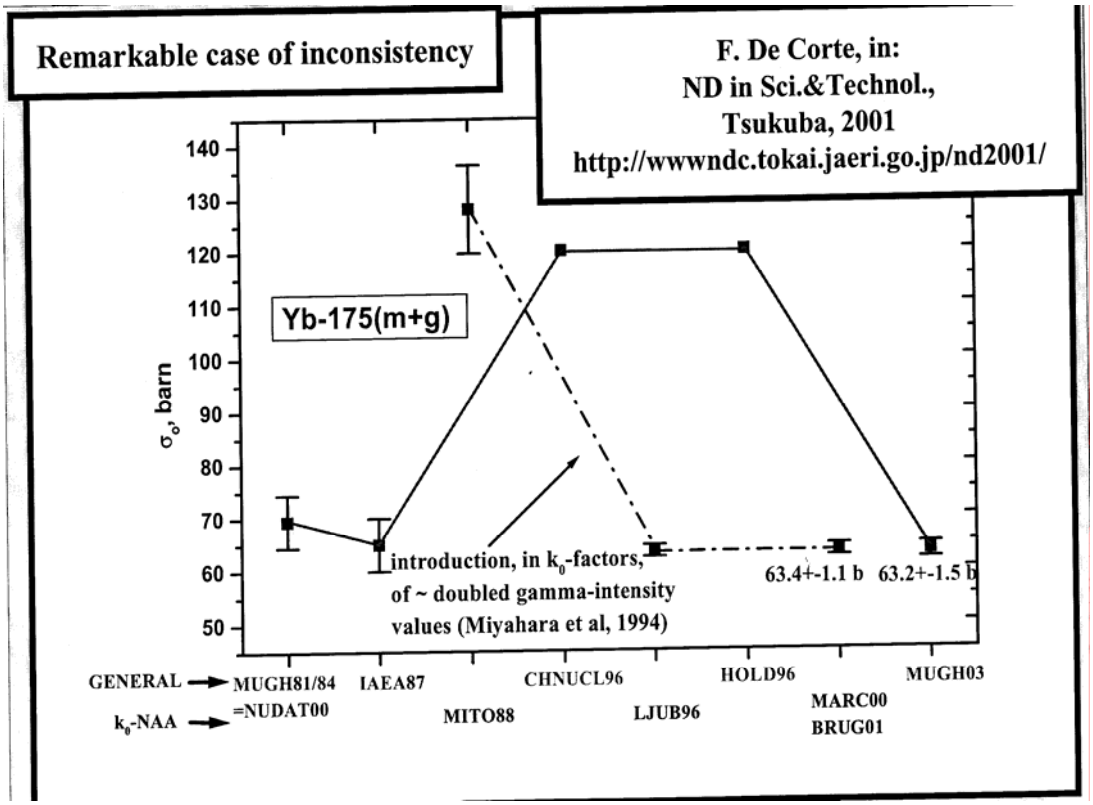

Fig. 1: Variations of Yb-175 thermal cross-section values

Similar discrepancies could be observed as one goes through the remaining elements of the periodic table. It could generally be said that Erdtman and Soyka, 1979 values (especially energy of radionuclide) are lower than the ones reported by M. Blaauw, 1996 and De Corte and Simonits, 2003. It therefore indicates that results obtained with equations 2 and 5 depend largely on the nuclear data source rather than the experimental procedure used.

Table 3: Analysis of nuclear data values as reported by three different sources

Key: Erdtman and Soyka, 1979. M. Blaauw, 1996. De Corte and Simonits, 2003.

	Energy (keV)			Half-life			E_r (eV)		Q_0	
	1979	1996	2003	1979	1996	2003	1996	2003	1996	2003
F	1633.1	1633.6	1633.6	11.0s	11.03s	11.16s	1.0	44,700	4.1	2.2
Na	2754.1	2753.9	2754.0	15.03h	14.9h	14.96h	3380	3380	0.6	0.59
Al	1778.0	1778.99	1778.9	2.31m	2.24m	2.24m	11800	11800	0.7	0.71
Ar	1293.6	1293.64	1293.6	1.83h	1.83h	1.82h	1.0	31000	0.7	0.63
Ca	3084.4	3084.54	3084.4	8.70m	8.72m	8.72m	1330000	1330000	0.5	0.45
Zn	1115.52	1115.52	1115.5	243.8d	244.1d	244.3d	2560	2560	1.9	1.908
Zr	756.72	756.73	756.7	64.4d	64.02d	64.02d	6260	6260	5.0	5.31
Zr	743.36	Nil	743.4	16.8h	16.9h	16.74h	338	338	248	251.6
Nb	16.6	Nil	871.0	6.29m	6.26m	6.26m	574	574	7.3	7.35
Au	411.8	411.8	411.8	2.697d	2.693d	2.695d	5.7	5.65	15.7	15.7

Table 4: Determine Reactor Flux Parameters using the Erdtman and Soyka 1979 Nuclear data

Channel (GHARR-1)	Monitor	*Energy (keV)*	$T_{1/2}$	σ_0 *(b)*	γ *(%)*	Q_o	E_r *(eV)*	E_{cd} *(eV)*	f (Expt.)
5 (Inner)	Au-197	411.8	2.697d	98.7	95.5	15.71	5.65	0.55	17.28
	Zr-96	743.36	16.8h	0.0213	98.0	251.6	338	0.55	
	Zr-94	756.72	64.4d	0.053	54.8	5.06	6260	0.55	
7 (Outer)	Au-197	411.8	2.697d	98.7	95.5	15.71	5.65	0.55	40.38
	Zr-96	743.36	16.8h	0.0213	98.0	251.6	338	0.55	
	Zr-94	756.72	64.4d	0.053	54.8	5.06	6260	0.55	

Table 5: The use of M. Blaauw 1996 Nuclear data to determine flux parameters

Channel (GHARR-1)	Monitor	*Energy (keV)*	$T_{1/2}$	σ_0 *(b)*	γ *(%)*	Q_o	E_r *(eV)*	E_{cd} *(eV)*	f (Expt.)
5 (Inner)	Au-197	411.8	2.694d	98.7	95.5	15.7	5.70	0.55	17.32
	Zr-96	743.36	16.9h	0.0213	98.0	248.0	338	0.55	
	Zr-94	756.72	64.02d	0.053	54.8	5.00	6260	0.55	
7 (Outer)	Au-197	411.8	2.694d	98.7	95.5	15.7	5.70	0.55	40.43
	Zr-96	743.36	16.9h	0.0213	98.0	248.0	338	0.55	
	Zr-94	756.72	64.02d	0.053	54.8	5.00	6260	0.55	

As observed from Table 1, it is apparent that both k_0-Website Operators and the Decay Data Evaluation Project (DDEP) require a combined evaluation of ^{75}Se data. Before that is done, NAA users are advice to look at both sources of information when estimating uncertainties in the decay scheme of Selenium. Table 3 revels that effective resonance energy, ratio of thermal cross-section to resonance integral (Q_0), half-life and branching ratio are the worst affected by inconsistencies. In some nuclides, the Q_0 values reported by Blaauw 1996 shows a difference of about 46.3% (Fluorine) from the one reported by Decorte and Simonits 2003. On the other hand, effective resonance energy values reported by Decorte and Simonits 2003 in some cases (Fluorine and Argon) are far highly than those of Blaauw 1996. It has been observed in some nuclides that half-life do not generally agree with each other (Table 3). Users are forewarned to be wary of very precise half-life value based on single measurements. The effects of all these could be seen in the differences of the thermal to epithermal flux values obtained in Tables 4 and 5 for flux measurements in the irradiation channel of an MNS reactor. The variation in the two results aroused from the differences in Q_0 values (derivable from the thermal cross-section and the resonance integral), half life and the effective resonance energy of the monitor nuclides as reported by the two authors of the nuclear data used.

Conclusion

In this study, the nuclear data reported by two authors was used to determine thermal to epithermal flux ratio in the irradiation channels of the Ghana Research Reactor-1. Results obtained with M. Blaauw 1996 data is 17.32 for inner irradiation channel 5 and 40.43 for outer channel 7. Using the Erdtman and Soyka 1979 nuclear data, we got 17.28 and 40.38 for the same channels respectively. From the variation in the flux ratio values obtained using the two nuclear data and the inconsistencies observed in the nuclear parameters reported by different authors, this study shows that nuclear data source significantly influence the precision of instrumental neutron activation analysis.

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