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# **Nuclear Reactions and Physical Models for Neutron Activation Analysis**

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# 1 Introduction

Due to its selectivity and sensitivity, neutron activation analysis (NAA) occupies an important place among the various analytical methods. It has proven to be a powerful non-destructive analytical technique for concentrations at or below the  $\mu\text{g/g}$  range, while up to 60 elements can be determined, performing two irradiations and several gamma-spectrum measurements after different decay periods. The main fields of NAA 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 candidate reference materials (RMs), providing e.g. the bulk of the literature data on the standard RMs of the National Institute of Standards and Technology [1] and reference materials of the International Atomic Energy Agency.

The  $k_0$  standardisation method of NAA ( $k_0$ -NAA), a concept launched in 1975, can be interpreted as an absolute standardisation method. It relies on  $k_0$  and  $Q_0$  factors and a few other parameters [2], which are composite physical constants that can be derived from the basic nuclear data. In practice they are usually determined by direct measurements, partly because equivalent constants derived from the basic data are often discrepant.

The purpose of this paper is to:

- define the reaction rate equations as used in NAA and their relation to the exact definitions from the basic nuclear data,
- identify sources of uncertainties and approximations and their propagation to calculated reaction rates.

The overall objective is to define the basis that will improve the understanding of the definitions and lead eventually to the improvement in the nuclear constants for NAA, as well as the basic nuclear data where accurately measured composite constants for NAA can provide additional constraints for the basic nuclear data evaluation process.

## 2 Definitions

### 2.1 Specific activities

When a material is placed in a neutron field, some nuclei of the material may capture neutrons to form excited nuclei, which return promptly to the ground state by emitting gamma radiation. The capture product nuclei are often radioactive. Decay by particle emission produces decay product nuclei in an excited state, which go into the ground state by again emitting gamma radiation. Gamma radiation associated with radioactive decay of a nucleus is actually the radiation of its decay product returning to the ground state. Different variants of the activation analysis as an analytical technique differ in the radiation that is being measured. Usually these are either prompt gamma rays emitted by the excited capture product or the delayed gamma rays emitted by the excited decay product nuclei.

If irradiation is performed in a neutron field with a significant fraction of high energy neutrons, it may happen that some threshold reactions on other nuclei present in the sample produce the same product nucleus as obtained by capture in the measured nucleus. Also, if some fissile material is present in the sample, such material may yield on fission a small amount of nuclei equal to the capture product nuclei. These are interference reactions and must be taken into account.

During irradiation some of the capture products decay and some may themselves interact with neutrons to form a different nucleus and are lost for the purpose of the measurement. At high neutron flux levels the target nuclei may become depleted, what also affects the production of the capture-product nuclei. The differential equation governing the rate of change of the concentration of the nuclei of interest is given by:

$$\frac{dN_c}{dt} = \phi N_m \sigma_m + \phi N_h \sigma_h + \phi N_f \sigma_f \gamma_f - \phi N_c \sigma_c - \lambda_c N_c$$

where  $\phi$  is the neutron flux,  $\sigma$  are the cross sections,  $N$  the nuclei number densities in the sample,  $\lambda$  the decay constant and  $\gamma$  the fission product yield. The suffixes are: “m” for the measured nuclei, “c” for the capture products, “f” for the fissile nuclei and “h” for nuclei that form nucleus “c” from high energy threshold reactions with cross section  $\sigma_h$ . A similar equation can be written for each type of nucleus, forming a system of coupled first order linear differential equations that can be solved numerically, or analytically (with some approximations, usually by neglecting all terms on the right-handside except the first and the last.).

Specific activity of a sample is the measured activity at the end of irradiation, corrected for the change of the concentration of the decaying nucleus due to measuring time, cooling time and irradiation time. It can be shown analytically that specific activity is proportional to the average reaction rate that produces the nucleus, which generates the emitted radiation that is being measured. Therefore, from the measured activity it is possible to determine the reaction rate and hence the concentration of the measured nucleus, provided we know the neutron flux and the corresponding nuclear constants. The remainder of this paper is devoted to the topic of reaction rate calculation and to associated nuclear constants.

## 2.2 Reaction rates

Reaction rate  $A$  of particles travelling through a material with nuclei of that material is parameterised by the reaction cross section  $\sigma(v)$ , which is the property of the material and the neutron flux spectrum  $\phi(v)$ , which is related to the density of the particles travelling through the material  $n(v)$  and their speed  $v$ :

$$\phi(v) = v n(v) . \tag{1}$$

Expressed in terms of the kinetic energy  $E$  of the incident particles, which is related to the speed  $v$  by the relation  $E = \frac{1}{2} m v^2$  (where  $m$  is the particle mass), the reaction rate is:

$$A = K \int_0^{\infty} \sigma(E) \phi(E) dE ; \quad \phi(1eV) = 1. \tag{2}$$

The normalisation of  $\varphi(E)$  is quite arbitrary and is chosen for convenience. The constant  $K$  ensures that the integral of  $\varphi(E)$  over energy results in the total neutron flux. In terms of neutron speed the equivalent expression for the reaction rate can be written as:

$$A = \int_0^{\infty} \sigma(v) v \cdot n(v) dv. \quad (3)$$

The integral can be split into the thermal part up to energy  $E_{cd}$  (corresponding to neutron speed  $v_{cd}$ ) and the epithermal part:

$$A = K \left[ \int_0^{E_{cd}} \sigma(E) \varphi(E) dE + \int_{E_{cd}}^{\infty} \sigma(E) \varphi(E) dE \right]. \quad (4)$$

Furthermore, the epithermal flux can be decomposed into the resonance part  $\varphi_r$  and the suitably normalised fast (fission) spectrum contribution  $\varphi_h$  for convenience:

$$\varphi(E) = \varphi_r(E) + h \varphi_h(E). \quad (4a)$$

Precise modelling of the fission spectrum contribution does not have a significant influence on calculated reaction rates in well-thermalised spectra; it might improve the modelling of reaction rates in irradiation facilities with a strong epithermal neutron spectrum component, but it is crucial for threshold reactions. The reaction rate equation becomes:

$$A = K \left[ \int_0^{E_{cd}} \sigma(E) \varphi(E) dE + \int_{E_{cd}}^{\infty} \sigma(E) \varphi_r(E) dE + h \int_{E_{cd}}^{\infty} \sigma(E) \varphi_h(E) dE \right]. \quad (4b)$$

The above expressions are *exact*; the problem is that neither the cross sections nor the neutron flux spectrum are known accurately enough due to the strong dependence of the parameters on neutron energy. For example, to represent the capture cross section of  $^{238}\text{U}$  to within 0.1% tolerance, several 100 000 data points are needed. Such detailed representation is necessary to ensure proper account of the Doppler broadening effect due to temperature and for the estimation of the self-shielding effects.

Without loss of generality, the integral equation (4b) can be cast into the expression commonly used in NAA by a suitable definition of constants:

$$A = \varphi_t \sigma_0 g G_t + \varphi_f (I G_f + J h) = \varphi_t \sigma_0 \left[ g G_t + \frac{1}{f} (Q G_f + H h) \right], \quad (5)$$

where the symbols have the following meaning:

- $\varphi_t$  thermal flux,
- $\varphi_f$  epithermal flux,
- $f$  ratio of thermal to epithermal flux  $\varphi_t/\varphi_f$ ,
- $\sigma_0$  thermal cross section at 2200 m/s neutron speed,
- $g$  generalised g-factor that measured the deviation of the thermal cross section from  $1/v$  shape,
- $I$  effective resonance integral,
- $J$  effective fission integral
- $Q$  ratio of the resonance integral and the thermal cross section  $I / \sigma_0$ ,

- $H$  ratio of the fission integral and the thermal cross section  $J / \sigma_0$ ,
- $h$  fission spectrum factor,
- $G_t$  thermal flux depression factor,
- $G_f$  resonance self-shielding factor.

The applicability and the accuracy of the above expression depend on the approximations involved in determining the constants. Correspondence and definitions of individual terms are discussed in the sections that follow.

To avoid the need to determine the neutron flux, the  $k_0$  standardization method of NAA relies on the measurement of the ratio of specific activities (and reaction rates) of the measured nuclide and some well-defined standard. The commonly applied standard is gold, because it has well-known cross sections and an associated gamma ray of accurately-known emission probability that is relatively easy to measure. The ratio of the specific activity of the sample  $A_a$  relative to the specific activity of the standard  $A_s$  is related to the ratio of reaction rates, given by the following expression:

$$\frac{A_a}{A_s} = k_{0,a} \frac{G_t f g_a + G_{f,a} Q_a + H_a h}{G_t f g_s + G_{f,s} Q_s + H_s h} \quad (6)$$

where

$$k_{0,a} = \frac{M_s \Theta_a \gamma_a \sigma_{0,a}}{M_a \Theta_s \gamma_s \sigma_{0,s}} \quad (7)$$

and the constants with index  $x$  equals  $a$  for sample and  $s$  for standard are:

- $M_x$  molar mass of sample,
- $\Theta_x$  natural atomic abundance,
- $\gamma_x$  gamma emission probability of the measured gamma ray,
- $\sigma_{0,x}$  thermal capture cross section.

### 2.3 Thermal cross-section $\sigma_0$ , g-factor and thermal flux depression factor $G_t$

Contribution of thermal neutrons to the reaction rate, expressed in “neutron speed” domain is given by:

$$A_t = \int_0^{v_{cd}} \sigma(v) v \cdot n(v) dv. \quad (8)$$

For a  $1/v$  absorber the cross section is:

$$\sigma(v) = \sigma_0 \frac{v_0}{v} \quad (9)$$

where the symbols are:

- $v_0$  thermal neutron speed, 2200 m/s by definition,
- $\sigma_0$  cross section at neutron speed  $v_0$ .

Substituting into the equation for  $A_t$

$$A_t = \sigma_0 v_0 \int_0^{v_{cd}} n(v) dv = \sigma_0 v_0 N_t, \quad (10)$$

where  $N_t$  is the total thermal neutron density (i.e. total number of neutrons per unit volume). Note that the reaction rate is independent of the neutron speed distribution  $n(v)$ .

In the energy domain the equivalent expression for the thermal reaction rate is

$$A_t = K \int_0^{E_{cd}} \sigma(E) \phi(E) dE \quad (11)$$

Substituting the expression for kinetic energy into equation (9) for a  $1/\nu$  absorber

$$\sigma(E) = \sigma_0 \sqrt{\frac{E_0}{E}} \quad (12)$$

where  $E_0$  is the energy of thermal neutrons corresponding to  $\nu_0$  and is equal to 0.0253 eV. Simplification of the integral for reaction rate in the energy domain is not possible. The reaction rate is proportional to the total thermal neutron density, but not to the total thermal neutron flux.

Assuming that the thermal neutron flux has Maxwellian distribution

$$\phi(E) = E e^{-E/kT}, \quad (13)$$

where  $k$  is the Boltzman constant  $T$  is the temperature and  $K$  is the flux scaling factor, the thermal reaction rate is given by:

$$A_t = K \int_0^{E_{cd}} \sigma(E) E e^{-E/kT} dE. \quad (14)$$

For a  $1/\nu$  absorber

$$A_t = K \sigma_0 \sqrt{E_0} \int_0^{E_{cd}} \sqrt{E} e^{-E/kT} dE. \quad (15)$$

The average thermal cross section  $\sigma_{th}$  is defined by

$$\sigma_{th} = \frac{\int \sigma(E) \phi(E) dE}{\int \phi(E) dE} = \frac{K \sigma_0 \sqrt{E_0} \int \sqrt{E} e^{-E/kT} dE}{K \int E e^{-E/kT} dE}. \quad (16)$$

Extending the integration limits from 0 to  $\infty$ , recognising the integral in the numerator as the gamma function  $\Gamma(3/2)$  and using the relation between the energy and the temperature  $E_0 = kT_0$ , the average thermal cross section  $\sigma_{th}$  is related to the thermal cross section  $\sigma_0$  by the relation:

$$\sigma_{th} = \frac{K \sigma_0 (kT)^{3/2} \sqrt{kT_0} \sqrt{\pi} / 2}{K (kT)^2} = \frac{\sqrt{\pi}}{2} \sigma_0 \sqrt{\frac{T_0}{T}}. \quad (17)$$

Note that this relation is strictly valid only for a pure  $1/\nu$  absorber in a Maxwellian spectrum. In practice, the cross sections may deviate from the  $1/\nu$  behaviour and the spectrum may be distorted (depending on the irradiation facility). Westcott attempted to correct for the non-ideal cross section behaviour by introducing the Westcott g-factor, but still assumed that the spectrum was of Maxwellian shape. He even took the trouble to extract the  $1/\nu$  part of the cross section contribution from the resonance range above the energy  $E_{cd}$ . At the time when the Westcott formalism was developed, the knowledge of cross section shapes was lacking and determining the spectral shape was based more on intuition and educated guessing than

anything else. Computational power posed additional limitations, which favoured analytical approaches. With many of these constraints relaxed, it is possible to introduce an alternative definition of the generalised g-factor, which can be used to calculate reaction rates without loss of generality and is applicable to non-1/v absorbers as well as spectra, which deviate from the Maxwellian shape. Comparing equations (4) and (5) see that:

$$\varphi_t \sigma_0 g G_t = K \int_0^{E_{cd}} \sigma(E) \varphi(E) dE . \quad (18)$$

Arbitrarily we define

$$\varphi_t = K \frac{\sqrt{\pi}}{2} \int_0^{E_{cd}} \varphi(E) dE . \quad (19)$$

Neglecting the thermal flux depression factor  $G_t$  for the time being (assuming it is equal to 1), the definition of the generalised g-factor follows:

$$g = \frac{\int_0^{E_{cd}} \sigma(E) \varphi(E) dE}{\sigma_0 \frac{\sqrt{\pi}}{2} \int_0^{E_{cd}} \varphi(E) dE} = \frac{2}{\sqrt{\pi}} \frac{\sigma_{th}}{\sigma_0} . \quad (20)$$

Substituting the integrals with the expression for  $\sigma_{th}$  it is easily seen that for a 1/v absorber in a Maxwellian spectrum the above definition gives the well-known Westcott g-factor relation:

$$g_w = \sqrt{\frac{T_0}{T}} g . \quad (21)$$

In addition to the applicability to arbitrary spectra, the main difference in the generalised definition of the g-factor is the upper integration limit  $E_{cd}$ , commonly taken as 0.55 eV. Normally this does not affect the value of the g-factors in Maxwellian spectra because the distribution function falls off very rapidly and the contribution to the integral above 0.55 eV is very small.

The generalised g-factor can be calculated easily from the cross sections, which are readily available for practically all nuclides of interest. The value of the calculated g-factor does not depend on the absolute magnitude of the cross sections, which may have significant systematic errors, but only on the shape.

Introduction of the generalised definition of the g-factor extends the applicability of the methods, which rely on simple expressions for reaction rates such as given in equation (5), to irradiation facilities with spectra that deviate significantly from the Maxwellian shape in the thermal region.

The thermal neutron flux depression factor  $G_t$  is often referred to as the ‘‘thermal self-shielding factor’’, but the term is misleading, because it implies primary dependence on the measured nuclide in the sample. This is indeed the case with resonance absorption in the epithermal range, but not in the thermal range, where neutron transport effects play a dominant role. The thermal neutron flux depression factor is therefore determined by the macroscopic cross sections of the sample material as a whole. It can be calculated by a direct



transport calculation or from parameterised expressions, which are discussed in more detail in the literature [3,4]

## 2.4 Resonance integral $I$ , cadmium transmission factor $F_{cd}$ and $Q$ value

The reference resonance integral  $I_0$  is usually defined by the product of the cross section and a pure  $1/E$  spectrum, integrated between some chosen *cadmium cutoff* energy  $E_c$  and an arbitrarily chosen upper limit  $E_3$ :

$$I_0 = \int_{E_{cd}}^{E_3} \sigma(E) \psi(E) dE \quad ; \quad \psi(E) = \frac{1}{E}. \quad (22)$$

Similarly, the reference  $Q_0$  value is given by:

$$Q_0 = \frac{I_0}{\sigma_0}. \quad (23)$$

This definition is rather artificial because such a spectrum with sharp cutoff energies cannot be produced experimentally. Measurements are usually done in thermal reactor spectra, which approximately follow the  $1/E$  behaviour in the epithermal energy range. If the irradiation position is separated from the fission source (usually the reactor core) by a relatively thick moderator material region, relatively few fission neutrons reach the irradiation position directly, so the fission peak in the spectrum is small. The fission spectrum falls off rather rapidly above the peak, so the energy around 2 MeV is the natural upper cutoff energy. At the low energy end, thermal neutrons can be filtered by a strong absorber like cadmium, which has a huge resonance at 0.178 eV and relatively weak resonances at higher energies. A 1 mm cadmium filter effectively removes most neutrons below 0.55 eV. The total cross section of cadmium is shown in Figure 1 part a). The resonance integral can be approximated by the reaction rate  $I_{cd}$  measured under a cadmium filter.

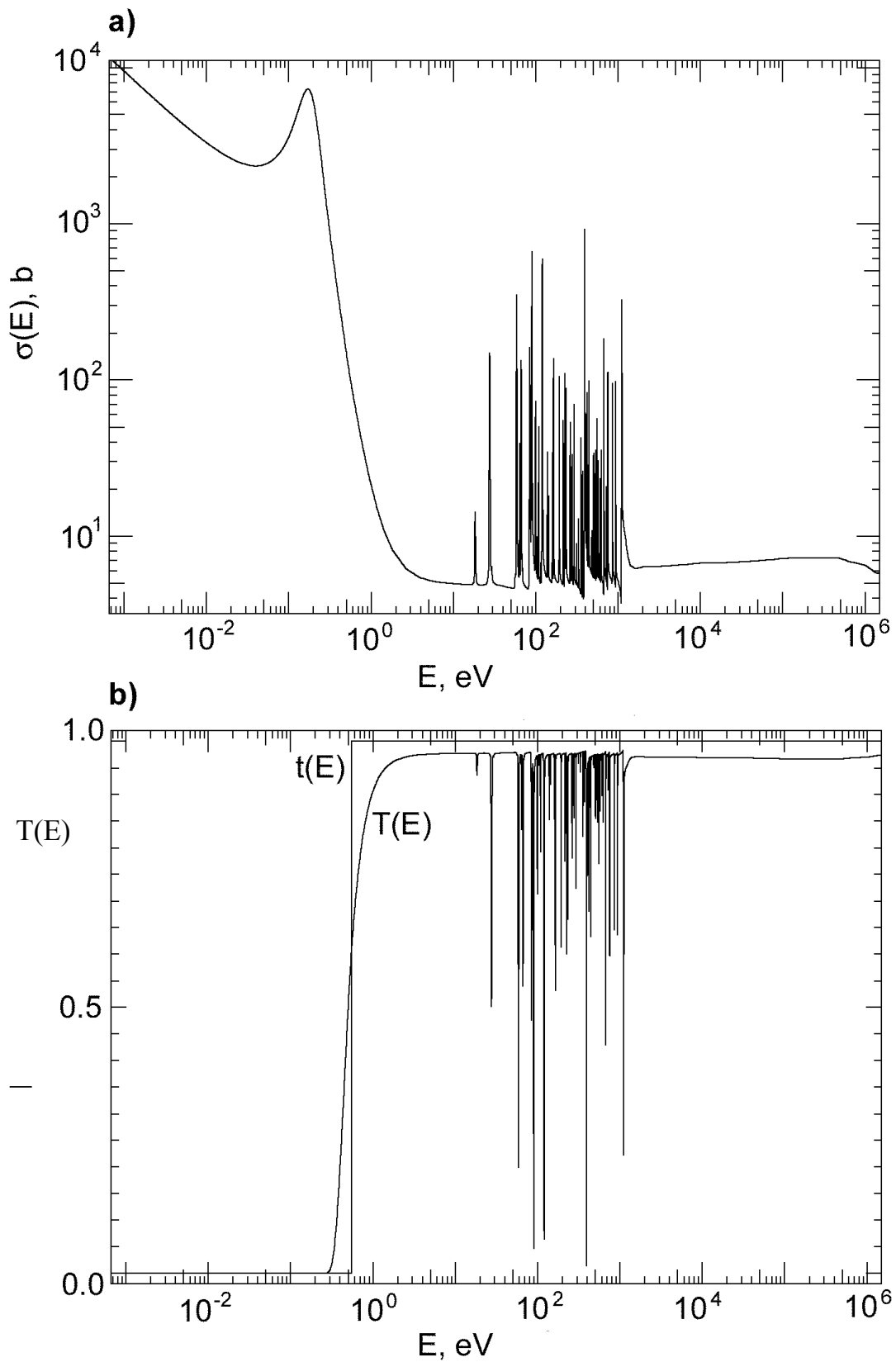


Figure 1: a) Total cross section of cadmium.  
 b) Cadmium transmission function for a 1mm thick cadmium cover.

Introducing the cadmium transmission function  $t(E)$ , a more precise definition of the measured resonance integral under cadmium cover  $I_{cd}$  in a real spectrum  $\varphi^*(E)$  is obtained:

$$I_{cd} = \int_0^{\infty} t(E)\sigma(E)\varphi^*(E) dE . \quad (24)$$

The above equation reduces to the previous idealised one if the range of integration is limited from  $E_{cd}$  to  $E_3$ , the spectrum is pure  $1/E$ , and  $t(E)$  is an idealised Heaviside function:

$$t(E) = \begin{cases} 0 & \text{for } E < E_{cd} \\ 1 & \text{for } E \geq E_{cd} \end{cases} \quad (25)$$

A more realistic form of the cadmium transmission function is obtained by assuming exponential attenuation of neutrons through cadmium:

$$t(E) = e^{-N_{cd} d \sigma_{cd}(E)} \quad (26)$$

where  $d$  is the cadmium cover thickness,  $\sigma_{cd}$  is the cadmium cross section and  $N_{cd}$  is the number density of cadmium atoms in the cover. It is calculated as:

$$N_{cd} = \frac{\rho_{cd} N_A}{M_{cd}} , \quad (27)$$

where  $\rho_{cd}$  is the density of cadmium,  $N_A$  the Avogadro number and  $M_{cd}$  the molar mass of cadmium.

In a collimated narrow neutron beam incident on a small target, any reaction on cadmium would remove a neutron from its path. Using the total cross section for  $\sigma_{cd}$  the cadmium transmission function would be valid exactly. In practice, the beam profile and target dimension are finite; the neutron field may be isotropic, in which case there is a high probability that scattered neutrons would also reach the target. In such cases only the absorption reaction really removes the neutrons so it may be more appropriate to define  $\sigma_{cd}$  as the absorption cross section. In reality the truth is somewhere in between. Note that the form of the cadmium transmission function is the first approximation in the definitions introduced so far.

Resonance integral defined by equation (23) is a measurable quantity. This is to be compared with the required form evident from equations (4) and (5). The cadmium transmission factor  $F_{cd}$  is introduced to compensate for the non-ideal shape of the cadmium filter, assuming the spectrum closely follows the  $1/E$  behaviour and ignoring (or subtracting out) the high energy contribution of the fission spectrum:

$$I = \int_{E_{cd}}^{E_3} \sigma(E)\varphi(E) dE = \frac{1}{F_{cd}} \int_0^{\infty} t(E)\sigma(E)\varphi(E) dE . \quad (28)$$

From this it follows that:

$$F_{cd} = \frac{\int_0^{\infty} t(E)\sigma(E)\phi(E) dE}{\int_{E_{cd}}^{E_3} \sigma(E)\phi(E) dE}. \quad (29)$$

Deviation of  $F_{cd}$  from unity arises from the cadmium transmission function and from the difference in the upper integration limit. The contribution of the later is small in the case of  $1/E$  spectrum with a small component of the fission neutrons in the spectrum. This is usually the case for irradiation facilities behind a reflector. Cadmium transmission factor values can be calculated from the cross sections by direct integration according to equation (29), assuming a  $1/E$  spectrum and choosing appropriate integration limits in the numerator and the denominator ( $E_3=2$  MeV, say); the lower integration limit  $E_{cd}$  is chosen to approximately match the effective cutoff of the cadmium cover. It depends on the cadmium thickness. The value 0.55 eV is usually adopted for a cadmium thickness of 1 mm.

Irradiation channels inside (or near) the reactor core may exhibit spectra with a significant contribution of fission neutrons. In such cases the neutron spectrum characterisation has to be done very carefully and the cadmium transmission factor calculated directly from the cross sections and the actual spectrum of the irradiation facility.

In NAA databases very few nuclides contain  $F_{cd}$  factors that deviate from unity, and even those have to be considered with care. For example, commonly adopted value for  $^{186}\text{W}$  is 0.908 and yields measured  $Q_0$  values, which disagree by nearly 10% from those calculated from the energy-dependent cross sections in evaluated nuclear data files [8]. Direct calculations, using cross sections to simulate the transmission of neutrons through 1 mm cadmium layer, results in a cadmium transmission factor that differs from unity by about 1%. Furthermore, the measured value of 0.908 is not given with the associated uncertainty. From the original paper on the measurement the uncertainty is likely to be high and the quoted measured  $F_{cd}$  is probably incorrect.

With the resonance integral uniquely defined, the  $Q$  value for a general neutron spectrum can also be defined in a way analogous to equation (23):

$$Q = \frac{I}{\sigma_0} \quad (30)$$

The reference  $Q_0$  for an ideal  $1/E$  spectrum is already defined by equation (23). Relation between the reference  $Q_0$  and the general  $Q$  is discussed in Section 2.5. Some comparisons between measured values and those calculated using cross sections from evaluated nuclear data files are given in Section 3.2.

## 2.5 Resonance self-shielding factor $G_f$

In the absence of strong absorbers the neutron spectrum as a function of energy is a smooth function. When resonance absorbers are present in significant quantities, the resonances tend to create dips in the spectrum shape. This phenomenon is well known in reactor physics and has been dealt with extensively in the so-called resonance theory. In the intermediate-

resonance approximation (IR) the real spectrum  $\phi^*(E)$  is expressed in terms of the spectrum unperturbed by the resonance absorber  $\phi(E)$  by the expression:

$$\phi^*(E) = \frac{\sigma_0 + \lambda \sigma_p(E)}{\sigma_0 + \lambda \sigma_a(E) + \sigma_s(E)} \phi(E) \quad (31)$$

where:

- $\sigma_0$  Bondarenko background cross section, which measures the effective dilution of the resonance absorber,
- $\sigma_a$  absorption cross section of the resonance absorber,
- $\sigma_s$  scattering cross section of the resonance absorber,
- $\sigma_p$  potential scattering cross section of the resonance absorber,
- $\lambda$  Goldstein - Cohen parameter - a "measure" of the resonance width,
- $\phi(E)$  smooth spectrum (unperturbed by the resonances).

The intermediate resonance approximation is an improvement to the narrow resonance (NR) approximation ( $\lambda=1$ , implying that the resonances are so narrow that any scattering event will decrease neutron energy sufficiently to fall outside the resonance) and the wide resonance (WR) approximation ( $\lambda=0$ , assuming that energy loss in a scattering event is small compared to the resonance width).

Resonance theory is based on the assumption that the absorber atom is surrounded by a moderator of approximately constant cross section, represented by the Bondarenko background cross section, which effectively measures the dilution of the absorber and is defined as the macroscopic potential cross section of the moderator per absorber atom:

$$\sigma_0 = \frac{1}{N_a} \sum_i N_i \lambda_i \sigma_i \quad (32)$$

where

- $N_a$  absorber atom number density,
- $N_i$  number density of the  $i$ -th moderator nucleus,
- $\sigma_i$  cross section of the  $i$ -th moderator nucleus,
- $\lambda_i$  parameter (related to the Goldstein-Cohen parameter) that measures the moderator effectiveness. By definition it is equal to 1 for hydrogen. Further details can be found in the documentation of the WIMS-D Library Update Project [5].

The above derivation is applicable to infinite homogeneous media, but irradiated samples are of finite dimensions. In the surrounding medium (analogous to a moderator without strong resonance absorbers) the spectrum is relatively smooth. The neutrons enter the sample (containing a resonant absorber), but their depth of penetration at resonance energies is limited due to the absorption in the resonances. The process is therefore similar to the one in an infinite medium. In reactor physics this is called the equivalence theorem. Equation for the Bondarenko background cross section is modified to include the so-called escape cross section  $\Sigma_e$ , which accounts for the finite dimensions of the sample:

$$\sigma_0 = \frac{1}{N_a} \left[ \Sigma_e + \sum_i N_i \lambda_i \sigma_i \right] \quad (33)$$

The escape cross section is given by the simple expression:

$$\Sigma_e = \frac{a}{l} \quad (34)$$

where

$a$  Bell factor (usually assumed constant with value 1.16),  
 $l$  mean chord length.

The mean chord length for a convex volume is given by

$$l = \frac{4V}{S} \quad (35)$$

where  $V$  is the volume and  $S$  is the surface area.

Epithermal self-shielding factor describing the effects of resonance absorption can be defined by:

$$G_f = \frac{\int_{E_{cd}}^{E_3} \sigma(E) \phi^*(E) dE}{\int_{E_{cd}}^{E_3} \sigma(E) \phi(E) dE} \quad (36)$$

With weighting spectrum  $\phi^*(E)$  defined by equation (31). A practical procedure is to generate a library of self-shielding factors for all nuclides of interest, and particularly the main likely constituents of sample materials with significant absorption properties, tabulated as a function of the Bondarenko background cross section  $\sigma_0$ . The user can then calculate the relevant value of  $\sigma_0$  from equations (33-34) and retrieve the required  $G_f$  by interpolation. The main approximations in this approach are those inherent in the IR resonance approximation and the assumption that  $G_f$  factors are not sensitive to small deviations in the weighting spectrum  $\phi(E)$ , which is usually assumed to be of the  $1/E$  form.

## 2.6 Effective resonance energy $E_r$

The resonance integral and the  $Q$  value depend on the shape of the neutron spectrum in the epithermal range. Assuming that the spectrum deviates only slightly from the  $1/E$  behaviour such that it can be represented by

$$\phi(E) = \frac{1}{E^{1+\alpha}}, \quad (37)$$

where  $\alpha$  is a constant, and assuming that resonances can be represented by the single-level Breit-Wigner formula, an analytical expression is commonly in use in NAA for the dependence of the  $Q$  value on  $\alpha$  in terms of the reference  $Q_0$  value in a pure  $1/E$  spectrum:

$$Q(\alpha) = \frac{Q_0 - 0.429}{(E_r)^\alpha} + \frac{0.429}{(2\alpha + 1)(0.55)^\alpha} \quad (38)$$

To verify the validity of the approximation for  $Q(\alpha)$ , exact values were calculated directly from the cross sections based on Equations (28) and (20) for a set of  $\alpha$  values in the range between  $-0.1$  and  $+0.1$ , and using idealised cadmium transmission function with cutoff at  $0.55$  eV. By inverting equation (38) an expression for  $E_r$  can be obtained:

$$E_r(\alpha) = \left\{ \frac{Q_0 0.429}{Q(\alpha) - \frac{0.429}{(2\alpha+1)(0.55)^\alpha}} \right\}^{1/\alpha} \quad (39)$$

The average  $E_r$  is defined by the integral

$$E_r = \frac{1}{\alpha_{hi} - \alpha_{lo}} \int_{\alpha_{lo}}^{\alpha_{hi}} E_r(\alpha) d\alpha \quad (39a)$$

and the integration limits  $\alpha_{hi}$  and  $\alpha_{lo}$  are chosen  $+0.1$  and  $-0.1$ , respectively.

The calculated  $E_r(\alpha)$  for different values of  $\alpha$  were found to vary by up to 30% from the average value  $E_r$ . The  $\alpha$ -dependent  $E_r(\alpha)$ , (normalised by the average  $E_r$ ) for different nuclides is shown in Figure 2. The calculated average effective resonance energy  $E_r$  also differs quite significantly from the values usually adopted for NAA, which were derived from the available resonance parameters. The comparison is shown in Table 1. In the case of  $^{94}\text{Zr}$  the difference exceeds a factor of two.

**Table 1: Comparison of Effective Resonance Energies  $E_r$  for Neutron Activation Analysis**

$E_r$ nuclide	Kayzero [eV]	This work [eV]	Dif. [%]
Mn-55	468.0	488.1	4.3
Co-59	136.0	122.4	-10
Zn-64	2560.0	4268.2	67
Zr-94	6260.0	15401.0	146
Zr-96	338.0	387.2	15
Mo-98	241.0	319.5	33
Mo-100	672.0	899.5	34
Au-197	5.7	5.7	0
Th-232	54.4	74.4	37
U-238	16.9	18.2	7.7

A similar analysis was performed for  $Q(\alpha)$ . Exact values calculated by direct integration of equations (22) and (23) were compared to the approximate ones based on equation (38), using  $Q_0$  and average  $E_r$  calculated as described before. For easier comparison between different nuclides all values were normalised with the corresponding  $Q_0$  value. The comparison is shown in Figure 3. Fortunately it turns out that the dependence of  $Q(\alpha)$  on  $E_r$  is rather weak. Although the  $\alpha$ -dependence of  $E_r$  is quite strong, the use of the average value in conjunction with equation (38) does not introduce a large error into the calculated  $Q(\alpha)$ . The differences are larger for nuclides with a higher effective resonance energies  $E_r$  and may exceed 3% in

some cases. It is interesting to note that equation (38) always leads to the underprediction of  $Q(\alpha)$ .

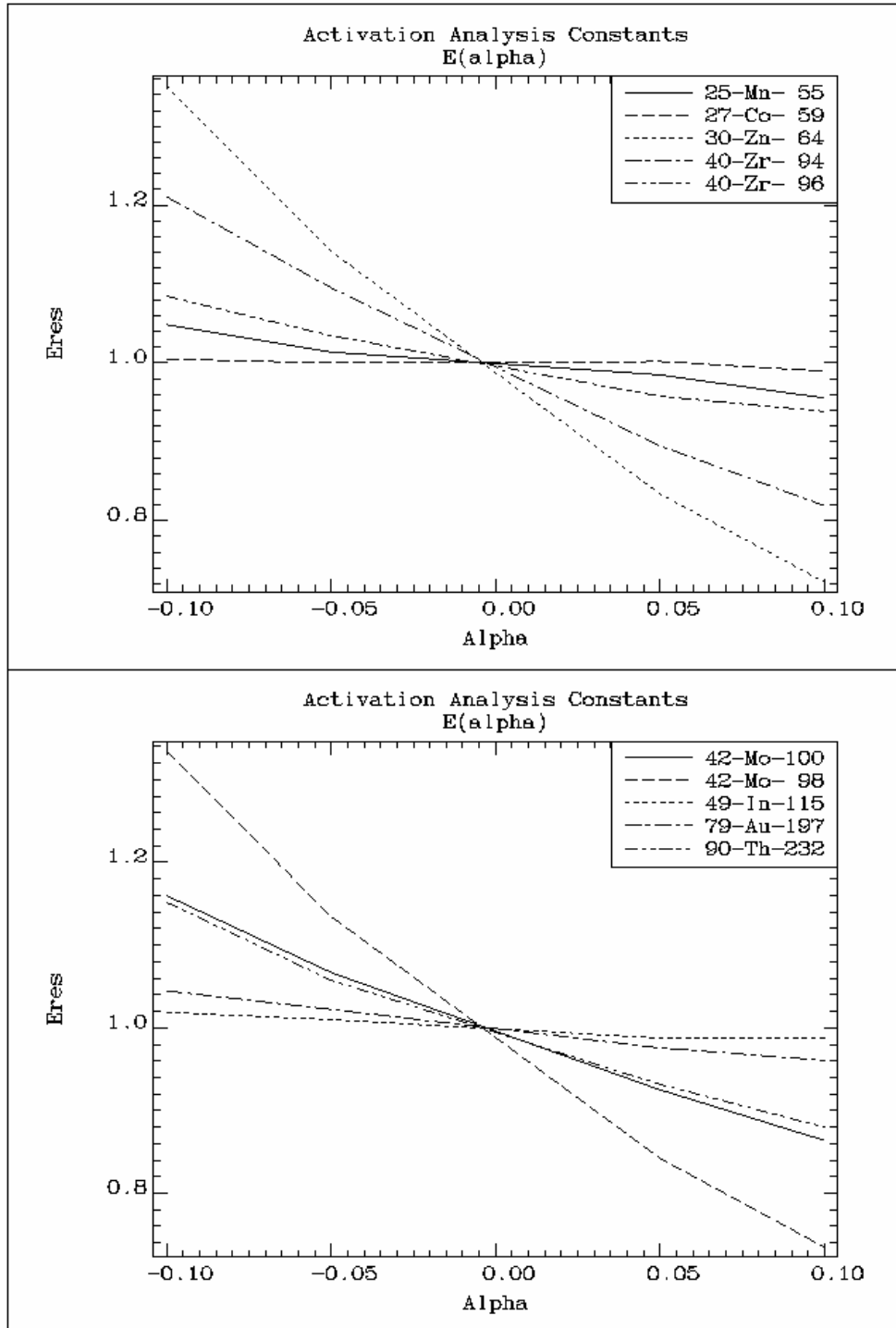


Figure 2: Variations of  $E_r$  with  $\alpha$  for different nuclides.



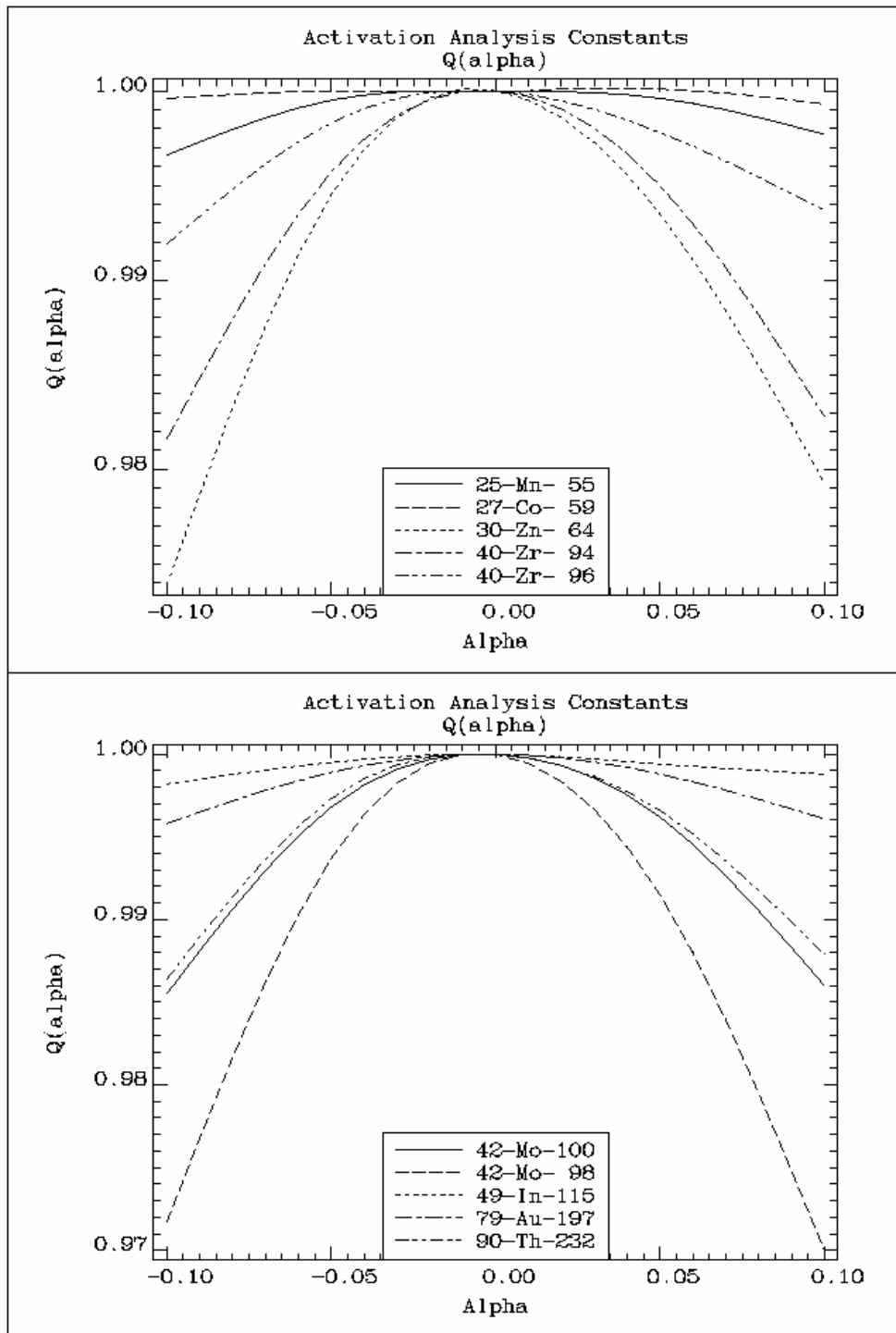


Figure 3: Comparison of exact and approximate  $Q(\alpha)$  values for different nuclides.

## 2.7 Fission Spectrum Contribution to Reaction Rate

Mathematically the fission spectrum integral is easy to define:

$$J = \int_{E_{cd}}^{\infty} \sigma(E) \varphi_h(E) dE = \frac{1}{h} \int_{E_{cd}}^{\infty} \sigma(E) [\varphi_r(E) + h \varphi_h(E)] dE - \frac{1}{h} \int_{E_{cd}}^{\infty} \sigma(E) \varphi_r(E) dE \quad (39b)$$

The fission spectrum fraction  $h$  depends on the spectrum normalisation. Once the fission spectrum is defined, the fission spectrum integrals can readily be calculated from the cross sections. Unfortunately this factor cannot be determined easily from the capture measurement, but we can make use of some threshold reactions for this purpose. An equation analogous to (6) can be written for threshold reactions:

$$\frac{A_a}{A_s} = k_{0,a} \frac{I_a + J_a h}{G_t f g_s + G_{f,s} Q_s + H_s h} \quad (39c)$$

where

$$k_{0,a} = \frac{M_s \Theta_a \gamma_a}{M_a \Theta_s \gamma_s \sigma_{0,s}} \quad (39d)$$

and the constants have their usual meaning. Note that in the above expression there is no thermal component. There might be a small contribution to the “resonance integral” for reactions with threshold below the upper cutoff energy for the resonance integral. Note also that the  $k_0$  factor is defined without the cross section in the numerator, because it is zero for threshold reactions.

The quantities in the denominator of the expression for  $k_0$  refer to the standard and are well known. The largest uncertainty probably originates from the gamma emission probability of the threshold reaction product. However, if threshold reactions are only needed to determine interference lines in the spectra, their uncertainties are less important.

## 2.8 Neutron Spectrum

Neutron spectrum in thermal reactors is determined by the fission neutron source, the slowing-down process at intermediate energies, and the thermal region where neutrons are in equilibrium with the surrounding crystal lattice. It is useful to define an analytical function that is representative of the general features of the spectrum. Typical light water reactor spectrum, which can be used as a weighting function for averaging cross sections and calculating reaction rates is approximated by the thermal Maxwellian part  $\psi_t$ , the epithermal region  $\psi_e$  and the fission spectrum  $\psi_f$  defined by:

$$\begin{aligned} \psi_t &= C_t E \left[ e^{-E/kT} + C_{t1} e^{-E/kT_1} + C_{t2} e^{-E/kT_2} \right] \\ \psi_e &= E^{-[1+\alpha_0+\alpha_1 \log E+\alpha_2 (\log E)^2]} \end{aligned} \quad (40)$$

$$\psi_f = C_f e^{-E/W} \sinh(\sqrt{EW_b}) \cdot \frac{1}{E^m} \quad \text{or}$$

$$C_f \sqrt{E} e^{-E/E_T} \cdot \frac{1}{E^{m_0+m_1 E}}$$

where:

- $k$  is the Boltzman constant,
- $\alpha_j$  are the constants that determine deviation from the  $1/E$  spectrum in the epithermal range,
- $W_a, W_b$  are the parameters of the Watt fission spectrum,
- $T$  is the temperature,
- $C_t, C_f$  are continuity constants such that  $\psi_t(E_t) = \psi_e(E_t)$  and  $\psi_f(E_f) = \psi_e(E_f)$ , respectively,
- $E_t, E_f$  are breakpoints between thermal, epithermal and the fast spectrum range,
- $m$  fast neutron slowing-down factor (equals 0 for no slowing-down and 1 for fast neutron sources surrounded by a moderator). It may vary linearly with energy, in which case the coefficients  $m_0$  and  $m_1$  are defined.

The full function  $\psi$  representing the spectrum is defined by:

$$\psi = K_t \psi_t + K_e \psi_e + K_f \psi_f \quad , \quad (41)$$

where

$$K_e = \begin{cases} 1 & \text{for } E_t < E < E_f \\ 0 & \text{otherwise} \end{cases}$$

$$\begin{aligned} K_t &= 1 + O_t - K_e \\ K_f &= 1 + O_f - K_e. \end{aligned}$$

Parameters  $O_t$  and  $O_f$  can be chosen arbitrarily to define overlap for a smooth transition between different regions (0 for no overlap, about 1 for a moderate overlap, typically).

The weighting function thus defined gives the spectrum shape with the required characteristics and a smooth transition between the thermal, epithermal and the fast energy range. It is equal to 1 at energy  $1 \text{ eV}$ .

Special features of the function are:

- Thermal region is a superposition of three Maxwellian functions at different temperatures, which allows one to model distortions in the spectrum at low energies.
- The  $\alpha$ - parameter that measures the deviation from the  $1/E$  shape is allowed to be energy-dependent.
- Fast fission spectrum can be the Maxwellian or the Watt function, with a correction to account for fast-neutron slowing-down.

## 3 Determination of parameters

### 3.1 Parameters determined from evaluated cross section data

As already mentioned, some of the parameters are difficult to determine experimentally. Having precise definitions of the constants from first principles allows us to calculate them from the basic nuclear data (particularly the energy-dependent cross sections in evaluated nuclear data files) at least in cases, when the parameters do not depend on the absolute accuracy of the cross section values but mainly on the shape in a particular energy range. Examples of such parameters are:

- $g$  generalised g-factor defined by equation (20) is usually very close to one. It is very difficult to design an experiment for a direct measurement of the g-factor that would be more accurate than the value calculated from cross section data, except perhaps in cases where the g-factor differs strongly from one. The ratio of the g-factors of the measured material to the standard can be determined from the measurements of the  $k_0$  factors in a thermal and a cold neutron beam (see next section below), knowing that at lower neutron energies the deviations of the cross sections from the  $1/v$  behaviour is smaller. The precondition for such measurement is good knowledge of the shape of the spectra. Generally it is preferable to use measurements of this kind for the validation of the g-factors calculated from the cross sections rather than their direct determination.
- $F_{cd}$  cadmium transmission factor defined by equation (29) accounts for the difference between the idealised and the measurable resonance integral, taking explicitly into account the interference between the absorber and the cadmium resonances. It would be possible to choose a definition of the resonance integral that would more closely match the measured one, but this would only obscure its subsequent application in the calculation of reaction rates. Defining the cadmium transmission factor to correct for the difference between the measured and the idealised resonance integral in a  $1/E$  spectrum between 0.55 eV and 2 MeV is a practical convenience.
- $G_f$  epithermal self-shielding factor accounts for detailed changes in the spectrum due to resonance absorption. Theoretical approach defined by equations (31) to (36) is well established in reactor physics, and gives good results even for absorbers in relatively high concentrations. There is no reason to question its applicability in NAA, where the levels of self-shielding are usually lower. Accurate direct measurements were reported for some monitor materials by irradiating samples of various thicknesses and extrapolating to zero, but such a procedure is not practical for general implementation to all materials that may occur in real samples; it may serve well for the validation of the resonance self-shielding factors calculated from the cross sections.
- $E_r$  effective resonance energy.
- $\sigma_h$  cross section in the fission spectrum energy range.

Contrary to the constants listed above, the thermal “self-shielding” (or flux depression) factor  $G_t$  is not a property of the measured nuclei but of the matrix in which they are embedded. The presence of strong absorbers may cause flux depression (and hence a decrease in the reaction rate), while abundance of organic materials may actually increase the thermal flux locally due to internal moderation. Empirical expressions for the calculation of  $G_t$  are described in the literature.

### 3.2 Parameters of the analytical spectrum function

If no additional information is available, the user can assume that the spectrum is purely Maxwellian with strength defined by factor  $f$  at thermal energies and deviates by a fraction  $\alpha$  from  $1/E$  in the resonance range up to 2 MeV, say. This is consistent with the traditional approach in NAA.

Rapid advances in computational power made possible the development of detailed full-core models (including irradiation facilities), with which the neutron spectrum can be calculated. A Monte Carlo simulation of the spectrum in the central channel of the TRIGA Mark-II reactor in Ljubljana is presented in Figure 4, labelled “Modulated fitting function”. Note the structure in the spectrum below the fission peak, which is due to the oxygen resonances.

The calculated spectrum was fitted with parameter  $\alpha$  having linear dependence, thermal spectrum given by parameter  $f$  with a small contribution of a secondary Maxwellian at 600 K to fit the shape at about 0.2 eV ...

The analytic function fit reproduces very well the overall shape of the spectrum, but not the fine details. To remedy this we define the *modulating function* as the ratio of the calculated spectrum with the analytic function. Obviously, scaling the analytic function with the modulating function reproduces exactly the calculated spectrum. Modulation of the analytic fitting function may be suppressed in the regions where the detailed shape of the calculated spectrum is unreliable (for example, below 0.4 eV and above 4 MeV).

If for some reason we need to change slightly the fitting function parameters, the overall characteristics of the spectrum change, but the detailed shape defined by the modulating function is preserved. The curve in Figure 4 labelled “Fit RR (All)” was obtained by allowing parameters  $f$ ,  $\alpha$  and  $m$  to vary so as to reproduce better the measured reaction rates of dosimetry monitors.

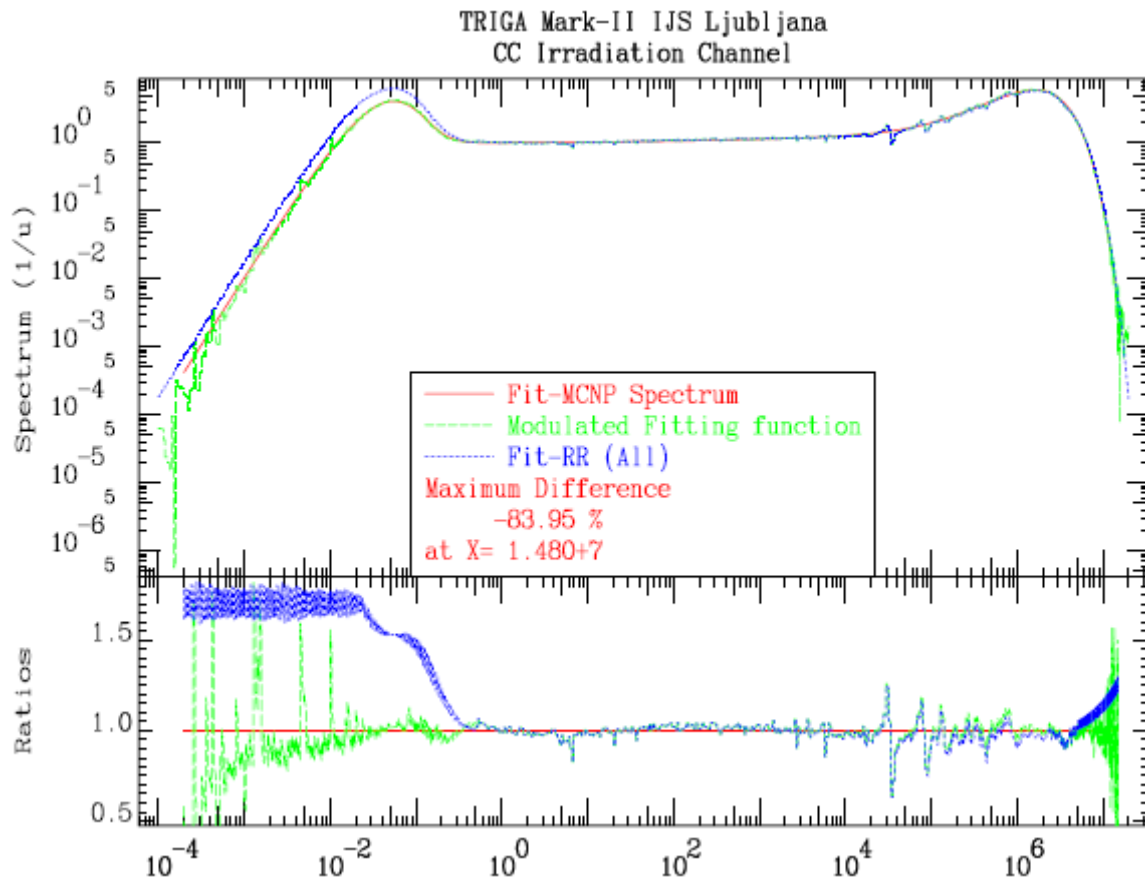


Figure 4: Calculated spectrum fitted with analytical spectrum function.

## 4 Experimental Measurements

### 4.1 $k_0$ measurements, thermal capture cross section and gamma emission probability

From equations (6) and (7) it follows that the  $k_0$  factor can be determined from the measured ratio of activities of the nuclide of interest (subscript  $a$ ) and the standard (subscript  $s$ ):

$$k_{0,a} = \frac{M_s \Theta_a P_{\gamma,a} \sigma_{0,a}}{M_a \Theta_s P_{\gamma,s} \sigma_{0,s}} = \frac{A_a}{A_s} \cdot \frac{G_t f g_s + G_{f,s} Q_s + H_s h}{G_t f g_a + G_{f,a} Q_a + H_a h}. \quad (42)$$

The accuracy of the measured  $k_0$  factor depends on the neutron spectrum in which the measurement is done. If the epithermal spectrum contribution is small, the  $f$  factor is large, making the contributions of the  $Q$  and  $H$  terms negligible. The only parameter influencing the result in addition to the measured ratio of specific activities is the ratio of the g-factors of the measured nuclide and the standard.

The measured  $k_0$  factor is proportional to the ratio of the partial gamma production cross sections  $\sigma_\gamma$ , defined by the product of the gamma emission probability  $P_\gamma$  and the capture cross section  $\sigma_0$ :

$$\frac{\sigma_{\gamma,a}}{\sigma_{\gamma,s}} = \frac{P_{\gamma,a} \sigma_{0,a}}{P_{\gamma,s} \sigma_{0,s}} = k_{0,a} \frac{M_a \Theta_s}{M_s \Theta_a}. \quad (43)$$

Partial gamma production cross sections can be used in combination with other experiments to determine the thermal cross section and the gamma emission probabilities. This possibility was generally not exploited, except in a few cases where the experimentalists explicitly reported the derived cross section values in the publication [6]. A more rigorous effort in this direction was made in the re-evaluation of the thermal capture cross section of  $^{238}\text{U}$ , where all available measurements of the cross sections, partial cross sections (including  $k_0$  values) and directly-measured gamma emission probabilities were analysed simultaneously by a generalised least squares procedure, taking correlations into account whenever possible [7]. This method yields a self-consistent set of cross sections, gamma emission probabilities, their uncertainties and correlations.

### 4.2 $Q_0$ measurements by the cadmium ratio method

The cadmium ratio is defined by the ratio of bare and cadmium covered reaction rates:

$$R_{cd} = \frac{A}{A_{cd}} = \frac{\phi_t \sigma_0 g G_t + \phi_f (I G_f + J h)}{\phi_f (I_{cd} G_f + J h)} = \frac{\phi_t \sigma_0 g G_t + \phi_f (I G_f + J h)}{\phi_f (I F_{cd} G_f + J h)}, \quad (44)$$

from which it follows that

$$R_{cd} = \left[ \frac{f g G_t + Q G_f + H h}{Q G_f F_{cd} + H h} \right]; \quad Q = \left[ \frac{g f}{R_{cd} F_{cd} - 1} \right] \frac{G_t}{G_f} - \frac{(R_{cd} - 1) H h}{(R_{cd} F_{cd} - 1) G_f}. \quad (45)$$

The first term in the expression for  $Q$  is well known in the literature on neutron activation analysis. The second term represents the correction for the fission spectrum contribution and vanishes if either the fission spectrum integral or the fission spectrum contribution tends to zero.

The reference  $Q_0$  can be obtained through relation derived from equation (38):

$$Q_0 = \left[ Q + \frac{0.429}{(2\alpha + 1)(0.55)^\alpha} \right] (E_r)^\alpha + 0.429 \quad (46)$$

The only assumption in this definition is that parameters  $F_{cd}$  and  $G_f$  are approximately independent of  $\alpha$  and that equation (38) adequately describes the dependence of  $Q$  on  $\alpha$ .

It is important to consider error propagation, which originates from the uncertainty  $\Delta f$  in the measured value of  $f$ ,  $\Delta R_{cd}$  in the measured cadmium ratio,  $\Delta h$  in the fission spectrum contribution and  $\Delta H$  in the fission spectrum integral. Note however, that the problem is ill-posed when the measured cadmium ratio is close to one.

$$\Delta Q = \frac{g}{R_{cd} F_{cd} - 1} \left[ \Delta f + \frac{F_{cd} \Delta R_{cd}}{R_{cd} F_{cd} - 1} \right] \frac{G}{G} + \frac{(R_{cd} - 1)}{(R_{cd} F_{cd} - 1) G_f} [H \Delta h + h \Delta H] \quad (47)$$

Different sets of evaluated nuclear data files were processed to obtain constants for a number of nuclides that are commonly used as monitors to determine the spectral parameters. Label EAF-99 refers to the European Activation Library, RNAL to the Reference Neutron Activation Library, JENDL/D-99 to the Japanese activation library and FENDL/A-2 to the FENDL-2 activation library. The Nudat results were retrieved from the IAEA web page "<http://www-nds.iaea.org/>". The corresponding values commonly applied in NAA are labelled Kayzero. The results are compared in Table 2. The column "+/- %" gives the specified uncertainty while the columns labelled "Dif [%]" give the percent difference from the Nudat values.

Comparison of the  $Q_0$  values shows that the Kayzero data for important monitor reactions agree very well with the Nudat values, except for zirconium. There is also good agreement with the values derived from evaluated data files. Particularly, excluding  $^{55}\text{Mn}$  and  $^{94}\text{Zr}$ , the EAF-99 values of  $Q_0$  are practically always within the experimental uncertainty of the Nudat values.

**Table 2: Comparison of  $Q_0$  nuclear constants for neutron activation analysis.**

	Kayzero +/- %		Nudat +/- %		EAF-99	Dif. [%]	RNAL	Dif. [%]	JENDL/D99	Dif. [%]	FENDL	Dif. [%]
Mn-55	1.053	2.6	1.053	3.7	0.845	-19.7	1.110	5.5	0.845	-19.7	1.110	5.5
Co-59	1.993	2.7	1.990	2.9	1.930	-3.1	1.893	-4.9	1.934	-2.8	1.925	-3.3
Zn-64	1.908	4.9	1.908	7.0	1.933	1.3	1.923	0.8	-	-	1.922	0.7
Zn-68	3.190	1.4	-	-	3.632	-	3.634	-	-	-	-	-
Zr-94	5.306	3.3	4.609	9.6	6.743	31.6	6.747	46.4	-	-	6.743	46.3
Zr-96	251.6	0.97	231.4	10.5	242.1	4.4	-	-	-	-	242.1	4.6
Mo-98	53.10	6.3	53.08	9.4	54.01	1.7	54.06	1.9	-	-	54.01	1.8
Mo-100	18.84	4.3	18.84	5.6	19.65	4.1	19.46	3.3	-	-	19.40	3.0
Au-197	15.71	1.8	15.71	1.9	15.48	-1.5	15.46	-1.6	15.45	-1.6	15.44	-1.8
Th-232	11.53	3.6	11.5	4.3	11.1	-4.1	-	-	10.9	-5.4	10.9	-5.5
U-238	103.4	1.3	103.4	1.8	100.0	-3.4	-	-	99.6	-3.6	99.2	-4.0

### 4.3 Determination of spectral parameters

The spectral parameters in NAA are mainly the spectral ratio  $f$  and the spectrum slope parameter  $\alpha$ . Equivalent spectral parameters implied by equation (40) are the energy breakpoint  $E_t$  between the thermal and the epithermal spectrum and the  $\alpha_j$  parameters. Note that  $\alpha$  is allowed to be energy-dependent, parameterised by second-order polynomial coefficients  $\alpha_0$ ,  $\alpha_1$  and  $\alpha_2$  in  $\log(E)$  domain. Normally the nuclear constants for NAA are not very sensitive to the other parameters that appear in equation (40).

Traditionally, the spectral ratio  $f$  is determined from the cadmium ratio of the gold standard, but measured cadmium ratios of other nuclides may be used as well. Similarly, the  $\alpha$  parameter (assumed constant) can be determined from a linear fit in the log-log scale of  $H_{(j)}$  as a function of  $\alpha$  for several monitor nuclides ( $j$ ) where  $H_{(j)}$  is given by:

$$H_{(j)}(\alpha) = \left\{ \frac{G_e}{G_t} (R_{cd(j)} - 1) Q_{(j)} (\bar{E}_{r(j)})^\alpha \right\}^{-1} \quad (48)$$

The fission spectrum fraction can be determined from reaction rates sensitive to the fission spectrum using equation (6). The obvious candidates are threshold reactions that also have well-defined constants for the capture process, which may serve as a secondary standard. The expression for the fission fraction is:

$$h = \frac{G_t f \left[ g_a k_0 - g_s \frac{A_a}{A_s} \right] + G_{f,a} Q_a k_0 - G_{f,s} Q_s \frac{A_a}{A_s}}{H_s \frac{A_a}{A_s} - H_a k_0} \quad (49)$$

The expression for the threshold  $k_0$  factor defined by equation (39d) is applicable and the standard in this case is a capture reaction for one of the isotopes of the same element. The above expression becomes much clearer if we note that for a threshold reaction  $g_a$  is zero by definition,  $Q_a$  is (close to) zero and  $A_a/A_s$  and  $H_s$  are usually small:

$$h = \frac{A_a}{A_s} \frac{G_t f g_s + G_{f,s} Q_s}{H_a k_0} \quad (50)$$

Note that equation (50) is given for clarity only. In practical calculations there is little penalty for using the full expression of equation (49).

Alternatively, the spectral parameters can be determined directly by minimising  $\chi^2$ , which is defined as the sum of the squares of the relative differences between the measured and calculated reaction rate ratios or specific activities. Reaction rate ratios can be calculated from energy-dependent cross sections and the parameterised neutron spectrum. The GRUPINT code was developed for this purpose: it calculates reaction rates by direct integration of the product of the differential cross sections with the parameterised neutron spectrum and performs  $\chi^2$ -minimisation by a systematic variation of the spectral parameters.



The additional advantage of direct fitting of reaction rates is that non-linear behaviour of parameters  $\alpha$  and  $m$  can easily be accommodated.

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