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Lawrence Livermore Laboratory

ABSOLUTE INSTRUMENTAL NEUTRON ACTIVATION ANALYSIS AT LAWRENCE LIVERMORE LABORATORY

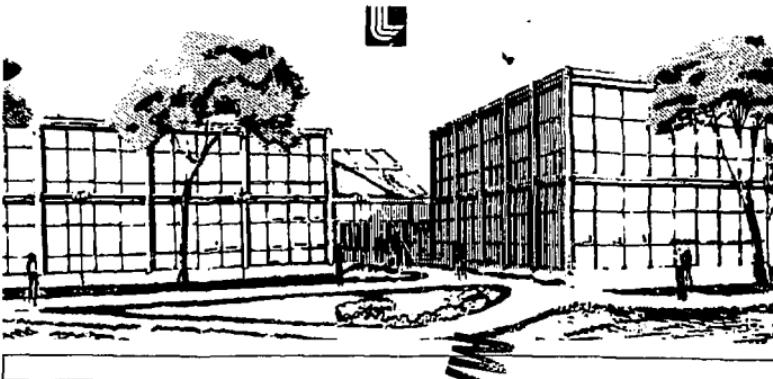
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Environmental Sciences Division

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Absolute Instrumental Neutron Activation Analysis at
Lawrence Livermore Laboratory

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ABSTRACT

The Environmental Science Division at Lawrence Livermore Laboratory has in use a system of absolute Instrumental Neutron Activation Analysis (INAA). Basically, absolute INAA is dependent upon the absolute measurement of the disintegration rates of the nuclides produced by neutron capture. From such disintegration rate data, the amount of the target element present in the irradiated sample is calculated by dividing the observed disintegration rate for each nuclide by the expected value for the disintegration rate per microgram of the target element that produced the nuclide.

In absolute INAA, the expected value for disintegration rate per microgram is calculated from nuclear parameters and from measured values of both thermal and epithermal neutron fluxes which were present during irradiation. Absolute INAA does not depend on the concurrent irradiation of elemental standards but does depend on the values for thermal and epithermal neutron capture cross-sections for the target nuclides.

A description of the analytical method is presented.

INTRODUCTION

Using the element cobalt and its neutron capture product ^{60}Co as examples, the basic equation for absolute INAA can be expressed as follows:

$$\text{Micrograms Cobalt} = \frac{\text{Measured DPM } ^{60}\text{Co}}{\text{Expected DPM } ^{60}\text{Co}/\mu\text{g cobalt}} \quad (1)$$

The numerator on the right side of the equation is determined by analyzing the gamma spectrum of an irradiated sample to find absolute disintegration rates for each of the contributing nuclides. At LLL this analysis is carried out by a computer program GAMANAL which was developed by R. Gunnink and J. Niday. GAMANAL is described in detail

photon energy and source geometry. The spectrum consists of 4096 channels at 0.5 Kev per channel. In the spectral analysis, GAMANAL determines the energies and absolute intensities of the gamma rays by determining a background and fitting peaks using a prescribed peak shape function. Complex groups are resolved by a least squares fitting method. At this point the spectrum has been reduced to a set of gamma ray energies and total intensities. The next step is to interpret these energies and intensities in terms of disintegration rates of specific nuclides.

Interpretation by GAMANAL is a two step process. First, a tentative identification of the radionuclides present is made by comparing the observed energies with energies listed in a decay scheme library. The library contains half-life and parent-daughter relationships for each nuclide and branching intensity of its gamma rays. GAMANAL uses energy, half-life vs sample age and associative gammas to generate a list of candidate nuclides which could have produced the set of observed gamma rays. In the final step, a matrix of equations is set up on the basis that each observed photopeak is a linear combination of one or more components arising from the list of nuclides tentatively identified. A least-squares solution of the set of equations produces the final result of the gamma spectrum analysis - a list of disintegration rates of specific nuclides.

In Figure 2, an abridged portion of the GAMANAL computer output is shown. This listing shows in some detail the basis for the disintegration rate assignments which were made. The second column of data in Figure 2, which is labeled "DPM", is actually the disintegration rate corrected to zero time. The nuclide labeled "HX" in column 1 is a dummy nuclide inserted in the decay scheme library to provide for the 511 Kev annihilation peak. Data for this peak are not used in the subsequent analysis.

In Figure 3, a second abridged portion of the GAMANAL output is shown. The values listed in the column headed "DPM AT COUNT TIME", together with nuclide identification, "PCT ERROR", time of start of count and length of count are put out in a data file format and are used as input to the NADAC analysis code.

NADAC COMPUTATIONS

A particular radionuclide may be produced both by decay of a radioactive parent and directly by neutron capture. This commonly occurs where both a ground state and a metastable isomer are formed during irradiation. The equations used to determine the relationship between the observed disintegration rate and the amount of element present in the sample are:

$$K_1 = \frac{T_p}{T_p - T_D} + \frac{T_p}{T_D},$$

$$K_2 = 1 - \frac{T_p}{T_p - T_D},$$

(9)

where T_p and T_D are half-lives of parent and daughter, respectively. DPM = disintegration of daughter owing to parental decay, and DPM' = disintegration of daughter owing to independent yield.

The solutions to equations (2) through (4) are:

$$DPM = fR_p (K_1 \alpha_p \beta_p + K_2 \alpha_D \beta_D) \text{ (via parent)}, \quad (10)$$

$$DPM' = R_D \alpha_D \beta_D \text{ (direct production)}. \quad (11)$$

The gamma detector measures the sum (DPMTOT) of these two disintegration rates. Hence the expected disintegration rate is:

$$DPMTOT = fR_p \left[K_1 \alpha_p \beta_p + \left(K_2 + \frac{R_D}{fR_p} \right) \alpha_D \beta_D \right]. \quad (12)$$

The α , β , and K terms in equation (12) depend only on time and half-life values which are known. To calculate DPMTOT we need the parent and daughter production rates, R .

It should be noted that equation (12) applies to the particularly complicated case of a daughter product with independent production of the daughter nuclide. Where the nuclide counted is directly produced, K_1 and K_2 become zero and equation (12) goes to:

$$DPMTOT = R \cdot \alpha \cdot \beta \quad (13)$$

which is the form we will use in the following discussion for simplicity of notation.

Rate of Nuclide Production During Irradiation

In a reactor, the neutron flux consists of three components: thermal neutrons, epithermal neutrons, and "high-energy" neutrons. The rate of nuclide production by (n, γ) or (n, f) reactions depends on thermal and epithermal fluxes. The rate of production by (n, p) or (n, α) reactions depends on the high-energy flux.

^{239}U based on thermal cross-sections and thermal flux equal to unity, we can solve the following pair of equations for the epithermal and thermal flux values.

$$\frac{(\text{DPM } ^{46}\text{Sc}/\mu\text{g})\text{OBSVD}}{(\text{DPM } ^{46}\text{Sc}/\mu\text{g})\text{thermal}} = \phi_{\text{TH}} + .44 \phi_{\text{EPI}} \quad (17)$$

$$\frac{(\text{DPM } ^{239}\text{U}/\mu\text{g})\text{OBSVD}}{(\text{DPM } ^{239}\text{U}/\mu\text{g})\text{thermal}} = \phi_{\text{TH}} + 102.9 \phi_{\text{EPI}} \quad (18)$$

The epithermal/thermal flux ratio observed for 3 irradiation positions used at the Livermore reactor are .0195, .039, .048.

Calculation of Total Micrograms

In the operation of NADAC a library of nuclear parameters is used. Stored parameters include parent and daughter half-lives and production rate values. These parameters together with the appropriate time information and disintegration rate data provided by GAMANAL are used to calculate "W" the total micrograms of element for each nuclide found. Combining equations (12) and (15) we have

$$\text{DPMTOT} = R_0 \times \alpha \times \beta \times \phi_{\text{TH}} \times W \quad (19)$$

In effect, DPMTOT/W is the expected disintegration rate of equation (1) hence

$$W(\mu\text{g}) = \frac{\text{DPM(GAMANAL)}}{R_0 \times \alpha \times \beta \times \phi_{\text{TH}}} \quad (20)$$

Correction for Interfering Reactions

The nuclide ^{27}Mg is produced both by (n, γ) reaction on ^{26}Mg and by (n, p) reaction on ^{27}Mg :

$$\begin{aligned} \text{DPMTOT}(\text{Mg}^{27}) = & \left[R_0(\text{Mg}^{26}) \phi_{\text{TH}} \cdot \alpha \cdot \beta \cdot W(\text{Mg}) \right] + \\ & \left[R_1(\text{Al}^{27}) \phi_{\text{TH}} \cdot \alpha \cdot \beta \cdot W(\text{Al}) \right] . \end{aligned} \quad (21)$$

We want to determine $W(\text{Mg})$, the net micrograms of Mg in the sample:

$$W(\text{Mg}) = \left[\frac{\text{DPMTOT}(\text{Mg}^{27})}{R_0(\text{Mg}^{26}) \phi_{\text{TH}} \cdot F(t)} \right] - \left[\frac{R_1(\text{Al}^{27})}{R_0(\text{Mg}^{26})} \right] W(\text{Al}) \quad (22)$$

The (n, p) production rate R_1 , introduced in Eq. (21) above, is determined for each reactor region by irradiating Al in that region. The ratio $R_1(\text{Al}^{27})/R_0(\text{Mg}^{26})$ is stored in the nuclear parameter library.

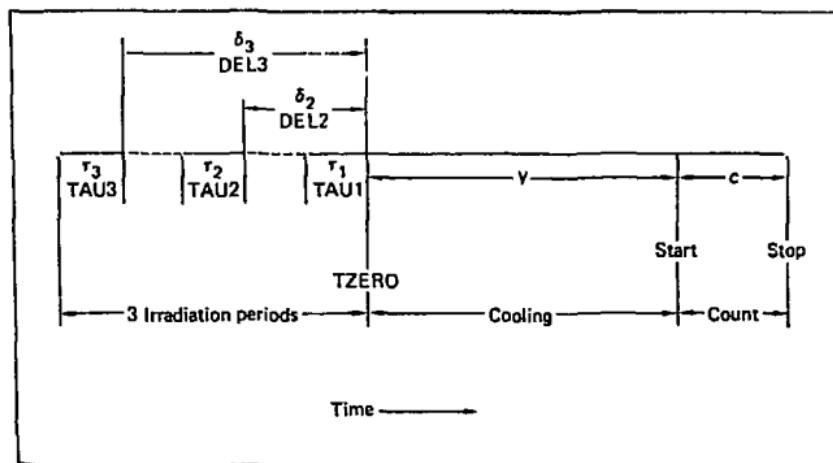
unknowns. In the absolute method the principle problem is to determine accurate values for all the nuclear parameters that enter into the computation. Basically, the absolute method is simpler and versatile, however, especially for the less common nuclides, the nuclear parameters reported by various workers do exhibit some fairly broad variation (and hence uncertainty). We have irradiated standard samples of the elements in various reactor regions. The gamma spectrometric data from these irradiations are being used to check branching intensity and half-life values. The disintegration rate data are being used to check thermal and epithermal cross-section values. Literature values of σ_{TH} and RI are summarized by R. Sher³ and H. Albinsson⁴ in the IAEA Handbook on Nuclear Activation Cross Sections. We plan to report the results of this work upon completion.

REFERENCES

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3. R. Sher, "2200 m/s Neutron Activation Cross Section," in Handbook on Nuclear Activation Cross Sections, International Atomic Energy Agency, Vienna, Austria, Technical Report Series No. 156 (1974).
4. H. Albinsson, "Infinite Dilution Resonance Integrals," in Handbook on Nuclear Activation Cross Sections, International Atomic Energy Agency, Vienna, Austria, Technical Report Series No. 156 (1974).

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IRRADIATION, COOLING, AND COUNTING TIMES



GE GAMMA ANALYSIS OF ST0261ST11 20000

DATE OF CALCULATION

EXPERIMENT NO. 2

11:33:06 4/13/77

SAMPLE NUMBER 00

ZERO TIME 83.529

SAMPLE WEIGHT = 1.000E+00

MIDTIME OF COUNT 97.731

SPECTRUM NO 097301

NORMALIZATION WEIGHT = 1.000E+00

DECAY TIME IS 14.208 DAYS

DETECTOR T SYSTEM 3

NORMALIZATION FACTOR = 1.000E+00

LIVE-TIME OF COUNT (TAKEN FROM CHANNEL 11) = 333.33 MINS

96BY33 / 94BY29

GEOMETRY IS 4.09 CM

LIVE-TIME OF COUNT (TAKEN FROM CHANNEL 11) = 333.33 MINS 96BY33 / 94BY29

** MLR LEAST-SQUARES RESULTS **

| NUCLIDE | DPM AT COUNT TIME | DPM AT ZERO TIME | ATOMS AT ZERO TIME | PCT. ERROR | SET NO. | QFIT | IDENTIFICATION | CONFIDENCE VALUE |
|---------|-------------------|------------------|--------------------|------------|---------|------|----------------|------------------|
| H X | 1.362E+03 | 1.408E+03 | 8.771E+08 | 20.2 | 1 | 1.0 | 0.46 | |
| SC 46 | 2.978E+04 | 3.349E+04 | 5.835E+09 | 0.9 | 2 | 1.2 | 0.97 | |
| CR 51 | 5.196E+04 | 7.413E+04 | 4.268E+09 | 2.1 | 3 | 1.0 | 0.75 | |
| FE 59 | 3.965E+03 | 4.945E+03 | 4.581E+08 | 3.3 | 2 | 1.2 | 1.00 | |
| CO 58 | 4.744E+03 | 5.447E+03 | 8.057E+08 | 2.7 | 4 | 1.0 | 0.62 | |
| CO 60 | 6.442E+03 | 6.475E+03 | 2.585E+08 | 1.8 | 2 | 1.2 | 0.93 | |
| ZN 65 | 8.441E+03 | 8.787E+03 | 4.471E+08 | 3.4 | 2 | 1.2 | 0.68 | |
| SE 75 | 9.327E+02 | 1.012E+03 | 2.523E+08 | 3.9 | 2 | 1.2 | 1.00 | |
| SR 85 | 2.055E+03 | 2.390E+03 | 3.236E+08 | 14.8 | 5 | 1.0 | 0.56 | |
| AG 410 | 2.200E+03 | 2.287E+03 | 1.202E+09 | 4.0 | 2 | 1.2 | 1.00 | |
| CD 115 | 2.151E+03 | 1.861E+05 | 8.534E+08 | 5.0 | 2 | 1.2 | 0.98 | |
| SB 122 | 8.124E+03 | 2.880E+05 | 1.651E+09 | 3.2 | 2 | 1.2 | 0.89 | |
| SB 124 | 5.456E+03 | 6.424E+03 | 8.045E+08 | 1.8 | 2 | 1.2 | 1.00 | |
| CS 134 | 1.487E+04 | 1.507E+04 | 2.332E+10 | 1.1 | 2 | 1.2 | 1.00 | |
| BA 131 | 4.896E+03 | 1.112E+04 | 2.772E+08 | 1.7 | 2 | 1.2 | 1.00 | |
| LA 140 | 9.145E+02 | 3.271E+05 | 1.130E+09 | 5.7 | 2 | 1.2 | 0.98 | |
| CE 141 | 3.257E+03 | 4.428E+03 | 2.978E+08 | 3.0 | 2 | 1.2 | 0.73 | |
| SM 153 | 3.168E+03 | 5.076E+05 | 2.045E+09 | 4.6 | 2 | 1.2 | 0.81 | |
| EU 152 | 4.813E+02 | 4.823E+02 | 5.126E+09 | 19.1 | 2 | 1.2 | 0.94 | |
| YR 159 | 9.200E+02 | 1.269E+03 | 8.092E+07 | 6.9 | 2 | 1.2 | 1.00 | |
| YB 175 | 6.894E+03 | 7.152E+04 | 6.254E+08 | 8.5 | 2 | 1.2 | 0.99 | |
| TA 182 | 9.169E+02 | 9.909E+02 | 2.306E+08 | 11.4 | 2 | 1.2 | 1.00 | |

FIG 5

PAGE 45

1 FINAL SUMMARY REPORT

187006

SAMPLE CODE

RC0078

DESCRIPTION

FC031MPC MK3025 06C 04 09:13 760218

SAMPLE AMOUNT

8.940E+00 SCF

CONTAINING MATERIALS

2.181E+01 SG CM COATED KENFOL

NOMINAL FLUX

2.846E+13

+ 9.500E+01 MG

POLY BAG

| NUCLIDE | ELEMENT | TOTAL UO | SD | MATERIALS | BLANK | UG | SD | N,F OR N,P | BLANK | UG | SD | NET UD/ | SCF | SD | FSD |
|---------|---------|-----------|-----------|-----------|-----------|----|-----------|------------|-----------|----|----|-----------|-----------|-----------|-----|
| 11024 | NA | 3.349E+02 | 5.694E+00 | 1.661E+00 | 1.103E-01 | 0. | 0. | 0. | 0. | 0. | 0. | 3.720E+01 | 6.370E-01 | 1.709E-02 | |
| 19042 | K | 1.576E+02 | 4.036E+01 | 6.542E-01 | 1.247E-01 | 0. | 0. | 0. | 0. | 0. | 0. | 1.754E+01 | 5.514E-01 | 2.574E-01 | |
| 21046 | SC | 3.571E-01 | 3.545E-03 | 5.011E-01 | 3.653E-05 | 0. | 0. | 0. | 0. | 0. | 0. | 3.989E-02 | 3.966E-04 | 9.943E-03 | |
| 23051 | FE | 1.946E+02 | 6.813E-02 | 4.057E-02 | 2.742E-03 | 0. | 0. | 0. | 0. | 0. | 0. | 1.261E-01 | 7.626E-03 | 3.583E-02 | |
| 26069 | Cr | 2.100E+02 | 5.457E+00 | 2.388E+00 | 2.693E-03 | 0. | 0. | 0. | 0. | 0. | 0. | 6.171E+01 | 1.188E+00 | 1.405E-03 | |
| 27060 | Co | 4.666E-01 | 9.372E-03 | 7.632E-04 | 2.672E-04 | 0. | 0. | 0. | 0. | 0. | 0. | 0.233E-02 | 1.049E-03 | 2.004E-02 | |
| 30065 | ZN | 1.190E+01 | 3.688E-01 | 1.093E-01 | 2.020E-02 | 0. | 0. | 0. | 0. | 0. | 0. | 1.311E+00 | 4.131E-02 | 3.133E-02 | |
| 31072 | GA | 3.576E+00 | 1.430E-01 | 0. | 0. | 0. | 0. | 0. | 0. | 0. | 0. | 4.000E-01 | 1.600E-02 | 4.000E-02 | |
| 33075 | AS | 9.200E-01 | 3.000E-02 | 0. | 0.889E-03 | 0. | 0.911E-04 | 0. | 0. | 0. | 0. | 2.052E-01 | 4.575E-02 | 1.600E-02 | |
| 34075 | SE | 4.099E-01 | 3.103E-02 | 0. | 0. | 0. | 0. | 0. | 0. | 0. | 0. | 1.052E-01 | 3.775E-02 | 1.000E-02 | |
| 37085 | RB | 1.073E+00 | 1.008E-01 | 0. | 0. | 0. | 0. | 0. | 0. | 0. | 0. | 1.205E-01 | 1.128E-02 | 9.400E-02 | |
| 38085 | SR | 1.124E+01 | 2.366E+00 | 0. | 0. | 0. | 0. | 0. | 0. | 0. | 0. | 1.287E+00 | 2.639E-01 | 2.100E-01 | |
| 40093 | ZR | 1.008E-01 | 2.478E+00 | 0. | 0. | 0. | 0. | 0. | 0. | 0. | 0. | 7.773E-01 | 2.713E-01 | 3.489E-01 | |
| 42099 | RD | 1.837E+00 | 2.196E-01 | 0. | 0.035E-03 | 0. | 1.410E-01 | 0. | 0. | 0. | 0. | 3.368E-01 | 2.163E-02 | 9.560E-02 | |
| 51122 | SB | 4.102E-01 | 1.034E-02 | 0. | 7.384E-04 | 0. | 7.731E-04 | 0. | 0. | 0. | 0. | 4.509E-02 | 1.157E-03 | 2.525E-02 | |
| 51124 | SB | 4.235E-01 | 1.143E-02 | 0. | 7.384E-04 | 0. | 7.731E-04 | 0. | 0. | 0. | 0. | 4.728E-02 | 1.279E-03 | 2.705E-02 | |
| 55134 | CG | 8.308E-02 | 6.978E-03 | 0. | 0. | 0. | 0. | 0. | 0. | 0. | 0. | 8.293E-03 | 7.808E-04 | 8.400E-02 | |
| 55131 | BA | 6.524E-01 | 1.449E+00 | 0. | 0.625E-02 | 0. | 0.882E-03 | 0. | 0. | 0. | 0. | 9.931E+00 | 1.621E-01 | 1.701E-01 | |
| 56140 | UF | 6.440E-01 | 2.544E-01 | 0. | 0. | 0. | 0. | 0. | 0. | 0. | 0. | 7.201E-02 | 2.845E-02 | 3.950E-01 | |
| 57140 | LA | 1.565E+00 | 2.204E-02 | 0. | 0.932E-04 | 0. | 1.216E-04 | 0. | 0. | 0. | 0. | 1.772E-01 | 2.466E-03 | 1.392E-02 | |
| 58141 | CG | 9.130E-02 | 6.897E-02 | 0. | 0. | 0. | 0. | 0. | 0. | 0. | 0. | 3.306E-01 | 7.741E-03 | 3.341E-02 | |
| 58147 | SD | 9.109E-01 | 6.193E-02 | 0. | 0. | 0. | 0. | 0. | 0. | 0. | 0. | 1.232E-01 | 9.563E-03 | 7.763E-01 | |
| 62153 | SD | 9.109E-01 | 6.193E-02 | 0. | 0. | 0. | 0. | 0. | 0. | 0. | 0. | 1.232E-01 | 9.563E-03 | 7.763E-01 | |
| 63152 | EU | 4.480E-02 | 1.914E-03 | 0. | 0.620E-04 | 0. | 0.401E-05 | 0. | 0.800E-05 | 0. | 0. | 2.339E-02 | 1.193E-02 | 9.300E-02 | |
| 63452 | EU | 5.597E-02 | 1.004E-02 | 0. | 0. | 0. | 0. | 0. | 0. | 0. | 0. | 0.805E-03 | 1.458E-03 | 5.022E-01 | |
| 70180 | YB | 8.465E-02 | 4.287E-02 | 0. | 0. | 0. | 0. | 0. | 0. | 0. | 0. | 1.061E-02 | 4.945E-02 | 4.500E-02 | |
| 70179 | YB | 9.050E-02 | 4.720E-03 | 0. | 0. | 0. | 0. | 0. | 0. | 0. | 0. | 1.061E-02 | 2.795E-04 | 3.00E-02 | |
| 71177 | LU | 7.365E-02 | 1.239E-03 | 0. | 0. | 0. | 0. | 0. | 0. | 0. | 0. | 1.643E-03 | 1.402E-04 | 1.300E-02 | |
| 72178 | HF | 1.780E-01 | 8.651E-03 | 0. | 0. | 0. | 0. | 0. | 0. | 0. | 0. | 0.002E-02 | 7.327E-03 | 3.660E-01 | |
| 72181 | HF | 5.333E-01 | 5.632E-03 | 0. | 0. | 0. | 0. | 0. | 0. | 0. | 0. | 2.609E-02 | 6.523E-04 | 2.500E-02 | |
| 73182 | TA | 8.935E-02 | 6.205E-03 | 0. | 0. | 0. | 0. | 0. | 0. | 0. | 0. | 6.639E-03 | 3.588E-04 | 3.400E-02 | |
| 74181 | TA | 6.937E-02 | 6.625E-03 | 0. | 1.235E-03 | 0. | 1.729E-04 | 0. | 0. | 0. | 0. | 5.019E-02 | 7.746E-03 | 1.404E-01 | |
| 74183 | TA | 6.658E-02 | 6.658E-03 | 0. | 0. | 0. | 0. | 0. | 0. | 0. | 0. | 7.495E-02 | 9.689E-04 | 3.300E-02 | |
| 83239 | CF | 6.274E-01 | 2.451E-02 | 0. | 0.235E-04 | 0. | 0.627E-04 | 0. | 0. | 0. | 0. | 7.008E-02 | 2.741E-03 | 3.912E-01 | |

NOTICE

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in UCRL-51061¹, and will be discussed briefly below.

The denominator term on the right side of equation (1) is determined by calculating the radionuclide production rate by both thermal and epithermal neutrons, and correcting for radioactive growth and decay processes as required by the irradiation, cooling, and counting times. The computations are carried out by means of a computer program, NADAC, developed by R. Heft and W. Martin and is described in detail in UCRL-52249². A brief description of the mathematical treatment used in NADAC is given below.

The overall analysis is carried out in the following manner. The sample to be analyzed is irradiated for a measured interval of time. The irradiation package includes dual flux monitor consisting of two nuclides. For one nuclide the ratio of the epithermal neutron capture cross-section to the thermal neutron capture cross-section must be small (e.g. ^{45}Sc with a ratio equal to 0.44. For the other nuclide the ratio must be high (e.g. ^{238}U with a ratio equal to 102.9). Following a cooling period, the gamma spectral analysis is started and continued for a measured real time interval. The gamma spectrum is analyzed by GAMANAL and the output listing of disintegration rates at counting time for the nuclides found is used as input to NADAC. A separate spectral analysis of the dual flux monitor is made in order to determine both thermal and epithermal neutron flux values. These and the required irradiation, cooling, and counting time values are also provided to NADAC which combines the experimental data with stored values of the nuclear parameters and computes total micrograms for each element, corrected as required for interference by (n,p) or (n,f) reactions. NADAC uses an absolute computation method and provides a correct calculation of elemental abundance in all cases including:

- Counting time is long compared to half-life of radionuclide counted.
- Sample is subjected to multiple or interrupted irradiations.
- Nuclide counted is daughter of nuclide produced, including the case where daughter is also produced directly.

GAMANAL COMPUTATIONS

In Figure 1, the time information required for the absolute computation is shown schematically. The time at which the last irradiation was completed (TZERO) is taken as the basis for decay calculations in order to simplify the growth-decay equations required in the calculations. The counting interval "C" is the real time of the count as opposed to the live-time used in calculating disintegration rates. The real time is needed to permit accurate decay calculations when the half-life of nuclide counted is short compared to the counting interval.

All of the gamma spectrometer systems used in this program have been calibrated to give absolute photons per minute as a function of

$$\text{Parent} \quad \frac{dN_p}{dt} = R_p - \lambda_p N_p \quad , \quad (2)$$

$$\text{Daughter} \quad \frac{dN_D}{dt} = f \lambda_p N_p - \lambda_D N_D \quad , \quad (3)$$

$$\text{Independent} \quad \frac{dN_D^1}{dt} = R_D - \lambda_D N_D^1 \quad , \quad (4)$$

where:

N_p = no. of atoms of parent nuclide.

N_D = no. of atoms of daughter nuclide produced by decay.

N_D^1 = no. of atoms of independently produced daughter nuclide.

R_p = parent production rate (atoms/min).

R_D = independent production rate of daughter (atoms/min).

λ_p = decay constant of parent (min^{-1}) ($= \ln 2/T_p$).

λ_D = decay constant of daughter (min^{-1}) ($= \ln 2/T_D$).

f = fraction of parent decays leading to formation of daughter.

T_p, T_D = half-lives of parent and daughter nuclides.

The relationship needed is the solution to these equations for a generalized set of irradiation, cooling, and counting times. The times involved are illustrated in Fig. 1.

In the diagram, three irradiation periods, τ_1 , τ_2 , and τ_3 are indicated. Zero time (TZERO) is set to equal the time of completion of the final irradiation, and cooling time (y) and counting time (c) are as shown. To simplify writing solutions to the equations, we introduce the following notation:

$$F(a, b, x) = (1 - e^{-ax})e^{-bx} \quad , \quad (5)$$

$$a_i = F(\tau_3, \delta_3, \lambda_i) + F(\tau_2, \delta_2, \lambda_i) + F(\tau_1, 0, \lambda_i) \quad , \quad (6)$$

$$b_i = F(c, y, \lambda_i)/\lambda_D c \quad , \quad (7)$$

where $i = P$ or D .

The rate of production by (n, γ) or (n, f) reaction is given by:

$$R = 3.614 \times 10^6 \left[(\phi_{TH} \times \sigma_{TH}) + (\phi_{EPI} \times RI) \right] \frac{W \cdot G}{M} . \quad (14)$$

where:

R = production rate of (n, γ) or (n, f) product in atoms/min.

$3.614 \times 10^6 = 60 \times 10^{19} \times$ Avogadro's number.

ϕ_{TH} = thermal flux in units of 10^{13} neutrons/cm²/sec.

ϕ_{EPI} = epithermal flux in units of 10^{13} neutrons/cm²/sec.

σ_{TH} = thermal-neutron cross section in barns.

RI = epithermal-neutron cross section in barns.

G = isotopic abundance of target nuclide in percent.

M = atomic weight of target element in grams.

W = mass of target element in micrograms.

We define:

$$R_0 = 3.614 \times 10^6 \left[\sigma_{TH} + \left(\frac{\phi_{EPI}}{\phi_{TH}} \right) RI \right] \frac{G/M}{M} \left(\frac{\text{atoms}}{(\text{min})(\mu\text{g})} \right) , \quad (15)$$

so that:

$$R = R_0 (\phi_{TH}) W .$$

The parameter R_0 is a function of constants except for the ratio ϕ_{EPI}/ϕ_{TH} , which varies with the location within the reactor. However, within a particular location, the ratio remains reasonably constant, and thus R_0 can be treated as a constant for that location. Values of R_0 for the individual nuclides can either be determined directly for a particular reactor region by irradiating elemental standards or they can be calculated from thermal and epithermal cross-sections and the epithermal/thermal flux ratio. The flux ratio can be determined by irradiating a dual flux monitor consisting of two nuclides which have widely different epithermal/thermal cross-section ratios. This ratio for ^{45}Sc is .44 and for ^{238}U is 102.9. Hence if we irradiate a Sc/U standard and calculate expected disintegration rates for ^{46}Sc and

The total correction is computed by NADAC by multiplying this ratio by the weight (micrograms) of aluminum after W has been computed for each element present. If Al is found, a Mg correction is made.

In samples where the Si:Al ratio is high, the Mg results will be incorrect, because ^{28}Al is formed both by (n, γ) on ^{27}Al and by (n, p) reaction on ^{28}Si . Therefore, the micrograms of Al used in Eq. (22) is high, the correction for Al (n, p) interference in the Mg determination is overstated, and the Mg result is low. NADAC has a built-in provision for modifying the Mg correction to allow for the Si contribution to the Al assay.

A second kind of interference reaction is the fission of ^{235}U by thermal and epithermal neutrons. The nuclide ^{95}Zr is produced both by fission of ^{235}U and by (n, γ) on ^{94}Zr :

$$\text{DPM}(FISS) = R_0(FISSION) \cdot \epsilon_{TH} \cdot \alpha \cdot B \cdot W(U) \cdot FY(95\text{Zr}), \quad (23)$$

where $FY(95\text{Zr})$ is the thermal fission yield of ^{95}Zr .

$$\text{DPM}(n, \gamma) = R_0(n, \gamma) \cdot \epsilon_{TH} \cdot F(t) \cdot W(Zr).$$

The observed disintegration rate for ^{95}Zr is the sum of these two reactions, so the total micrograms of Zr is:

$$\begin{aligned} \text{UGTOT}(Zr) \equiv W(Zr) = & \left[\frac{\text{DPMTOT}(95\text{Zr})}{R_0(n, \gamma) \cdot \epsilon_{TH} \cdot \alpha \cdot B} \right] - \\ & \left[\frac{R_0(FISSION) \cdot FY(95\text{Zr})}{R_0(n, \gamma)} \right] W(U). \end{aligned} \quad (25)$$

The ratio $R_0(FISSION) \cdot FY(95\text{Zr}) / R_0(n, \gamma)$ is stored in the nuclear parameter library.

When the irradiated sample has been counted more than once, a weighted mean for the total micrograms of each element found is calculated before the interference blank correction is made. NADAC also makes provision for correcting for the contribution to total micrograms by packaging materials used. In Figures 4 and 5, abridged portions of NADAC computer output are shown to illustrate the data handling for a sample which was counted twice.

CONCLUSION

INAA can be carried out on a comparative basis or on an absolute basis. In the comparative method standards for the elements sought are irradiated and counted along with the samples to be analyzed. Problems that may arise are that standard solutions may be unstable and that the spectral resolution may be different for standards and

Figure Captions

Figure 1 Irradiation, Cooling, and Counting Times

Figure 2 GAMANAL Computer Output Part 1

Figure 3 GAMANAL Computer Output Part 2

Figure 4 NACAC Computer Output Part 1

Figure 5 NADAC Computer Output Part 2

** DETAILED MLR RESULTS ** (BASED ON LIBRARY FILE:ACTL15377)

EVALUATION OF INDIVIDUAL CONTRIBUTIONS AT ZERO TIME FOR PEAKS GREATER THAN 50.0 KEV

| NUCLIDE | DPM | ERRSR | GAMMA ENERGY | DPM | FRACTION OF PEAK | PEAK ERROR (STD DEVS) | RESIDUAL (STD DEVS) | INTERFERENCES PRESENT (AND OTHERS CONSIDERED) |
|---------|-----------|-------|--------------|-----------|------------------|-----------------------|---------------------|---|
| H X | 1.408E+03 | 20.2 | 510.9 | 1.408E+03 | 1.000 | 20.2 | 0.0 | |
| SC 46 | 3.349E+04 | 0.9 | 889.1 | 3.377E+04 | 0.992 | 1.2 | 0.7 | (TRIVIAL: TE132) |
| | | | 1120.4 | 3.321E+04 | 0.998 | 1.2 | -0.7 | PLUS: TA182 |
| CR 51 | 7.413E+04 | 2.1 | 320.1 | 7.413E+04 | 1.000 | 2.1 | 0.0 | |
| FE 59 | 4.945E+03 | 3.3 | 149.6 | 4.945E+03 | 0.021 | 2.6 | -0.0 | PLUS: CE141 YB176 |
| | | | 192.4 | 4.823E+03 | 0.025 | 21.6 | -0.1 | |
| | | | 336.3 | 4.967E+03 | 0.009 | 4.9 | 0.1 | PLUS: CD115 |
| | | | 1999.1 | 4.964E+03 | 0.998 | 4.2 | 0.1 | |
| | | | 1291.5 | 4.931E+03 | 0.993 | 4.2 | -0.1 | PLUS: TA182 (TRIVIAL: TE132) |
| CO 60 | 8.447E+03 | 2.7 | 810.6 | 5.446E+03 | 1.000 | 2.7 | -0.0 | (TRIVIAL: TE132) |
| | | | 883.6 | 1.273E+04 | 0.428 | 100.0 | 0.6 | |
| CO 60 | 8.475E+03 | 1.8 | 311.9 | 6.423E+03 | 0.010 | 3.5 | 0.6 | PLUS: PA233 |
| | | | 1179.1 | 6.631E+03 | 0.973 | 2.2 | 1.2 | (TRIVIAL: TE132) |
| ZN 65 | 8.787E+03 | 3.4 | 1302.4 | 8.360E+03 | 1.016 | 1.9 | -1.0 | |
| | | | 1115.3 | 8.787E+03 | 0.984 | 3.0 | -0.0 | PLUS: EU152 (TRIVIAL: TA182) |
| SE 75 | 1.012E+03 | 3.9 | 98.5 | 9.941E+02 | 0.042 | 4.9 | -0.4 | PLUS: SM153 PA233 (REJECTED: QD153) PLUS: EU132 |
| | | | 121.3 | 9.039E+02 | 0.604 | 11.8 | -1.0 | |
| | | | 136.1 | 1.022E+03 | 0.990 | 4.0 | 0.2 | (REJECTED: RE168) PLUS: YB169 TA162 |
| | | | 198.1 | 9.542E+02 | 0.039 | 9.3 | -0.7 | PLUS: LA140 TA182 |
| | | | 264.7 | 1.036E+03 | 0.914 | 7.2 | 0.3 | (TRIVIAL: TE132) (REJECTED: BA140) |
| | | | 279.7 | 1.002E+03 | 1.011 | 15.7 | -0.1 | (REJECTED: HG209) |
| | | | 400.4 | 8.951E+02 | 0.821 | 36.5 | -1.6 | PLUS: SB124 PA233 |
| SR 85 | 2.390E+03 | 14.8 | 614.0 | 2.390E+03 | 1.000 | 14.8 | 0.0 | (TRIVIAL: YB169) |
| AB 410 | 2.287E+03 | 4.0 | | | | | | |

1 CALCULATION OF WEIGHTED MEAN OF TOTAL MICROGRAMS
 SAMPLE CODE 187006 RCD07B

| NUCLIDE | *****1 | *****2 | WT. MEAN | PCNT SD | N |
|----------|-----------|-----------|-----------|-----------|---|
| 11024 NA | 3.349E+02 | 0. | 3.349E+02 | 1.700E+00 | 1 |
| | 5.694E+00 | 0. | 5.694E+00 | | |
| 19042 K | 1.876E+02 | 0. | 1.876E+02 | 2.560E+01 | 1 |
| | 4.036E+01 | 0. | 4.036E+01 | | |
| 21046 SC | 3.756E-01 | 3.568E-01 | 3.571E-01 | 9.928E-01 | 2 |
| | 3.118E-02 | 3.568E-03 | 3.545E-03 | | |
| 24051 CR | 0. | 1.846E+00 | 1.846E+00 | 3.500E+00 | 1 |
| | 0. | 6.813E-02 | 6.813E-02 | | |
| 26059 FE | 0. | 7.328E+02 | 7.328E+02 | 1.400E+00 | 1 |
| | 0. | 1.026E+01 | 1.026E+01 | | |
| 27080 CO | 0. | 4.866E-01 | 4.866E-01 | 2.000E+00 | 1 |
| | 0. | 9.072E-03 | 9.372E-03 | | |
| 00065 ZN | 0. | 1.190E+01 | 1.190E+01 | 3.100E+00 | 1 |
| | 0. | 3.688E-01 | 3.688E-01 | | |
| 31072 DA | 3.576E+00 | 0. | 3.576E+00 | 4.000E+00 | 1 |
| | 1.430E-01 | 0. | 1.430E-01 | | |
| 33076 AS | 2.560E+00 | 0. | 2.560E+00 | 1.600E+00 | 1 |
| | 4.095E-02 | 0. | 4.095E-02 | | |
| 34075 SE | 0. | 9.408E-01 | 9.408E-01 | 3.300E+00 | 1 |
| | 0. | 3.105E-02 | 3.105E-02 | | |
| 37086 RB | 0. | 1.073E+00 | 1.073E+00 | 9.400E+00 | 1 |
| | 0. | 1.008E-01 | 1.008E-01 | | |
| 38085 SR | 0. | 1.124E+01 | 1.124E+01 | 2.100E+01 | 1 |
| | 0. | 2.380E+00 | 2.380E+00 | | |
| 40095 ZR | 0. | 1.088E+01 | 1.088E+01 | 2.230E+01 | 1 |
| | 0. | 2.426E+00 | 2.426E+00 | | |
| 42099 MO | 2.071E+00 | 1.820E+00 | 1.871E+00 | 5.813E+00 | 2 |
| | 4.660E-01 | 1.238E-01 | 1.156E-01 | | |
| 51122 SB | 4.138E-01 | 4.094E-01 | 4.102E-01 | 2.522E+00 | 2 |
| | 2.400E-02 | 1.148E-02 | 1.034E-02 | | |
| 51124 SB | 0. | 4.235E-01 | 4.235E-01 | 2.700E+00 | 1 |
| | 0. | 1.143E-02 | 1.143E-02 | | |
| 55134 CS | 0. | 8.308E-02 | 8.308E-02 | 8.400E+00 | 1 |
| | 0. | 8.978E-03 | 8.978E-03 | | |
| 56131 BA | 0. | 8.524E+01 | 8.524E+01 | 1.700E+00 | 1 |
| | 0. | 1.449E+00 | 1.449E+00 | | |
| 56140 UF | 0. | 6.440E-01 | 6.440E-01 | 3.950E+01 | 1 |