Nuclear Inst. and Methods in Physics Research, A 908 (2018) 155–158

Contents lists available at ScienceDirect

Nuclear Inst. and Methods in Physics Research, A

journal homepage: www.elsevier.com/locate/nima

Technical Notes

New method for measuring the time integral of neutron flux in a reactor

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ARTICLE INFO

Keywords:
Neutron flux
Gamma detection
^{58}Ni(n,γ) reaction
^{59}Ni(n,γ) reaction
HPGe

ABSTRACT

A new method for measuring the time integral of thermal neutron flux of a nuclear reactor is proposed. This method utilizes two consecutive (n,γ) reactions on the ^{58}Ni isotope of a natural nickel sample placed near the core of a reactor, ^{58}Ni(n,γ)→^{59}Ni(n,γ)→^{60}Ni(n,γ). Natural nickel contains 68% ^{58}Ni producing an intense 8.999 MeV γ-line leading to the ^{59}Ni ground state via the ^{58}Ni(n,γ) reaction. The ^{59}Ni isotope produced in this manner undergoes another (n,γ) reaction, emitting a γ-line at 11.388 MeV leading to the ^{60}Ni ground state. By measuring the intensity ratio of the two γ-lines at 8.999 and 11.388 MeV using a HPGe detector, it is possible to deduce the integral over time of the neutron flux necessary for producing the ^{59}Ni isotope. Good accuracy can be obtained with this method due to the low gamma background at such high energies.

In the present work, we propose a new method for measuring the time integral of thermal neutron flux in a nuclear reactor. This method involves neutron irradiation utilizing two consecutive (n,γ) reactions on the ^{58}Ni isotope of a natural nickel sample placed near the core of a reactor: ^{58}Ni(n,γ) → ^{59}Ni(n,γ) → ^{60}Ni(n,γ). Natural nickel contains 68% ^{58}Ni producing an intense 8.999 MeV γ-line through thermal neutron capture via the ^{58}Ni(n,γ) reaction leading to the ^{59}Ni ground state. This is illustrated in Fig. 1. The intensity of the 8.999 MeV line is proportional to the thermal neutron cross section and to the amount of ^{58}Ni occurring in the sample. The ^{59}Ni isotope produced in this manner undergoes a subsequent (n,γ) reaction, emitting a γ-line at 11.388 MeV leading to the ^{60}Ni ground state (Fig. 1).

Note that ^{59}Ni does not occur in natural nickel and has a very long lifetime of 76,000 years. Obviously, the intensity of the emitted 11.388 MeV gamma rays is also proportional to the amount of the ^{59}Ni isotope occurring in the nickel sample. Since the intensity of the 11.388 MeV line is zero at the beginning of n-irradiation of natural Ni and it increases with time upon breeding ^{59}Ni, it is thus a measure of the accumulated amount of ^{59}Ni in the sample. Hence by measuring the intensity ratio of the two γ-lines at 8.999 and 11.388 MeV emitted from a Ni sample placed near a reactor core, it is possible to deduce the integral over time of the neutron flux producing the ^{59}Ni isotope.

After an extended length of time of neutron irradiation, the accumulated amount of ^{59}Ni is high enough for revealing the 11.388 MeV line in the γ-spectrum of the ^{59}Ni(n,γ) reaction. This line is shown in Fig. 2 which also shows the intense 8.999 MeV γ-line related to that of the ^{58}Ni(n,γ) reaction; the corresponding spectrum was measured using a HPGe detector. In this specific case, the natural metallic Ni sample was placed along a tangential beam tube and near the core of the Israel Research Reactor-2 (IRR-2); it was irradiated by thermal neutrons for few years. Note that the intensity of the gamma line at 11.388 MeV resulting from the double neutron capture of ^{58}Ni is not weak and could be quite intense depending on the total influx of neutrons. In fact, this γ-line has been produced in the past in three nuclear centers and was used for γ-scattering measurements as was done at: IRR-2 [1], at Argonne National Laboratory, USA [2,3], and at ILL, Grenoble, France [4].

The γ-spectrum from the Ni(n,γ) reaction (Fig. 2), reveals apart from transitions to the ground states in ^{59}Ni, ^{60}Ni and other final isotopes of natural Ni, several other γ-lines originating from transitions to excited states in all final nuclei. It should be remarked that because the energies of the 8.999 and 11.388 MeV lines are so high, the contribution of the gamma background to this energy region is obviously quite small. This enables good experimental accuracy for the determination of the total neutron flux by measuring the gamma line intensity ratios of 8.999 and 11.388 MeV.

Note that this experimental method places some strict requirements in the sense that the Ni sample must be placed inside a tube which is *tangential* to the core of a reactor in a high neutron flux. Such an arrangement ensures that the Ni sample is viewed directly by the Ge detector. In such a geometry, the gamma background is relatively low and the (n,γ) spectrum from the Ni sample is expected to show up clearly. It should be noted in this connection that placing the sample along a beam tube *radial* to the reactor core should be avoided because the amount of gamma background emerging from the core is so huge that it overwhelms any signal emitted by the Ni(n,γ) reaction from the sample.

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https://doi.org/10.1016/j.nima.2018.08.045
Received 25 January 2018; Received in revised form 3 July 2018; Accepted 15 August 2018
Available online xxxx
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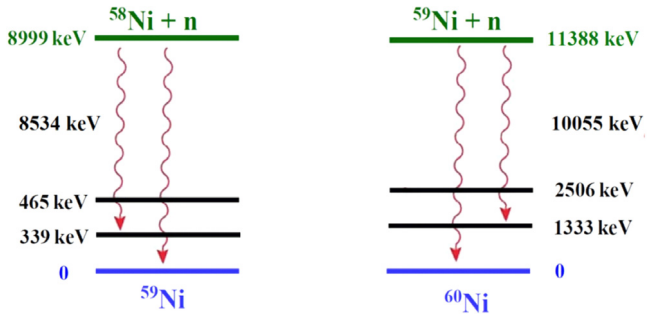

Fig. 1. Schematic view (not to scale) of the de-excitation of the n-capture states following the ^{58}Ni(n,γ), ^{59}Ni and ^{59}Ni(n,γ) ^{60}Ni reactions. The 8.999 MeV and 8.534 MeV γ-rays correspond to primary transitions to the ground state and the 0.465 MeV excited level in ^{59}Ni, while the 11.388 MeV and 10.055 MeV γ-rays correspond to primary transitions to the ground state and the 1.333 MeV excited level in ^{60}Ni.

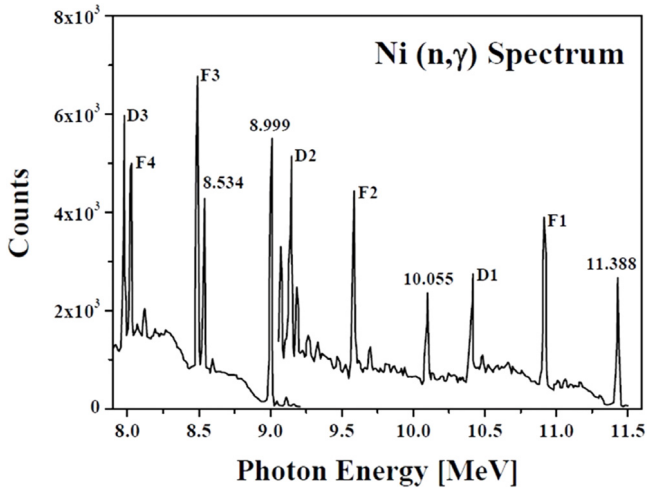

Fig. 2. The Ni(n,γ) spectrum obtained from a metallic nickel sample placed near the reactor core, and along a tangential beam tube of the IRR-2 reactor, using a 140 cm^3 HPGe detector. The γ-spectrum was measured after passing the beam through lead absorbers and about 20 cm of borated paraffin. This was necessary to cut down the gamma intensity to levels tolerated by the Ge detector. The n-irradiation time of the Ni sample was few years. Note that the 8.999 MeV and the 11.388 MeV γ-lines correspond to ground state transitions from the capture states in ^{59}Ni and ^{60}Ni while the 8.534 and 10.055 MeV correspond to inelastic transitions to excited states in those isotopes as indicated in Fig. 1. Note that only the energies of the photo peaks are indicated while the first escape and second escape peaks of the various lines are labelled by (F_i) and (D_i) (i = 1, 2, 3, 4). The scale of the right part of the spectrum, after the break at 9.1 MeV, corresponds to a maximum of 400 counts.

In the following we show how the intensity ratio of the lines at 9.0 and 11.4 MeV is calculated. As noted above, the ^{59}Ni is produced via the ^{58}Ni(n,γ) reaction; it partially decays with a half-life of 76,000 years via electron capture and partially lost by n-absorption through the ^{59}Ni(n,γ) reaction creating ^{60}Ni. Thus the creation rate of the ^{59}Ni isotope may be written as:

$$\frac{d}{dt}N_{59}(t) = N_{58}\sigma_{58}\Phi_{\rm th}(t) - \lambda_{59}N_{59}(t) - \sigma_{59}\Phi_{\rm th}(t)N_{59}(t) \quad (1)$$

where $N_{58} = MF_{58}N_A/58$, the number of atoms of ^{58}Ni isotope in the metallic nickel sample (with M, its mass—it is assumed that this number is practically constant during n-irradiation, being correct only for moderate and low neutron fluxes); F_{58} the weight fraction of the ^{58}Ni isotope; N_A, Avogadro's number; $N_{59}(t)$ - the number of atoms of the ^{59}Ni isotope, which is a function of time t and is deduced by solving Eq. (1) with the boundary condition $N_{59}(0) = 0$. We deal with long irradiation periods, where the thermal flux is expected to vary with time. $\Phi_{\rm th}(t)$ is the thermal neutron flux; σ_{58} and σ_{59} are the thermal neutron capture cross sections for the ^{58}Ni(n,γ) and ^{59}Ni(n,γ) reactions; λ_{59}, the decay constant of ^{59}Ni. This decay can be neglected because of the very long decay time of ^{59}Ni (~76 000 years). The third term is generally small (some numerical estimates are given below) and may be neglected. Hence, Eq. (1) may be written as:

$$\frac{d}{dt}N_{59}(t) = N_{58}\sigma_{58}\Phi_{\rm th}(t) \quad (2)$$

Thus the solution for the number of ^{59}Ni atoms is given by:

$$N_{59}(t) = N_{58}\sigma_{58}\int \Phi_{\rm th}(t)dt \quad (3)$$

If $\Phi_{\rm th}$ is assumed to be constant, then N_{59} will be linear in time t.

In a similar manner the creation rate of ^{60}Ni via the ^{59}Ni(n,γ) reaction may be written as:

$$\frac{d}{dt}N_{60}(t) = \sigma_{59}\Phi_{th}(t)\,N_{59}(t) - \sigma_{60}\Phi_{th}(t)\,N_{60}(t) \quad (4)$$

The second term on the right hand side produces ^{61}Ni. Natural Ni contains about 26.2% ^{60}Ni which is stable. In principle, if ^{61}Ni is to be accounted for, the above equation has to be solved with the initial condition: $N_{60}(0) = \frac{MF_{60}}{60}N_A$. For the present purpose, we are only interested in the rate of production of the ^{60}Ni nuclei at a given time t, that is $dN_{60}(t)/dt$. Thus as in Eq. (1), the second term in the right hand side is neglected, giving:

$$\frac{d}{dt}N_{60}(t) = \sigma_{59}\Phi_{th}(t)N_{59}(t) \quad (5)$$

Experimentally, the intensities of the specific gammas: 8.999 and 11.388 MeV emitted by the two isotopes ^{59}Ni and ^{60}Ni are measured (Fig. 2). These intensities can be calculated using the creation rates (i.e. the left sides of Eqs. (2) and (5)) because as soon as the nuclei are created, via neutron absorption, they de-excite immediately via γ-emission. The rate of creation $R = dN(t)/dt$ is the same as the rate of emission and the measured activity will be

$$A = GRf\varepsilon\Omega$$

where ε is the detector efficiency for the specific gamma ray energy, Ω, the solid angle subtended by the detector, and f is a fraction of the ground state transition intensity from the capture state relative to all γ-transition intensities to the ground and excited levels in the same isotope. G is a geometrical factor accounting for sample geometry and flux non-uniformity. More often than not, G corrects for the absorption in metal shielding, placed in front of the detector, for counting rate reduction. While the absorption coefficients at the two energies are different, for sufficiently thin absorbers the values of G in the two cases will be practically equal. We assume this to be the case for the following discussion. Hence the ratio R_A of the two gamma line activities (denoted $A_{11.388}$ and $A_{8.999}$) at a time t from the point in time when the Ni sample is placed for the first time in the neutron field is:

$$R_A = \frac{A_{11.388}(\Phi_{th},t)}{A_{8.999}(\Phi_{th},t)} = \sigma_{59}\frac{\varepsilon(11.388)}{\varepsilon(8.999)}\frac{f_{11.388}}{f_{8.999}}\int \Phi_{th}(t)dt. \quad (6)$$

The value of $\int \Phi_{\rm th}(t)dt$, being the integral of the thermal neutron flux over time t, may be determined from Eq. (6). Note also that for a constant $\Phi_{\rm th}$ a linear dependence of the ratio on t, is evident.

We illustrate the calculation of $\int \Phi_{\rm th}(t)dt$ for the case where numerical values were taken from experiment [5,6]; the efficiencies were calculated using polynomial parameters (given in Fig. 2a of Ref. [6] without uncertainties). From the discussion presented in Ref. [6], a global error of 2% is assumed for the fitted efficiencies, yielding a 3% error for the efficiencies ratio in Eq. (6).

The fractions $f_{8.999}$ and $f_{11.388}$ appearing in Eq. (6) were estimated using measured data taken from Ref. [5]: the value of σ_{58} in ^{58}Ni is 4.13 b while that corresponding to the 8.999 MeV γ-line is 2.082 b, yielding $f_{8.999} = 50.4\%$ with an error of 1.9%. Similarly, In Ref. [5] the thermal cross section σ_{59} is: 73.7 ± 1.8 b, i.e a 2.5% error while that corresponding to the 11.388 MeV γ-line is 21.5 b, hence $f_{11388} = 29.2\%$ with an error of 4.5%. Hence their ratio has a 4.9% error. Summing