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Nickel Foil as Transmutation Detector for Neutron Fluence Measurements

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Abstract. Activation detectors are very often used for determination of the neutron fluence in reactor dosimetry. However, there are few disadvantages concerning these detectors; it is the demand of the knowledge of the irradiation history and a loss of information due to a radioactive decay in time. Transmutation detectors TMD could be a solution in this case. The transmutation detectors are materials in which stable or long-lived nuclides are produced by nuclear reactions with neutrons. From a measurement of concentration of these nuclides, neutron fluence can be evaluated regardless of the cooling time.

1. Introduction

One task of reactor dosimetry is the determination of the neutron field characteristics like neutron spectrum and neutron fluence. The activation detector method (ADM) is very often used for that purpose. Small metallic foils are used as detectors for ADM. A choice of material depends on the duration of irradiation, the type of reaction and the expected neutron fluence value. For the AMD method, the reaction (n, γ), (n, α) and (n, p) are very often used. The favourite materials are Ti, Mn, Fe, Co, Ni, Cu, Nb, In and Au. The idea of AMD consists in the neutron capture reaction in material where unstable and gamma-emitting nuclides are produced. The activity of these nuclides is determined by standard gamma spectrometry and servers as input data for further evaluation. Some issues are caused by lack of information about irradiation conditions, especially, when the neutron fluence rate dependence on time or shape of neutron spectra are unknown. In that case approximation of input spectra has to be used for adjustment with computer codes. The activation method with input spectrum calculation permits neutron spectrum adjustment using more reactions with different cross-section energy dependencies and thresholds. The approximation of input spectrum, the unknown time dependence of neutron fluence rate and the loss of information due to decay are disadvantages of the method [1].

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The mentioned loss of information led to a question if there is a possibility to use other type of detection. The solution offered here are detectors based on such reactions that generate stable or nearly stable nuclides. The most convenient reactions are (n, γ), (n, α) and (n, 2n). The (n, p) reaction is not so frequent because of β^- decay to the original target nucleus. The measured value could be the mass concentrations of the produced nuclides. Unfortunately, the trace concentrations of the generated nuclides are so low that only a very sensitive analytical method has to be used. The mass spectrometry, neutron activation analysis and nuclear magnetic resonance are prospective analytical methods.

The aim of the presented experiment was to confirm the possibility of using a nickel foil as such transmutation detector. Two isotopes of nickel with sufficiently long half-lives are produced during neutron irradiation in nickel with a natural abundance. Those are ^{63}Ni and ^{59}Ni with half-lives of 100 and 7×10^4 years, respectively. The isotopes are produced in reactions ^{62}Ni(n, γ)^{63}Ni and ^{58}Ni(n, γ)^{59}Ni by thermal neutrons. Both of them emit only low energy beta particles. Thus, there is no possibility of detecting them by gamma spectrometry as in the case of activation detectors. Therefore, it is necessary to use other analytical method, which is sufficiently and selectively sensitive for isotopic concentration measurement.

2. Experimental Background

An experimental verification was carried out in Research Centre Řež (CV Řež) at the LVR-15 research reactor. Nickel foils were irradiated in a vertical irradiation channel of the research reactor LVR-15 for one reactor campaign (21 days). Together with the nickel foils, other material samples were placed inside the irradiation capsule. Many of them were used for neutron fluence evaluation by the classical activation method. A preliminary estimate of the quantities of isotopes was performed by the calculation. This way, the concentrations of emerging nuclides were determined after one reactor campaign, long neutron irradiation. Due to the low concentration of ^{59}Ni and ^{63}Ni, in a range of hundreds of ppm, the Prompt Gamma Neutron Activation Analysis (PGNAA) method has been chosen as the most appropriate for the concentration assessment.

The PGNAA method is based on a (n, γ) reaction and successive production of nuclei in excited state. Due to the transition to lower energy level, characteristic gamma quanta are emitted. They are called prompt gamma-rays and are detected by a usual HPGe detector. The gamma spectrum acquired by a standard multichannel analyser includes peaks with characteristic energy for every isotope occurring in the irradiated sample. A ratio of the peak-areas for different isotopes was used for the concentration calculations. More details about the concentration evaluation are described by Tomandl et al. in reference [2].

Since the expected concentration of the transmuted isotopes was very small (in ppm-range), high-intensity neutron flux was essential for the sample irradiation within reasonable time (about 8 hours). Therefore, the sample measurement was performed at the Forschungs-Neutronquelle Heinz Maier-Leibnitz (FRM II) in Garching, Germany.

From the obtained concentration values and the knowledge of the microscopic cross-section the neutron fluence value was acquired and compared with the value obtained from the measurement with activation detectors.

2.1 LVR-15 Measurements

The first attempts with TMD were done in our institute in the year 2009. The set of materials was continuously irradiated in the vertical channel F6 for the whole period of 21 days by the thermal reactor power of 9 MW and the neutron fluence rate of $3 \times 10^{14}\,\mathrm{cm}^{-2} \cdot \mathrm{s}^{-1}$. There were placed nickel and iron activation foils for neutron spectra unfolding and neutron fluence evaluation. The evaluation was calculated with program SAND II. Values of activity for the gamma-emitting nuclides and neutron

Table 1. Comparison of expected and measured concentration of nickel nuclides.

Target nuclide	Transmuted nuclide	Expected conc. (ppm)	Measured conc. (ppm)	Unc. (%)	Ratio Exp./Meas.
^{58}Ni	^{59}Ni	490	483	6	1.01
^{62}Ni	^{63}Ni	83	110	6	0.75

Table 2. Measured activity of nuclides like input data for ADM unfolding.

Reaction	Activity (Bq)	Unc. (%)
^{54}Fe(n,p)^{54}Mn	9.41E+05	1
^{58}Fe(n,γ)^{59}Fe	1.25E+07	2
^{58}Ni(n,p)^{58}Co	4.51E+07	1

spectrum in irradiation position were used as input data. The activity was measured in a spectrometric laboratory in CV Řež: the ^{54}Fe(n, p)^{54}Mn and ^{58}Fe(n, γ)^{59}Fe reactions for the iron detector, and ^{58}Ni(n, p)^{58}Co for nickel detector. The same nickel foil was later used like the transmutation detector for the PGNAA measurement performed at FRM II.

2.2 PGNAA Measurements

The PGAA instrument at FRM II is installed at the end of a 51-m long, curved cold-neutron beam guide NL4b. The thermal neutron flux can reach up to $6.1 \times 10^{10}\,\mathrm{cm}^{-2} \cdot \mathrm{s}^{-1}$ thermal equivalent. The rectangular cross-section of the neutron beam at the irradiation position was $12 \times 24\,\mathrm{mm}^2$. TMD samples in form of metallic foils were packed into thin Teflon foils and inserted into cold neutron beam at the distance of 33 cm from the HPGe detector.

The prompt and delayed spectra for the nickel foil were taken within the 2 measurement days. Prompt gamma lines from transmuted isotopes ^{59}Ni, ^{63}Ni and ^{64}Ni were observed in these spectra. Based on the counting rate of these gamma lines, the preliminary concentrations of these transmuted isotopes agree with expected concentrations. This comparison is given in Table 1. The TMD foils were irradiated by the thermal neutron flux of $4.0 \times 10^{10}\,\mathrm{cm}^{-2} \cdot \mathrm{s}^{-1}$ for the Ni foils. The gamma-ray spectra were acquired with a standard Compton-suppressed spectrometer consisting of an HPGe with relative efficiency of 60% and a BGO scintillator surrounding the HPGe detector crystal and connected in anti-coincidence mode [2].

3. Results

The unfolding of spectra was done with two types of detectors, activation detectors and transmutation detectors. The same input spectrum was used for neutron spectrum deconvolution or adjustment. The well-defined input spectrum, which is used for evaluation of neutron fluence measurements in horizontal irradiation channel, was calculated with the Monte Carlo transport code MCNP. The reaction rates were derived from the activity measurements of Fe and Ni foils in case of activation detector method. The derived reaction rates were used as input date for unfolding programme SAND II. The measured activity of nuclides is shown Table 2. It may be seen that the uncertainties have lower dispersion than for concentrations in Table 1. The main component of uncertainty is statistical error of peak area in spectra. The concentrations are related to the total number of nickel nuclei.

For transmutation detector method, the reaction rates were obtained from concentration measured at FRM II. The determination of concentration was carried out for two generated nickel nuclides ^{59}Ni and ^{63}Ni.

Table 3. Neutron fluence rate obtained with activation detector method (ADM) and with transmutation detectors (TMD).

Energy interval	Neutron fluence rate ($cm^{-2} \cdot s^{-1}$)		
	ADM	TMD	TMD/ADM
(0; 0.501 eV)	1.48E+14	1.08E+14	0.73
(0.501 eV; 10 keV)	4.45E+13	3.24E+13	0.73
(10 keV; 1 MeV)	1.07E+13	7.56E+12	0.71
(0.1 MeV; 20 MeV)	4.86E+13	3.14E+13	0.65
(0.5 MeV; 20 MeV)	3.58E+13	2.27E+13	0.64
(1.0 MeV; 20 MeV)	2.68E+13	1.67E+13	0.62

The spectrum unfolding was calculated with programme SAND II [3] for both types of methods. The standard deviation of measured-to-calculated reaction rates were 16% and 22% for ADM and TMD, respectively. The comparison for both methods is presented in Table 3. The comparison of neutron fluence rate shows a ratio of approximately 0.7, which could be caused by the concentration value of ^{63}Ni, at least insofar as the ratio between expected and measured concentration points.

4. Conclusions

The main aim of the experiment has been to show that it is possible to use transmutation detectors as an alternative or support for the activation foil detectors method. Although both nickel isotopes are produced by thermal neutrons, we can use them for demonstration because we have well-defined input spectrum for irradiation position. In case of unknown neutron spectrum, there should be used more transmutation detectors covering wider energy range. Despite of the relatively high discrepancy in evaluated neutron fluence rates, it seems that concentration instead of activity, or transmutation detectors instead of activation detectors, can be combined with unfolding code SAND II. The big advantage of TMD compared to AMD is no need for the knowledge about the irradiation history of the detector; in other words the information about irradiation is saved in the detector because of no short term radioactive decay. The transmutation detector method can be used for long-term neutron fluence measurement in nuclear fission reactors. The drawback of TMD is the trace concentrations of transmuted nuclides (hundreds of ppm) in the detectors and so the demand for a very sensitive analytical method for the concentration determination.

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