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Transmutation detectors for fast neutron fluence measurements

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Abstract. Transmutation detectors (TMDs) are based on a similar principle as neutron activation detectors (NADs). In both cases, the neutron fluence is estimated from the number of reactions in the detectors. The main differences between these two detectors are the type of product created by the nuclear reaction and the evaluation of the irradiated detectors. In TMD, the transmuted nuclide is stable, and the number of reactions is gained by analytical methods suitable for measuring low concentrations. In NAD, the nuclear reaction leads to the production of radionuclides and the number of reactions is evaluated from the activity measured by radiometric instruments. While TMDs for thermal neutrons can be easily prepared and evaluated due to their high thermal neutron cross sections for isotopes such as ^{10}B, ^{115}In or ^{197}Au, the requirements for fast neutron TMDs are higher. Cross sections for nuclear reactions suitable for the TMD method with fast neutrons are in the range of 1 to 100 mb for the fission spectrum. This leads to concentrations from 0.01 ppm to 1 ppm for fast neutron fluence of 1E19 cm^{-2}. These low concentrations imply requirements for detector material purity and sensitivity of concentration evaluation equipment. Materials for fast neutron TMDs were proposed, where the relevant parameters, such as energy threshold and minimal detectable fluence are known and convenient. An experimental comparison of some TMD and NAD responses was made in the LVR-15 research reactor using secondary ion mass spectrometry to evaluate the concentration of TMDs.

1 Introduction

Nuclear power is one of the primary sources of electricity worldwide, providing significant amounts of energy while emitting relatively low levels of greenhouse gases. However, the safe operation of nuclear power plants is of utmost importance, as any accident significantly damages the environment and human life. Neutron fluence is an important factor that is considered in the assessment of the safety of nuclear power plants. The measurement of neutron fluence is a critical input value for assessing the structural integrity of the reactor vessel and ensuring that it remains within safe limits during the operation of the nuclear power plant. This is particularly important due to the degradation of the material

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caused by the fast neutrons and can potentially lead to fragile fracture if not monitored. Therefore, the accurate and periodic measurement of neutron fluence is essential to ensure the safety and lifetime estimation of nuclear power plants.

Nowadays, neutron fluence and fluence spectrum adjustments are monitored using neutron activation detectors (NADs). The transmutation detectors (TMDs) [1] are investigated to improve the precision of neutron fluence evaluation.

The study of the TMDs was executed at Research Centre Řež (CVR) and irradiated in the LVR-15 research reactor. The LVR-15 reactor is a light water tank-type research reactor placed in a stainless-steel vessel under a shielding cover. It has a forced cooling, IRT-4M fuel and an operational power level of 10 MWt. Reactor operations run in 3 to 4 weeks cycles, followed by a maintenance period of 10-14 days. Demineralised water is used as both the moderator and coolant. Either water or a beryllium block (depending on the operation configuration) is used as a reflector. [2]

The Be, Si_3N_4 on Si, SiO_2 on Si, CaF_2, and Si TMDs were irradiated in the reactor, and afterwards, the number of reactions per atom *R* during the irradiation was determined with a secondary ion mass spectrometer (SIMS). Along with the TMDs, a set of NADs was also irradiated. The set of NADs contained detectors that are widely used in reactor dosimetry and whose neutron cross sections are well known, specifically Fe, Ni, Co, and Ti foils. The neutron spectrum was adjusted by STAYSL code and the number of TMD reactions was estimated by numerical code FISPACT-II for comparison.

2 Materials

The TMD and NAD materials should be mechanically, chemically and radiation resistant. The TMDs should also produce nuclides, that are stable in the detector (they should remain in the detector after irradiation). Furthermore, the TMD material should be chosen mainly regarding the purity and should have well known cross sections for the fast neutrons (but nowadays there are a many uncertainties with the cross sections of TMD reactions). The NADs should also have well known cross section for the fast neutrons and the half-life of the produced nuclide should not be too short (to be able to measure it on the High-Purity Germanium (HPGe) detector).

Due to the above properties, Si, Ge and CaF_2 crystal, Be foil, Si_3N_4 on Si and SiO_2 on Si were suggested as transmutation detectors, and Fe, Ni, Co, and Ti foils as activation detectors.

3 Evaluation

The transmutation detectors for neutron fluence and spectrum measurements are different from the activation detectors, specifically in evaluating detector response after the irradiation. The TMDs use stable transmuted isotopes and measure their concentration, while activation detectors use radioactive isotopes and activity is measured.

In general, if the ratio of reaction-product nuclides to target nuclides (measured by SIMS in the case of TMDs or gamma-ray spectrometry in the case of NADs) and the spectrum-averaged cross section are known, neutron fluence can be determined by the following formula:

$$\Phi = R / \bar{\sigma}_R, \tag{1}$$

where *R* is the ratio number of transmuted nuclides to the number of target nuclides (reactions per nucleus) and $\bar{\sigma}_R$ is the spectrum-averaged cross section for the reaction of

interest in the TMD or NAD. The formula (1) is valid if the burn-up of the target nuclide is negligible, which is true if $R << 1$ and is valid for all investigated cases. But in real cases, the energy spectrum is unknown and is a subject of measurement. In the general case, relation (1) is rewritten to

$$R = \int_0^\infty \Phi(E) \cdot \sigma_R(E) \cdot dE, \tag{2}$$

where $\Phi(E)$ is the differential neutron fluence and $\sigma_R(E)$ cross section for the energy E. To calculate R from (2), it is necessary to evaluate $\Phi(E)$. Then, results obtained from NAD and TMD can be compared or combined.

3.1 Activation detectors

The neutron activation detectors are devices used to measure neutron fluence in a particular place (reactor vessel, reactor core, etc.). This detection method is based on the bombardment of a material with neutrons (from a nuclear reactor, neutron generator, etc.) to induce a nuclear reaction that creates radioactive isotopes. The resulting radioactive isotopes emit characteristic radiation, precisely gamma rays or beta particles, which can be detected and measured using various techniques.

For each reaction, the ratio number of the activated nuclides to the number of the target nuclide R is calculated. The determination of the input quantities and the calculation of the R is in accordance with ASTM standards [3], [4], [5], and [6]. For the calculation of the ratio R, the history of irradiation and the activity per nucleus (A_{PN}) is used. The formula for the calculation of the ratio number R is as follows (for the constant fluence rate):

$$R = A_{PN} \cdot \lambda^{-1} / (1 - e^{-\lambda T_{irr}}), \tag{3}$$

where A_{PN} is the activity of the measured nuclide per nucleus and $1-e^{-\lambda T_{irr}}$ represents radionuclide decay correction for the course of irradiation, λ is the decay constant and T_{irr} is irradiation time.

3.2 Transmutation detectors

As said at the beginning of this chapter, the TMDs use stable transmuted isotopes and measure their concentration via various analytical methods. With SIMS, the R can be evaluated directly from the definition.

The number of stable nuclides generated by transmutation in the detector is measured using analytical methods, such as mass spectrometry (e.g., SIMS [7] or Resonance Ionization Mass Spectrometry (RIMS) [8]), neutron activation analysis (e.g., Prompt Gamma Neutron Activation Analysis (PGNAA) [9]) or nuclear magnetic resonance (NMR). TMDs have been investigated to improve the precision of neutron fluence evaluation.

The main advantage of TMDs is their independence from the irradiation history, TMDs retain information about neutron fluence for an unlimited amount of time and TMDs usually display insignificant appendant activity the end of irradiation. On the other hand, the high-purity material and the sensitivity of the analytical methods are needed. Additionally, NADs are more sensitive to fluence rate closer to the end of the irradiation period (TMDs are not sensitive to the end of the irradiation). Therefore, NADs can give a response that is weighted more towards the later irradiation history if the fluence rate is not constant. The benefit of the TMDs development should be an increase in the accuracy of

long-term neutron fluence measurements and consequently, for example, to extend the operation of nuclear power plants while maintaining operational safety.

3.3 Secondary ion mass spectrometry

The Secondary Ion Mass Spectrometry (SIMS) is a method based on the emission of ionized atomic species from surfaces of solid samples bombarded with energetic ions with kinetic energies in the keV range. Most of the emitted (sputtered) atomic species are in the electrically neutral form. Positive or negative secondary ions comprise only a small fraction of the total (~1% depending on the species). These (secondary) ions are then accelerated in an electrostatic field to keV-energies, extracted into the spectrometer, mass analysed by mass to charge ratio and finally detected with an electron multiplier.

The beam of primary ions is focused to a small spot, typically between 1 μm and 100 μm, and raster scanned on the surface over a square area. A micro crater is formed when measuring a typical size of 100 μm x 100 μm and the depth from a few tens of nanometres to several micrometres depending on the analytical task. The measurement must be done in the ultra-high vacuum, which imposes the vacuum compatibility requirements on the samples.

There are several operating modes of SIMS. Firstly, it is possible to acquire a mass spectrum, which provides information about the presence of elements and isotopes in the sample and under certain conditions also molecules on the surface. A layer-by-layer sputtering within the raster-scanned area enables to measure the secondary ion signal as a function of depth, thus providing a concentration depth profile, which is the most natural operating mode of SIMS. Raster scanning of the primary ion beam allows for obtaining the elemental 2-dimensional distribution on the sample surface – the SIMS imaging mode. Combining the latter two modes allows for a 3-dimensional spatial elemental distribution. Finally, the SIMS method enables the measurement of isotopic ratios of chosen isotopes from small areas on the sample with high accuracy and precision.

The SIMS method has a prominent position among chemical analytical techniques based on the combination of extreme sensitivity (ppm to sub-ppb), good imaging resolution, the ability to analyse all elements and isotopes of the periodic table, and to obtain the elemental and isotopic information from small sample volumes.

However, a quantitative analysis can be done only if calibration standards, i.e., the samples of the same composition of major (matrix) elements as in the analysed sample, are available.

4 Experimental Work

~~A~~n experiment investigating transmutation detectors for fast neutrons was performed. The set of Si and CaF_2 crystal, Be foil, Si_3N_4 on Si and SiO_2 on Si, and a set of the activation detectors (Fe, Ni, Co, and Ti foils) was irradiated in the research reactor LVR-15. The ratios *R* were determined by SIMS for transmutation detectors. The activity of each NAD was measured by HPGe detectors. The results were used to evaluate received neutron fluence in TMD during irradiation.

4.1 Irradiation in the LVR-15 research reactor

The TMDs and NADs were irradiated in the vertical channel of the LVR-15 reactor, specifically in channel H5 in the second irradiation position. The maximum power of the reactor was 10 MW, and the mean power during irradiation was 8 MW.

Before irradiation, the TMDs and NADs were wrapped in an aluminium foil and placed in a cylindrical aluminium irradiation capsule with a diameter of 3.6 cm and a length of 12 cm.

The detectors in the capsule were irradiated in the vertical channel for 21.6 days at fluences of 9.9E19 cm^{-2} for thermal and 2.1E19 cm^{-2} for fast (>1 MeV) neutrons. After irradiation, the detectors were measured, and the number of produced isotopes of the investigated reactions was evaluated.

4.2 Detector measurements and calculations

The measurements of TMDs were performed using a double-focusing magnetic sector SIMS IMS 7f (Cameca). All the SIMS measurements were performed in a bulk analysis SIMS mode, a depth profiling mode at high sputter rate. In the case of the Si crystal, the pre-sputtering was applied before the SIMS data acquisition to reduce the level of contamination from the Al foil wrap (the Al foil wrap was in contact with the surface of the Si crystal during irradiation). The sputterring time of the SIMS measurement was so long that all signals levelled off to be free from the surface effects and that the levelled part of the profile provided sufficient counting statistics.

Moreover, not only the irradiated TMD was measured, but also the blank TMD, which is identical, but non-irradiated TMD made from the same material as the irradiated one. It was measured under the same SIMS setup. Measurements of both irradiated and non-irradiated were done for each TMD material.

The activity of the NADs was measured using a gamma spectroscopy instrument with a HPGe detector (Canberra), calibrated, and evaluated according to ASTM E181-17 [10]. Using the formula (3), the number of produced nuclides R was calculated and utilized in the STAYSL PNNL software for $\Phi(E)$ estimation. The input also included the neutron flux spectrum during the irradiation and was obtained from the MCNP calculations. Lastly, the input involved neutron activation cross sections obtained from IRDFF nuclear data library.

For the ratio of the produced nuclides R evaluation, the FISPACT-II software using formula from (2) was used. The integral in (2) was approximated by the sum of values corresponding to 709 energy groups, $\Phi(E)$ was obtained from STAYSL PNNL using NAD measurements and $\sigma_R(E)$ from various nuclear data libraries.

5 Results and discussion

The ratio R of the NAD was gained from formula (3) and are in Table 1 together with E_{05} lower energy range (5% of detector response occurs below this energy) [3]. As stated above, the R served as an input in programme called STAYSL PNNL. From this computational programme, the neutron spectra $\Phi(E)$ was received and utilized in the computational software FISPACT-II.

From FISPACT-II and neutron spectrum gained with NAD using STAYSL, the R_C using formula (2) was obtained and are summarized in Table 2. Additionally, the TENDL-14 nuclear data library was applied.

Table 1. Ratio R and E_{05} of the NADs.

Detector	Reaction	E_{05} (MeV)	R (ppm)
Fe	$^{54}Fe(n,p)^{54}Mn$	2.32	2.37E+00
Ni	$^{58}Ni(n,p)^{58}Co$	2.05	2.69E+00
Co	$^{59}Co(n,\gamma)^{60}Co$	0	4.49E+03

Ti	^{46}Ti(n,p)^{46}Sc	3.76	3.53E-01

Table 2. Ratio R and E_{05} of the TMDs (using TENDL-14).

Detector	Reaction	E_{05} (MeV)	R_C (ppm)
Be	^{9}Be(n,α)^{6}He (β-) ^{6}Li	1.0	9.40E-01
Si_3N_4 on Si	^{14}N(n,α)^{11}B	0.7	2.26E+00
SiO_2 on Si	^{16}O(n,α)^{13}C	3.2	1.93E-01
CaF_2	^{19}F(n,α)^{16}O	3.0	3.34E-01
Si	^{28}Si(n,α)^{25}Mg	2.8	1.10E-01
	^{29}Si(n,α)^{26}Mg	2.4	1.28E-01

Furthermore, the R_E (represents experimental value) for the Si crystal via SIMS was measured, and the results were taken from [7]. The ratio R_E for the Si crystal measured with SIMS was compared with the ratio R_C (represents calculated value) utilizing various nuclear data libraries. The comparison of these ratios is shown in Table 3.

Table 3. Comparison of R_E and R_C gained by SIMS and FISPACT-II.

Detector	R_E (ppm)	Nuclear Data Library	R_C (ppm)	R_E/R_C
^{28}Si(n,α)^{25}Mg	8.46E-02	TENDL-17	9.56E-02	0.88
		TENDL-14	1.10E-01	0.77
		JEFF-3.2	7.08E-02	1.19
		JENDL-4	7.80E-02	1.08
		ENDF-VIII	8.70E-02	0.97
^{29}Si(n,α)^{26}Mg	2.07E-01	TENDL-17	1.37E-01	1.51
		TENDL-14	1.28E-01	1.62
		JEFF-3.2	1.52E-01	1.36
		JENDL-4	1.50E-01	1.38
		ENDF-VIII	1.52E-01	1.36

Ideally, the ratio of R_E and R_C should be 1 – the ratio of the produced nuclides should be identical either using measurement of TMD (e.g., with SIMS) or NAD with numerical software (e, g, FISPACT-II). Different ratio values seen in Table 3 mean, that it depends, which nuclear data library is used. The uncertainty of gained R_C consists of the uncertainty of fluence $\Phi(E)$ and the uncertainty of $\sigma_R(E)$. The uncertainties for the activation detectors are estimated to 5% for the number of the ratio R and up to 15% for the neutron spectra obtained from STAYSL PNNL. Moreover, the uncertainty of R_E is estimated to 5% and the uncertainty of the cross sections of the reactions utilizing Si crystal between 15%-60%. Larger uncertainties could be obtained using cross sections values from several nuclear data libraries. For example, the JEFF-3.2 Nuclear Data Library provides the R_C 54.5% higher than R_C gained using TENDL-14 for ^{28}Si(n,α)^{25}Mg reaction.

The ratio closest to number 1 is given by the library ENDF-VIII, as seen in Table 3, specifically for the reaction ^{28}Si(n,α)^{25}Mg. The ratios R_E/R_C of the reaction ^{29}Si(n,α)^{26}Mg range from approximately 1.36 to roughly 1.62. This could be due to the contamination

from the Al foil wrap (the Al foil wrap was in contact with the surface of the Si crystal during the irradiation) or the inhomogeneous distribution of the ^{26}Mg product. Therefore, a further SIMS measurement of this reaction is proposed. The other reactions have not yet been measured by SIMS and are also the subject of future evaluation.

6 Conclusion

In this article, materials for transmutation detectors sensitive to fast neutrons were proposed. The TMDs were irradiated and the ratio of the transmuted nuclides to the number of the target nuclides R_C using FISPACT-II was calculated. Additionally, the ratio R_E of two reactions, specifically ^{28}Si(n,α)^{25}Mg and ^{29}Si(n,α)^{26}Mg was taken from [7].

Table 3 shows that there is a strong dependence on the used nuclear data library and refinement of the cross-sections is needed. Also, the most promising transmutation detector for fast neutrons appears to be TMD using ^{28}Si(n,α)^{25}Mg reaction. The rest of the detectors will be measured using SIMS in the future.

Overall, the TMD method is relatively new, so further development can be expected in the future, including the specification of tabulated values of the cross-sections, the development of pure materials or further development of analytical methods.

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