

DETERMINATION OF ^{54}Mn AND ^{59}Fe SPECIFIC
ACTIVITIES IN IRON PLATES ACTIVATION DETECTORS

Lyuben Dobrev, Mladen Mitev, Dobromir Dimitrov,
Tzvetana Nonova, Zahari Zahariev, Desislava Dimitrova,
Srgjan Krstevski, Bozhidar Slavchev

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Abstract

In reactor dosimetry it is well known that the empirical determination of the neutron flux is performed by determining the isotope ^{54}Mn generated in a (n,γ) nuclear reaction. This study presents a procedure for the simultaneous determination of the radioisotopes ^{54}Mn and ^{59}Fe , developed to obtain more complete information about the process. The basis of the method is the irradiation of iron-plate detectors with a neutron flux generated in a VVER-type nuclear reactor and subsequent gamma spectrometric analysis of the samples to determine the two radionuclides obtained in two simultaneous (n,x) and (n,γ) nuclear reactions. A very important stage of the assay is the preliminary radiochemical treatment of the samples involving the removal of oxides and corrosion products and the dissolution of the plates in strong mineral acids.

The results show that our approach of simultaneous determination of ^{54}Mn and ^{59}Fe significantly improves the time of analyses, reduces the radiation dose, and increases the accuracy and efficiency of neutron fluence determination. The procedures are relatively easy to perform and are suitable for application in routine studies.

Key words: neutron fluence, iron-59, manganese-54, reactor dosimetry, gamma-spectrometry

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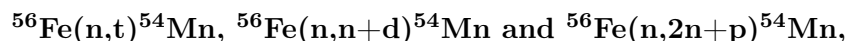
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Introduction. During the nuclear reactor operation, neutron radiation from the reactor core causes embrittlement of the reactor vessel. Embrittlement refers to a decrease in the fracture toughness of reactor pressure vessel materials and affects the vessel materials in the vicinity of the reactor fuel, referred to as the vessel's "beltline".

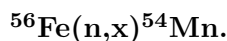
The neutron fluence is an important parameter for predicting the embrittlement of the metal. Its determination requires a study of the content of ^{54}Mn obtained by irradiating iron plate detectors with a stream of neutrons generated in the nuclear reactor during the fuel campaign.

This approach is based on one of the main experimental methods for studying neutron fluence used in reactor dosimetry and can be applied to validate the calculated neutron fluence of VVER-1000 reactors at Kozloduy NPP [1].

The production of the ^{54}Mn during the process of neutrons interaction with ^{56}Fe is implemented by three different nuclear reactions:



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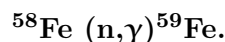


The energy thresholds (Eth) of the reactions are as follows:

- Eth = 12.14309 MeV for the (n,t) reaction,
- Eth = 18.51319 MeV for the (n,n+d) reaction and
- Eth = 20.77788 MeV for (n,2n+p) reaction.

The cross sections for the reactions are included into IRDFF-2 library as described by ZOLOTAREV [2] and TRKOV et al. [3].

Because of the presence of ^{58}Fe in the natural iron isotope mixture, a parallel nuclear reaction of generating ^{59}Fe is running simultaneously:



^{58}Fe is also activated by neutrons with energies close to those in the main reaction.

As is well known, natural iron is a mixture of four stable isotopes: ^{54}Fe – 5.85%, ^{56}Fe – 91.75%, ^{57}Fe – 2.12%, and ^{58}Fe – 0.282%. The iron plates used in our study contain the stable isotopes of iron in the same proportions as in the natural mixture.

This article presents the results obtained in determining the neutron fluence by gamma-spectrometric analysis of two sets of 26 samples, in which the content of ^{54}Mn and ^{59}Fe was determined after preliminary isolation from a complex mixture using radiochemical methods. This approach is related to the application

of a destructive method to separate the two studied isotopes from the main matrix, preparation of laboratory samples for measurement and subsequent gamma spectrometric analysis to determine the specific activity of the isotopes. The two sets of samples were selected and irradiated in consecutive standard 11 month fuel campaigns of the same high-power nuclear reactor. Set 1 was irradiated for two fuel campaigns and Set 2 for one fuel campaign. The samples were positioned in the same places, fixed, and irradiated.

Experimental. Two different sets of samples were irradiated for two and one fuel campaign, respectively, behind a welding steam located just in front of the core of a 1000 MW VVER nuclear reactor. For a better assessment of the disturbances resulting from corrosion and the accompanying reactions from metal impurities, two types of iron plates were used – pure for analysis iron (99.99 %) and technical iron with an iron content of 98.5% and about 1.5% impurities. The differences in the analytical spectra obtained from the measurement of the plate of pure iron and of technical iron arise from the presence of more than 1% impurities in the second one (Fig. 1). There are no peaks of impurities in the spectrum of the pure iron plates.

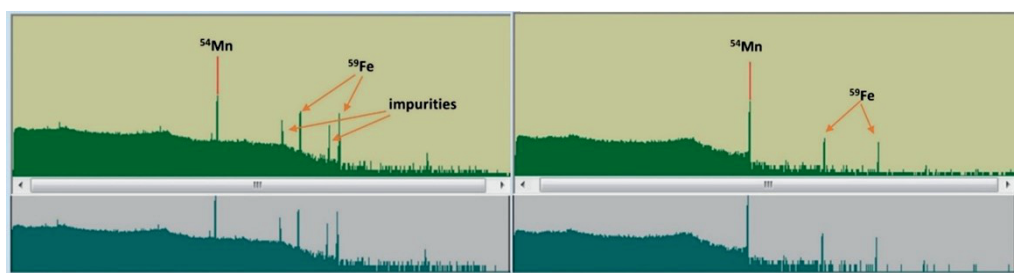


Fig. 1. Left – Spectrum of iron plate sample. Right – Spectrum of pure iron plate sample. Both measured by Ortec gamma-spectrometry system

The activation of the impurities leads to the generation of some radioactive isotopes such as ^{60}Co , which contributes to the overall gamma spectrum, but due to its discrete nature does not significantly affect the obtained results.

The plates were packed and placed in a 10 cm long aluminium strip, positioned inside an aluminium pipe, and placed on a frame stand perfectly aligned with the welding steam. Each individual set of iron plates was fixed in the same place. After completion of the specified exposure time, the tape with the iron plates was removed, the plates were dismantled and stored in separate containers for subsequent analysis.

The iron plates were pretreated to remove the corrosion products and then a radioanalytical procedure was applied to dissolve the samples and isolate the radioisotopes studied. The elimination of corrosion products is an important stage of the pretreatment and ensures accurate and reproducible results. The formed black coating of iron oxides on the surface of the plates is removed using diluted

HCl and an ultrasonic bath. The initial mass of each plate was determined and subsequently used to calculate the specific activity of the samples. A mass of 50 mg was measured for the pure iron plates and 200 mg for the technical iron plates.

Every plate was transferred in a separate glass beaker and 10 ml concentrated HCl, 5 ml concentrated HNO₃, and 1 ml 30% H₂O₂ were added. The beakers were placed on a heating plate and heated to 150 °C. After 10 min to each solution we added 10 mg of stable manganese as a solution of MnCl₂ for enriching the sample with manganese. This is necessary because of the low mass quantity of the generated ⁵⁴Mn, and for ensuring homogeneity of the sample. The samples were heated for about 90 min until the plates were completely dissolved. After cooling to room temperature, the solutions were quantitatively transferred in 250 ml polypropylene standardized containers, filled up to 250 ml with double distilled water and measured by gamma spectrometry. The main steps of the radiochemical sample preparation procedure are presented schematically in Fig. 2.

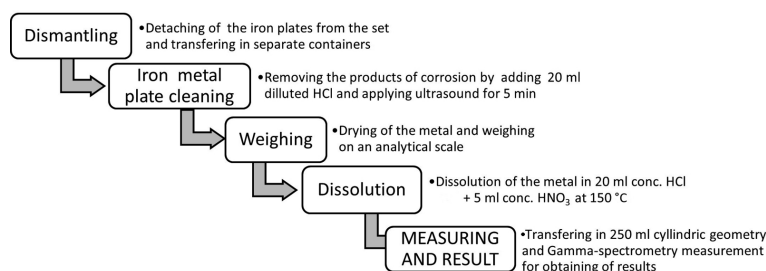


Fig. 2. Scheme of the analytical procedure for preparation of samples for gamma-spectrometry measurement

Gamma spectrometric measurements were performed with a low background HPGe (GEM) detector with 13% relative efficiency and a resolution of 1.6 keV for 1.33 MeV (Co-60) as in [4].

The efficiency calibration of the detector was done using a prepared secondary standard calibration source with a density of 1 g.cm⁻³ and geometry cylinder with volume of 250 cm³.

Results and discussion. The choice of a destructive method for the preparation of the samples for measurement complicates the procedure to a certain extent, but it makes it possible to completely isolate the two isotopes under investigation, which in turn guarantees high accuracy of the data obtained about their content in the matrix of the plates. The applied approach also enables an easier and more accurate calibration of the efficiency, due to avoiding the interference of the imprecise geometry of the plates.

The studied experimental data for the content of ⁵⁴Mn and ⁵⁹Fe in the two studied sets were normalized and a comparison was made with the theoretically calculated data. For this purpose, the actual previously provided detailed data describing the relevant fuel campaign and the validated TORT computer code [5,6]

were used. To obtain reliable results, the latest cross-sectional data defining the appropriate activation response are included in the calculations. The calculation results were obtained using the cross-section database IRDFF [3,7,8].

Figure 3 shows the normalized results for ^{54}Mn and ^{59}Fe from two sets of samples, irradiated for two and one consecutive fuel campaigns, respectively. The normalization was performed by scaling the data to the maximum value (maximum normalization). The normalization formula for the activity is as follows:

$$A_{\text{norm}} = A_i / A_{\text{max}},$$

where A_i is the i -th activity from the data set and A_{max} is the maximum activity from the same data set.

In Figure 3 the numbers of the analyzed samples from 1 to 26 for every set are pointed on the abscissa.

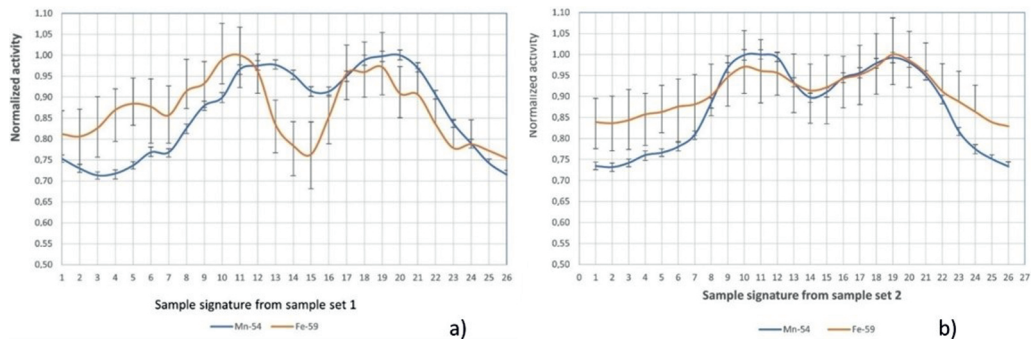


Fig. 3. a) Normalized results for ^{54}Mn and ^{59}Fe , obtained from analysis of sample set 1, irradiated for two fuel campaigns. (b) Normalized results for ^{54}Mn and ^{59}Fe , obtained from analysis of sample set 2, irradiated for one fuel campaign

The results for ^{59}Fe show that the general trend of the curve of data is respected (Fig. 3). The differences in the neutron fluence and in the time of irradiation are not determinative for the significant difference in the data sets for ^{54}Mn and ^{59}Fe . The discrepancy came from the minimal quantity of the ^{58}Fe (0.3%) in the iron plate matrix, against the great amount of ^{56}Fe (91.7%).

To assess the possibility of using plates made of the significantly cheaper technical iron, a comparison of the data obtained for the two types of investigated plates was made. Figure 4 shows the comparison of the normalized results obtained from pure iron plates and technical iron plates. The results show a difference between the two data sets within 3%. This difference is negligibly small and shows that plates of technical iron can be successfully used in experiments to determine the neutron fluence, and from the data obtained from their analysis, an adequate prediction can be made about the brittleness of the material of the nuclear reactor vessel.

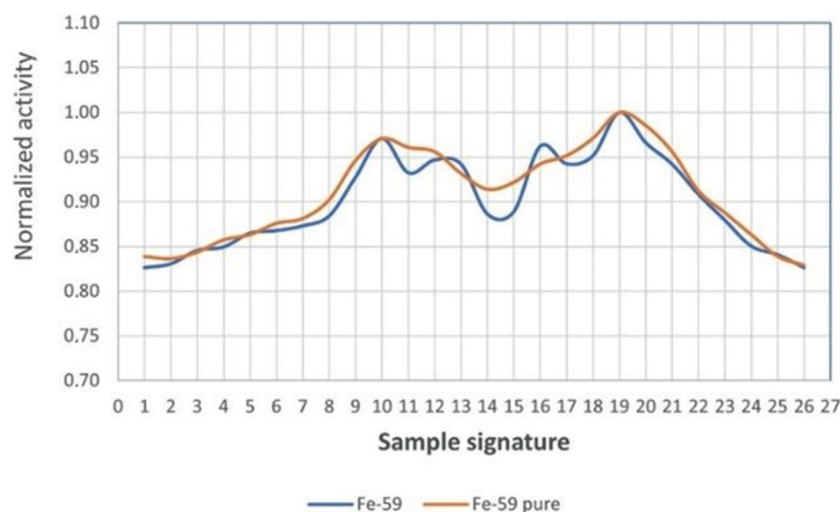


Fig. 4. Normalized results for ^{59}Fe in technical and pure irradiated iron plates, obtained from analysis of the samples in the same set

Conclusions. The obtained by us data for neutron fluence by the reaction $^{58}\text{Fe}(n,\gamma)^{59}\text{Fe}$ are comparable to the data obtained by the classical method of determining the neutron fluence by the reaction $^{56}\text{Fe}(n,x)^{54}\text{Mn}$ (see Fig. 3), which is a proof of the effectiveness of our applied approach of using destructive radiochemical analysis in the preparation for measuring the iron plates. The trends found for the $^{58}\text{Fe}(n,\gamma)^{59}\text{Fe}$ reaction are similar but the low ^{58}Fe content of the iron plates used leads to a lack of sufficient material and degrades the accuracy of the analyses. The different irradiation time used – one fuel campaign for the first set of samples and two fuel campaigns for the second set – shows that the time has no significant effect on the reliability of the results obtained for the ^{54}Mn content, but it is important for ^{59}Fe . However, they depend on time and show different trends. Longer exposure period resulted in better model fit, followed by ^{54}Mn . This indicates that to obtain more reliable results, it is necessary to use iron material with a higher ^{58}Fe content. However, the second set of data obtained for the parallel reaction generating ^{59}Fe cannot be used as an independent and reliable method for determining the neutron flux. Therefore, the neutron flux as well as the brittleness of the nuclear reactor vessel material can be estimated more accurately by examining the ^{54}Mn content and radiochemical preparation of the samples is a suitable approach.

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Data availability statement. Data supporting this study are normalized and not publicly available in their full range because they represent sensitive information about the safety operation of the nuclear power plant.

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Institute for Nuclear Research and Nuclear Energy
Bulgarian Academy of Sciences
72 Tsarigradsko Shosse Blvd
1784 Sofia, Bulgaria
e-mails: ltdobrev@gmail.com
mlmitev@inrne.bas.bg
nonova@inrne.bas.bg
zzahariev@gmail.com
dessy-d@inrne.bas.bg
reskim_2004@yahoo.com
bobislavchev@inrne.bas.bg