Available online at www.sciencedirect.com

SCIENCE DIRECT®

Journal of Nuclear Materials 345 (2005) 86–88

journal of nuclear materials

www.elsevier.com/locate/jnucmat

Letter to the Editors

Burnup effect on ^{95}Nb/^{95}Zr ratio-cooling time correlation

Kh. Haddad *

Nuclear Engineering Department, Atomic Energy Commission, P.O. Box 6091, Damascus, Syria

Received 11 March 2005; accepted 30 May 2005

Abstract

The axial ^{95}Nb/^{95}Zr ratio distribution in a fuel assembly of the typical research reactor (IRT) was determined experimentally by gamma scanning. The results showed that this ratio is stable along the fuel assembly axis regardless of the position of the scanned section. This allows to limit gamma scanning of the whole assembly on the measurement of the central section only. This will save time, efforts and experimentalist's exposure to radiation. In addition, the effect of burnup on the ^{95}Nb/^{95}Zr-cooling time correlation was investigated. The results showed that, using this correlation to determine cooling time, will include a systematic error of about 12%.

1. Introduction

The ^{95}Nb/^{95}Zr ratio is used to determine the cooling time of irradiated nuclear fuels for safeguard purposes when the cooling time is not higher than one year. Gamma scanning of irradiated fuel is one of the most widespread and preferred nondestructive methods for irradiated fuel research [1,2]. In this work we have analyzed the results of gamma scanning, which we have carried out on three fuel assemblies of different burnups in the IRT reactor of the Moscow State Engineering Physics Institute (MEPHI). The IRT belongs to medium power research nuclear reactors and is of the heterogeneous thermal water reactor pool type. Moderator and partially reflector serve ordinary distilled water, which is simultaneously used, as a coolant [3]. We have studied the ^{95}Nb/^{95}Zr ratio stability along the fuel assembly axis and the effect of burnup on the ^{95}Nb/^{95}Zr ratio-cooling time correlation. The fuel assembly contains eight coaxial tubular fuel elements. Each fuel element is composed of a three-layer pipe (core and coverings). The material of the fuel element cores is uranium dioxide dispersed in an aluminum matrix. The initial fuel assembly enrichment is 90% and its active layer length is (580 ± 20) mm.

2. Stability of the ^{95}Nb/^{95}Zr ratio along the axis of the fuel assembly

The gamma spectrum of each scanned section of the irradiated fuel assembly was acquired and peak areas of ^{95}Zr (756.7 keV) and ^{95}Nb (765.8 keV) were measured (Fig. 1). The main components of the experimental device are the hot cell inside which the scanning system, collimator system and spectrometer system were constructed. The spectrometer system includes a coaxial HPGe detector with resolution of 2.41 keV for the gamma line of ^{60}Co (1173.2 keV) and an efficiency of 10%. The lead protection surrounding the detector has kept the ratio background/peak less than 1%.

The ^{95}Nb/^{95}Zr ratio was calculated using the relation [4]

---

* Tel.: +963 11 2132581; fax: +963 11 6112289.
E-mail address: scientific@aec.org.sy

0022-3115/$ - see front matter © 2005 Elsevier B.V. All rights reserved.
doi:10.1016/j.jnucmat.2005.05.014

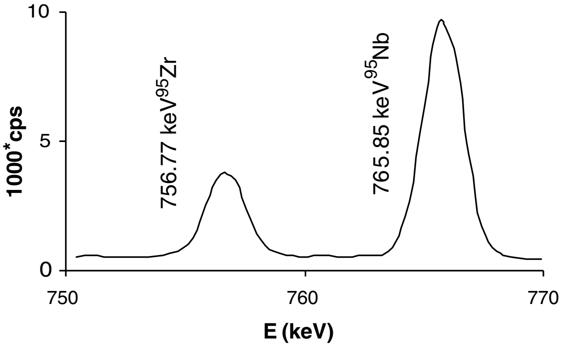

Fig. 1. Gamma spectrum of fuel assembly.

$$\frac{N_{\mathrm{Nb}}}{N_{\mathrm{Zr}}}=\frac{\lambda_{\mathrm{Zr}}}{\lambda_{\mathrm{Nb}}}\cdot\frac{\varepsilon(E_{\mathrm{Zr}})}{\varepsilon(E_{\mathrm{Nb}})}\cdot\frac{B_{\mathrm{Zr}}}{B_{\mathrm{Nb}}}\cdot\frac{P_{\mathrm{Nb}}}{P_{\mathrm{Zr}}},\tag{1}$$

where N_i – number of atoms of the i-th fission product at cooling time t_c; λ_i – decay constant; P_i – the net peak area under the energy peak of the i-th fission product; B_j – branching ratio; $\varepsilon(E_i)$ – efficiency of the measuring setup.

Noting that all parameters other than P_{Nb}, P_{Zr} in relation (1) are constants, we can use the value of $P_{\mathrm{Nb}}/P_{\mathrm{Zr}}$ as an indicator of the ratio $N_{\mathrm{Nb}}/N_{\mathrm{Zr}}$.

The rate of the fission product generation in a fuel assembly depends mainly on the fission rate. The fission rate depends above all on the neutron flux and the fissile material concentration, i.e. burnup. It is well known that neither the neutron flux nor the burnup has constant values along the axis of the fuel assembly. The studies showed that the neutron flux is maximum at the centre of the fuel assembly [1–3]. Thus the axial distribution of the specific fission product will certainly depend on the axial distributions of neutron flux and burnup. The effects of these distributions on the stability of the ^{95}Nb/^{95}Zr ratio along the axis of the fuel assembly were investigated in this work. Axial gamma scanning was carried out for fuel assemblies 133, 185 and 183. The values of P_{Nb} and P_{Zr} were measured for each scanned section (Table 1).

The analysis of Table 1 indicates the following results:

(a) the (P_{j}) values pertain their maxima in the central sections ($z = 0$) of the fuel element;
(b) neither P_{Nb} (or N_{Nb}) nor P_{Zr} (or N_{Zr}) has constant values along the axis of the fuel assembly;
(c) the standard deviation of the $P_{\mathrm{Nb}}/P_{\mathrm{Zr}}$ axial values along the fuel assembly does not exceed 1.33%, which is less than the experimental error. According to relation (1) we can say also that
(d) the standard deviation of the $N_{\mathrm{Nb}}/N_{\mathrm{Zr}}$ axial values along the fuel assembly does not exceed 1.33%.

These results allow us to reduce the scanning of the whole fuel assembly to the measurement of the central section only. This reduction will save time, efforts and the experimentalist's exposure to radiation.

3. The effects of burnup on the ^{95}Nb/^{95}Zr ratio-cooling time correlation

The theoretical ^{95}Nb/^{95}Zr-cooling time correlation is given by the expression [2]

$$t_c=\frac{1}{\lambda_{\mathrm{Zr}}-\lambda_{\mathrm{Nb}}}\mathrm{Ln}(f/f_0),\tag{2}$$

where $f = N_{\mathrm{Nb}}/N_{\mathrm{Zr}} + \lambda_{\mathrm{Zr}}/(\lambda_{\mathrm{Zr}} - \lambda_{\mathrm{Nb}})$, $f_0 = N_{\mathrm{Nb0}}/N_{\mathrm{Zr0}} + \lambda_{\mathrm{Zr}}/(\lambda_{\mathrm{Zr}} - \lambda_{\mathrm{Nb}})$, N_{Nb0} and N_{Zr0} are the numbers of ^{95}Nb and ^{95}Zr atoms at the end of the last irradiation ($t_c = 0$). To study the effect of burnup on the ^{95}Nb/^{95}Zr

Table 1
Axial distribution of ^{95}Nb and ^{95}Zr and $P_{\mathrm{Nb}}/P_{\mathrm{Zr}}$ ratio

Fuel assembly no.	Burnup (%)	z (mm)		Fission product					
				^{95}Zr		^{95}Nb			
			E (keV)	757		766		Ratio	
			t_c (d)	P_{Zr} (cps)	e[a] (%)	P_{Nb} (cps)	e (%)	$P_{\mathrm{Nb}}/P_{\mathrm{Zr}}$	e (%)
133	46.50	−220	71.58	10.19	3.3	28.17	4.6	2.76	5.7
		0	71.59	10.94	3.8	27.91	4.1	2.83	5.6
		220	71.60	5.40	3.9	15.28	3.0	2.83	4.9
185	10.09	−220	75.56	11.11	4.3	31.55	4.6	2.84	6.3
		0	75.54	11.52	5.5	33.01	5.9	2.87	8.1
		220	75.57	5.21	2.1	14.72	2.2	2.82	3.0
183	47.69	−220	78.52	6.13	0.5	17.97	1.4	2.93	1.5
		0	78.53	6.84	3.3	19.78	2.9	2.89	4.4
		220	78.54	4.01	2.0	11.77	1.9	2.93	2.8

[a] – Relative standard deviation (%).

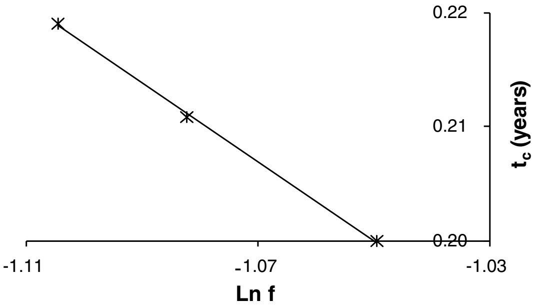

Fig. 2. The variation of Lnf versus cooling time.

ratio-cooling time correlation, the value of N_{Nb}/N_{Zr} was determined experimentally using relation (1) and consequently the value of f. The curve Lnf versus t_c was plotted in Fig. 2.

Fitting this experimental results corresponding to the three eight-tube-fuel assemblies 133, 183 and 185, which had different burnups using the least-squares method, we got the linear relation

$$t_c = -0.346 \, \mathrm{Ln} f - 0.167. \tag{3}$$

The coefficient of determination (R^2) for the resulted line is equal to 0.999. This linearity agrees very well with the theoretical relation. So the different burnups of the fuel assemblies have not disturbed the linear ^{95}Nb/^{95}Zr ratio-cooling time correlation. This result indicates that different burnups do not cause any arbitrary error.

To estimate the effects of burnup on the ^{95}Nb/^{95}Zr ratio-cooling time correlation quantitatively, the ratio of the experimental slope (relation (3)) to the theoretical one (relation (2)) was determined. This ratio was 1.12. This means that the difference is about 12%. The result indicates that different burnups cause a systematic error of about 12%.

These results are interpreted as follows. Full energy gamma peaks of ^{95}Nb and ^{95}Zr rest on a background continuum caused by the Compton scattering of higher gamma rays. This background masks parts of the ^{95}Nb and ^{95}Zr peaks and consequently causes errors in the determination of the fission product quantity. The measured fuel assemblies were irradiated simultaneously in the reactor core during the last operation period [3]. However, before the last irradiation which lasted 4 months, these assemblies were not irradiated longer than 14 months. So, the background is mainly based on the radiation of fission products and activation products formed during the last period only. Therefore the effect of this background is the same regardless of the burnup values. Thus, the resulting error is the same in fuel assemblies of different burnups. It means that different burnups will cause a systematic error and that which we have got in our work.

4. Conclusion

The stability of the ^{95}Nb/^{95}Zr ratio along the axis of the fuel assembly was investigated experimentally. The results showed that this ratio is stable along the fuel assembly axis regardless of the position of the scanned section. This allows to limit gamma scanning of the whole assembly on the measurement of the central section only. This will save time, efforts and experimentalist's exposure to radiation. In addition to that, the effect of burnup on ^{95}Nb/^{95}Zr-cooling time correlation was studied. The results showed that this correlation for the cooling time will include a systematic error of about 12% and does not cause any arbitrary error.

Acknowledgements

The author would like to thank Professor I. Othman (G.D. of AECS) for his encouragement and support. The author also expresses his gratitude to Professor A. Bushuev and the staff of the IRT reactor in MEPHI.

References

[1] H. Graber, G. Hofmann, S. Nagel, A. Keddar, Gamma-spectrometric determination of burnup and cooling time of irradiated ECH-1 fuel assemblies. in: Proceedings of the Symposium on Nuclear Material Safeguards, IAEA, Vienna, 2–6 October 1978, IAEA Vienna, 1979, vol. 1, p. 353.

[2] L. Hanna, Safeguards verification of spent materials testing reactor (MTR) fuel using gamma-ray spectrometry. in: Proceedings of the Symposium on Nuclear Material Safeguards, IAEA, Vienna, 2–6 October 1978, IAEA Vienna, 1979, vol. 1, p. 369.

[3] A. Bushuev, Kh. Haddad, V. Zubarev, A. Portnov, M. Shurovskaya, A. Kojin,V. Alferov, Experimental and calculation research of heat generation field and burnup of fuel IRT, Atomnaya-Energiya (Moscow) 2000, vol. 88, p. 4261.

[4] IAEA-TECDOC-633, Determination of research reactor fuel burnup, IAEA Vienna, 1992.