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PHOTOACTIVATION OF NUCLEAR ISOMERS FOR ASSAYING IRRADIATED REACTOR FUEL

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Abstract: Indium samples were activated by (γ,γ') method along the central longitudinal axis of spent fuel assemblies seven weeks after their discharge from the core of a pressurized water reactor type WWER-440 in Paks, Hungary. Measuring $^{115}In^{m}$ isomeric activities excited by gamma radiation of fission products (^{140}Ba-^{140}La) allowed scanning fuel assemblies axially by this method enabling reactor power distribution to be determined. A comparison was made between the results of the WWER-440 cassette mockup measurement and those of the fuel assembly measurements. The method -in addition to the computerized burnup calculations- can be useful for reactor operational and safeguards purposes.

Introduction

Several experimental methods and calculations exist for burnup and fissile content determination requiring different technical background and offering various degree of accuracy. Dragnev[1] and Hsue[2] have given a general review of non-destructive methods, among those gamma-spectroscopy is one of the most widely used. Here we report a novel method related closely to the gamma-spectroscopic one, it has several favourable features. It is based on the fact, that hard gamma radiation from fission products activates several nuclides producing nuclear isomers via inelastic (γ,γ') resonance scattering[3]. We measured research reactor fuel assemblies and preliminary results show suitability of the method for power plant assemblies too[4, 5, 6].

Isomer Excitation by Hard Gamma Radiation of Fission Products

Isomer excitation occurs when the energy of exciting gamma quanta corresponds to that of one or more activation levels, from where target nucleus comes to the isomeric state via various cascade transitions. The resonance energy gamma quanta are produced by Compton scattering mainly in the source (i.e. spent fuel) itself, and in the surrounding material (e.g. other fuel elements).

Among other candidates such as ^{77}Se, ^{111}Cd, ^{167}Er and ^{197}Au, ^{115}In proved to be the most suitable target isotope having a metastable state with reasonable half-life (4.5h) It was selected by model experiments carried out with 20 TBq (540 Ci) ^{140}La sources[4]. ^{140}La, which is the daughter of ^{140}Ba, represents well the high energy gamma spectrum of the spent fuel. In the case of ^{115}In the energy of the first activation level is 1.078 MeV. Gamma quanta of energy below this threshold are ineffective in isomer activation. Only few fission products have intense gamma-rays higher than 1 MeV together with a relatively long half-life and considerable cumulative fission yield (see Table I.). Contribution of superthreshold energy gammas other than ^{140}Ba-^{140}La is less than 10% in the cooling time interval of 10-60 days[5]. Detection of 336 keV (48%) gamma transition of $^{115}In^{m}$ provides a relative method for monitoring ^{140}Ba content of the fuel.

Gamma Activation Experiments at NPP Paks

The experiments were started in the spent fuel store where the hexagonal fuel assemblies were stacked vertically in the storage rack in the pond water. In order to avoid the influence of the surrounding assemblies, the actual measurements were carried out in the adjacent supervision mine filled also with borated water (boric acid concentration of 12 g/l), where the individual assemblies were transported one by one by the reloading machine.

The experimental arrangement is shown in Fig. 1. The irradiations were implemented in the 8.9 mm diameter central hole of the assemblies. A 13 m long special tube guiding the sample holder capsules was lowered down by the crane and fitted to the headpiece of the fuel assembly. Through this tube a long steel wire was led, on which a chain of 10 capsules was attached, each containing 2 cm diameter 0.5 mm thick metallic indium samples of 1.1 to 1.5 g masses. The stainless steel capsules were of 7.5 mm diameter to be introduced into the central hole for axial scanning. The indium samples were folded up and inserted in the capsules for irradiation. The capsules were positioned at 27.5 cm (centre-to-centre) from each other thus the 255 cm long chain of capsules covered the full "active" (UO_2 filled) length of the assembly.

Table 1. Hard γ radiation (>1 MeV) emitting fission products of high cumulative yields and half-lives longer than 4 days.

Fission products	Half-life	Cumulative yield from ^{235}U (%)	γ-rays above 1.08 MeV	Intensities (%)
^{106}Ru-^{106}Rh	372d	0.4	1.13	0.42
			1.19	0.06
			1.56	0.17
			1.80	0.03
			2.11	0.04
			2.36	0.02
^{134}Cs	2.06y	6.75(^{133}Cs)	1.17	1.80
			1.37	3.04
^{140}Ba-^{140}La	12.75d	6.3	1.60	95.5
			2.35	0.86
			2.52	3.44
^{144}Ce-^{144}Pr	284.5d	5.48	1.39	0.06
			1.49	2.61
			2.19	6.63
^{154}Eu	8.586y	0.164(^{153}Eu)	1.27	35.5
			1.60	1.67

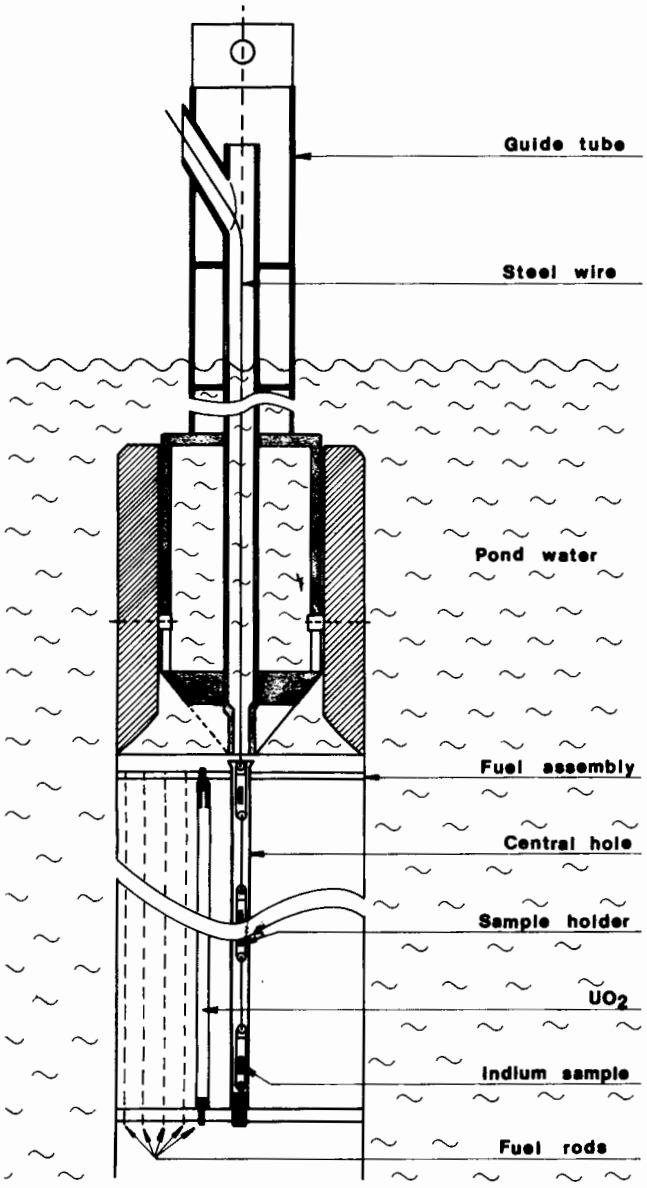

Fig.1 The experimental arrangement

Irradiation times varied between 1 to 10 hours. After irradiations the indium foils were unfolded to become flat again and placed in contact with a 35 cm^2 Ge(Li) detector surface. The gamma-spectra of the samples were taken by a Silena CATO 4K MCA. Counting periods were 5000 or 10000 s.

In addition to isomer activation by (γ,γ') process neutron activation also took place because of the n-emission of the spent fuel. The $^{116}In^m$ activity formed by n-capture was also identified through its gamma-spectrum, but the 336 keV peak of $^{115}In^m$ could easily be resolved and distinguished from the peaks of $^{116}In^m$, or they can be eliminated by covering the samples by Cd foil. The utilization of neutron signatures of indium sample for nuclear material safeguards purposes will be published elsewhere.

The data of the investigated fuel assemblies of 3.6% initial enrichment are given in Table 2.

Table 2. Data of fuel assemblies investigated. The count rates of 336 keV γ-peak of the In samples positioned at midpoint of fuel assemblies are extrapolated to saturation and corrected for delay and counting periods. (Statistical uncertainties related to 90% confidence level are in brackets.)

Fuel assembly serial number	BU MWd/KgU	Cooling time days	Specific count rate cps/g
13616	30.29	48	1.35(0.12)
13673	31.99	49	1.28(0.08)

Results and Discussion

The specific count rates of the 336 keV γ-peak of the samples positioned at 10 heights along the fuel assemblies were extrapolated to saturation and corrected for the time delay until the counting period started. The intensity decay of the sample during the counting period was also taken into account. The axial profiles taken along the assemblies is this way are shown in Fig. 2. The error bars represent statistical

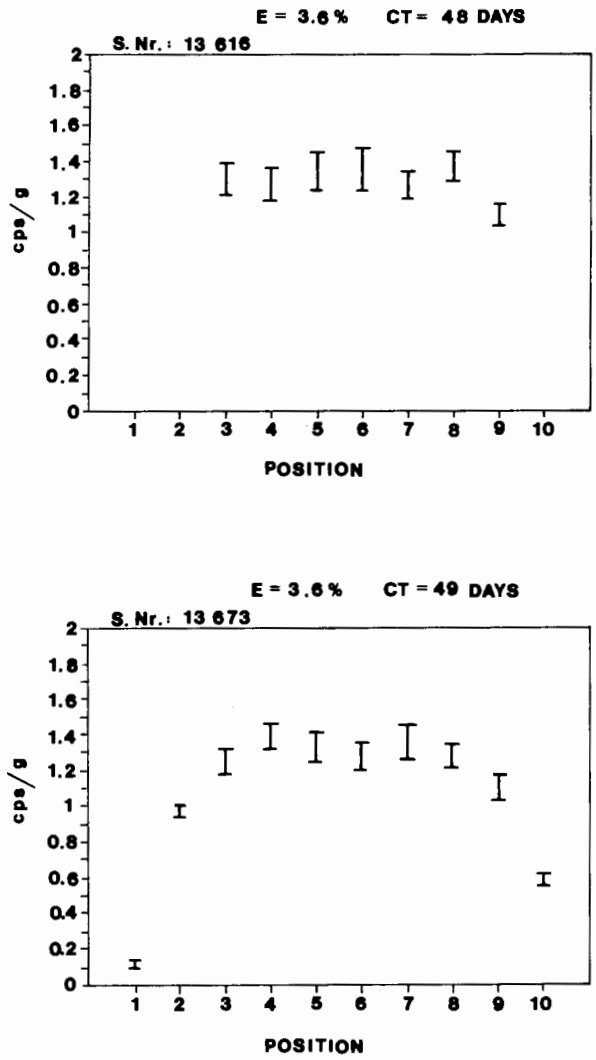

Fig.2 Axial profiles of the assemblies taken with 336 keV gamma signatures of $^{115}In^m$.

uncertainties related to 90% confidence level. The measurement positions are numbered from the top to the bottom of the fuel assemblies. Signatures measured at the midpoint (6. position) are indicated in Table 2.

According to the calculations[5], the intensity ratio of ^{140}Ba-^{140}La to all fission product gammas of energies above 1.08 MeV emitted by spent WWER-440 fuel is about 84% at 48 days cooling time. If the experiments had been carried out after 2 weeks cooling, the ^{140}Ba-^{140}La intensity ratio would have been 94%, whereas at the same time the isomeric activity would have been 6.3 times higher than was in the present case. So the gamma-signatures of the assemblies are characteristic of the ^{140}Ba-^{140}La fission product at short cooling times. This means, that these profiles reflect the power distribution of the reactor preceding shutdown.

A comparison can be made with the previous estimate of the expected $^{115}In^{m}$ intensities based on mockup experiments modelling WWER-440 assembly configuration[4]. For 3 years irradiation period (about 30 MWd/kgU burnup) the estimate was 14 cps/g, which related to a well type NaI(Tl) scintillation detector of 35% efficiency for the 336 keV gammas, 30 min irradiation of indium and 2 weeks cooling time. Our data are related to a Ge(Li) detector of 2% efficiency for this energy and 7 weeks cooling time, extrapolated to saturation. Making the comparison it can be observed, that the ^{140}Ba content reduces to 6% after 35 days cooling, the Ge detector efficiency is 5.7% of that of the well crystal, so 14 cps/g reduces to 0.048 whereas extrapolating to saturation results in 0.65 cps/g which is about one half of the measured value. The discrepancy may be due either to uncertainties of the estimation or to (n,n') contribution. To confirm this, further experiments are in progress. If the latter is proved, it will mean that the time limit should be more strict, i.e. the assay should be carried out 8-10 days after shutdown.

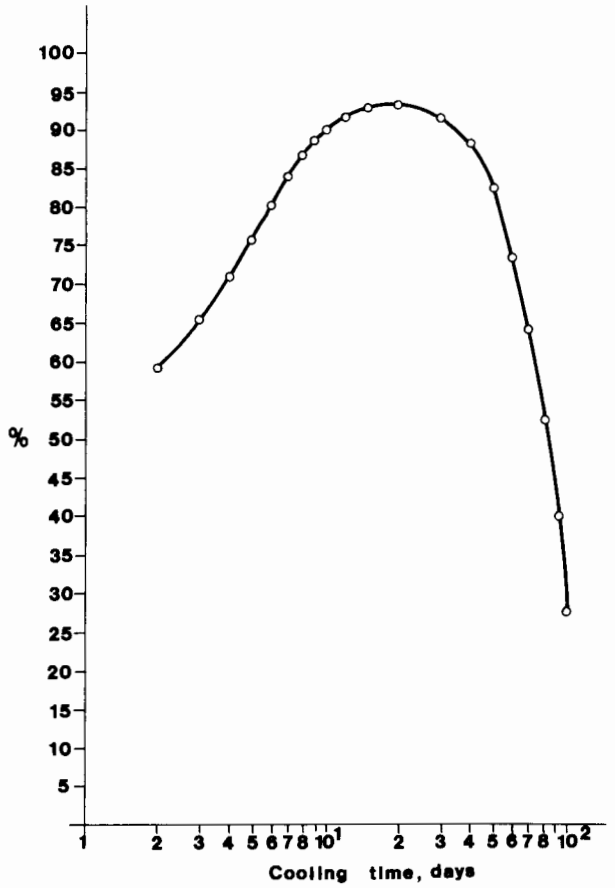

Fig.3 Intensity ratio of ^{140}Ba-^{140}La to all fission product γ-s above 1.08 MeV emitted from spent WWER-440 assembly vs. cooling time.

Conclusion

It can be concluded from the experiments, that hard gamma radiation from fission products can be scanned by a novel activation method which is sensitive first of all to ^{140}Ba fission product, shortly after reactor shutdown.

Fissile content in spent fuel assemblies can be deduced by combining these results with an independent information on local neutron flux.

The fuel assemblies can be measured in a few days after discharge from the core, in contrast with gamma spectroscopy, where usually at least a year is needed.

The scanning can be performed in the central hole of WWER-440 spent fuel assemblies thus shortening the time necessary for taking azimuthal distribution by other methods.

The method is simple and sensitive to inner rods which are out of reach of usual gamma-spectroscopy.

The features and advantages mentioned above make the method promising for reactor operational purposes as well as in the safeguards of nuclear materials.

Acknowledgement

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