Studies of Nuclear Fuel by means of Nuclear Spectroscopy Methods

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Licentiate thesis

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Abstract

This paper, which is a thesis for the title 'teknologie licentiat', is a summary text of several works performed by the author regarding spectroscopic measurements on spent nuclear fuel. Methods for determining the decay heat of spent nuclear fuel by means of gamma-ray spectroscopy and for verifying the integrity of nuclear fuel by means of tomography is presented. A summary of work performed regarding gamma-ray detector technology for studies of fission gas release is presented.

List of papers

1. P. Jansson, A. Håkansson, A. Bäcklin and S. Jacobsson, 'Gamma-ray Spectroscopy Measurements of Decay Heat in Spent Nuclear Fuel ', To be published, (2000)

2. S. Jacobsson, A. Håkansson, P. Jansson, A. Bäcklin, 'Experimental Investigation of a Tomographic Method for Verification of the Integrity of Spent Nuclear Fuel', Submitted for publication in Nuclear Technology, (2000)

3. P. Jansson, S. Jacobsson, A. Håkansson, A. Bäcklin, 'A Feasibility Study of BGO Scintillation Detectors for Tomographic Measurements on Nuclear Fuel ', ISV-1/2000, (2000)

4. I. Matsson, P. Jansson, B. Grapengiesser, A. Håkansson, A. Bäcklin, 'Fission Gas Release Determination Using an Anti-Compton Shield Detector', Nuclear Technology, NUTYBB **122** (3) 276-283 (1998)

Abstracts of papers

Paper 1

A method for determining the residual thermal power in spent nuclear fuel using gamma-ray spectroscopy has been applied on 35 boiling water reactor and 34 pressurized water reactor spent fuel assemblies. Measurements performed from 1995 through 1999 have shown that the method can be used to determine the residual thermal power with an uncertainty of about 2% for BWR and about 2.5% for PWR spent fuel.

Paper 2

A tomographic method for verification of the integrity of used LWR fuel has been experimentally investigated. The method utilises emitted gamma rays from fission products in the fuel rods. The radiation field is recorded in a large number of positions relative the assembly, whereby the source distribution is reconstructed using a special-purpose reconstruction code. An 8×8 BWR fuel assembly has been measured at the Swedish interim storage CLAB, using installed gamma-scanning equipment modified for the purpose of tomography. The equipment allows the mapping of the radiation field around a fuel assembly with the aid of a germanium detector fitted with a collimator with a vertical slit. Two gamma-ray energies were recorded, 662 keV (^{137}Cs) and 1274 keV (^{154}Eu). The intensities measured in 2520 detector positions were used as input for the tomographic reconstruction code. The results agreed very well with simulations and significantly revealed a position containing a water channel in the central part of the assembly.

Paper 3

A study of BGO detectors has been performed. The purpose of the study was to determine geometrical shape of the scintillator crystals in order to be suited for use in tomographic measurements on nuclear fuel. Computer calculations using Monte Carlo techniques were used. High count-rate experiments were performed on three nuclear fuel assemblies with the shapes of the crystals determined by the calculations. The resulting characteristics of the detectors show that they are suitable in a tomographic measurement.

Paper 4

Poolside measurements of fission gas release (FGR) in fuel pins have been made using gamma-ray spectroscopy with a Ge detector, measuring ^{85}Kr activity in the fuel rod plenum. The gamma-ray energy spectra from irradiated nuclear fuel are characterized by prominent Compton distributions that can obscure the weak 514-keV ^{85}Kr peak. To improve the sensitivity, the detector has been provided with an anti-Compton shield of six $Bi_3Ge_4O_{12}$ detectors. Laboratory tests of the detector system showed that the maximum peak-to-Compton (p/c) ratio was improved by a factor of $\approx$6. The results of the poolside measurement p/c ratio showed a somewhat smaller improvement (a factor of $\approx$4) because of scattered gamma radiation from the surrounding material. However, the precision in the poolside FGR measurements was improved substantially utilizing the Compton shield.

1 Introduction

Spent fuel from nuclear facilities in Sweden is planned to be finally stored in a deep geological repository, marking the end-point of a long process. In designing storages for spent fuel, care must be taken so that the fuel is separated from the biosphere for sufficiently long times. The Swedish concept is to encapsulate the spent fuel assemblies in steel and copper canisters embedded in bentonite clay at a depth of about 500 meters, ref. [1]. According to the present planning of the repository, the spent fuel will be stored in the order of 30 to 40 years prior to the final encapsulation. Eventually, about 35.000 spent fuel assemblies will be located in the deep geological repository, ref. [2].

To ensure that the clay can function as a buffer between the spent fuel and the biosphere, the temperature of the surface of the canisters must remain within certain limits. Thus, to cope with the demands of the repository, a fast method of measuring the decay heat of the spent nuclear fuel using a technique that as little as possible disturbs the fuel matrix would be beneficial. In addition, safeguard issues require verification of operator declared values on e.g. fuel integrity, burnup, enrichment and cooling time. Independent measurements to verify these parameters are therefore of interest both for authorities and for operators. The use of non-destructive measurements implies several advantages compared to destructive methods. First, large economical savings can be made if the fuel does not have to be taken appart. Second, the security in handling of the fuel is increased when the fuel can be measured as a whole. This thesis focuses on two types of non-destructive measurement methods.

Non-destructive measurements of nuclear fuel using gamma-ray spectroscopy involves measuring on a highly heterogenous and highly radioactive source. This implies that self absorption of the gamma radiation is to a large degree direction dependent and the highly radioactive measuring object implies that a fast data aquisition system to collect gamma-ray spectra must be used in combination with a carefully designed detector and collimator system.

This licentiate work is a summary of investigations performed in the following three areas.

1. A project aiming at finding an experimental method to determine the decay heat of spent nuclear fuel. This work comprises theoretical and experimental work summarized in papers 1.

2. The second is a project aiming at the development of a tomographic method to verify the activity content in nuclear fuel. Such a method

can be used for integrity verification. Computer simulations have been performed in order to develop the gamma-ray detectors used for the tomographic method. This work was complemented with some experimental work. Papers 2 and 3 describes this work.

3. The third area represents development work of detectors with a potential for improving the quality of mearurements in nuclear technology. The work is reported in paper 4.

2 Decay heat measurements

The magnitude of the decay heat of spent nuclear fuel is of importance for the design of a safe encapsulation of spent fuel for a deep geological storage. The surface temperature of the canister is limited to 80 degrees C and the maximum total decay heat of the fuel assemblies placed within the canister is accordingly limited to 1800 W, ref. [2]. After 40 years of cooling, typical decay heat of a spent fuel assembly are in the order of 100 W and 500 W for BWR and PWR fuels, respectively. There may, however, be substantial fluctuations between fuel assemblies depending on their burnup and power history in the core. It is anticipated that the encapsulation process can not rely entirely on decay heat calculations only. Calculations involves various models which in principle may fail when complicated power histories or other circumstances are involved. In addition, large financial savings can be achieved if precise measurements of the decay heat can be performed, since each canister is expensive and must be optimally utilised. Therefore, a method of experimentally determining the decay heat of spent nuclear fuel by means of fast gamma-ray measurements has been developed and evaluated.

The decay heat of spent nuclear fuel is generated by radioactive decay of the elements produced by the neutron irradiation of the fuel during operation. Decay heat can be calculated with computer codes, for instance Origen (ref. [3]), which has been used extensively in this work to calculate the contribution to the decay heat by various sources. Origen is a computer code capable of calculating the dynamics of the isotope production and depletion as the fuel is irradiated in the reactor. The code is part of the SCALE package (ref. [4]) which is used in computer analyses for licensing.

A summary of the contributions to the decay heat, calculated by using the Origen code, is shown in table 1. Some elements are long-lived and contribute to the decay heat for a long time while other elements are relatively short-lived and dominate the decay heat at short cooling times. After about 40 years of cooling time, heavy elements and actinides produced in the irradi-

ation process contribute by about 30% to the decay heat mostly by means of α decay and this contribution increases with time due to their long half-lifes. Fission products contribute with about 70% to the decay heat at the time of encapsulation, mostly by means of β decays and γ deexciations. This contribution decreases with time. Elements produced from irradiated construction materials in the fuel, e.g. ^{60}Co, do not contribute significantly to the decay heat.

When the fuel has been stored for about 40 years, the only significant gamma radiating isotope is ^{137}Ba, following β^- decay of ^{137}Cs. About 35%, of the total heat generated in spent nuclear fuel arises from the decay of ^{137}Cs. Using the Origen code, it is shown that the relative contribution (f) to the heat from the decay of ^{137}Cs is nearly constant with respect to variations in the irradiation history of the fuel and in the cooling time, see table 1. Therefore, by measuring the gamma radiation of 662 keV energy from ^{137}Ba, a reasonably good measure of the decay heat can be expected to be obtained.

In order to increase the accuracy of the determined decay heat, a first-order correction to the assumption of constant heat fraction of ^{137}Cs+^{137}Ba is made. The factor f has been determined by using the Origen code. Using the computer results, a linear function of burnup and cooling time is used to interpolate the correction factor for a particular fuel assembly.

A number of fuel elements have been measured both with regards to the emitted gamma radiation and with regards to the decay heat using a calorimetric method. For the gamma measurements, a high purity germanium detector system (ref. [5]) was used to measure the average intensity of ^{137}Cs over the whole fuel assemblies. The decay heat measurements was performed by the staff at the interim storage for spent fuel (CLAB). Using data from these measurement, a first objective of the project has been to establish the accuracy with which the decay heat can be measured. Paper 1 and references [6, 7] gives a detailed report of the achievements in this project.

The detector system enabled measurements of the 662 keV gamma radiation at a counting rate of 1000 cps in the full absorption peak. The total counting rate was about 100 kcps. Total measuring time was typically 10 minutes. A useful feature of the method is that burnup and cooling time is determined simultaneously with decay heat, see ref. [5].

It is shown in the papers that the decay heat can be determined with a relative uncertainty of about 3% for BWR and PWR spent fuel assemblies by using the gamma radiation from ^{137}Ba.

Table 1: Relative contributions from various isotopes to the decay heat as a function of cooling time (t), in percent. Calculated with Origen-S for a 8×8 BWR fuel with burnup of 37 GWd/tU and 2.6% initial ^{235}U enrichment. 4 power cycles were used.

t [y]	**10**	**15**	**20**	**25**	**30**	**35**	**40**	**45**	**50**
^{90}**Sr**	4.8	4.9	4.8	4.7	4.6	4.4	4.3	4.1	3.9
^{90}**Y**	22.7	23.4	23.1	22.5	21.8	21.1	20.4	19.7	18.8
^{134}**Cs**	4.7	1.0	0.2	0	0	0	0	0	0
^{137}**Cs**	7.5	7.8	7.8	7.6	7.5	7.3	7.1	6.9	6.6
^{137m}**Ba**	25.1	26.0	25.9	25.4	25.0	24.3	23.6	22.9	22.1
^{154}**Eu**	2.3	1.8	1.3	1.0	0.7	0.5	0.4	0.3	0.2
^{238}**Pu**	9.1	10.2	10.9	11.5	12.2	12.9	13.5	14.1	14.7
^{239}**Pu**	0.6	0.7	0.8	0.9	0.9	1.0	1.1	1.2	1.3
^{240}**Pu**	1.3	1.6	1.8	1.9	2.1	2.3	2.5	2.8	3.0
^{241}**Am**	4.5	6.8	9.0	11.1	13.1	15.2	17.3	19.2	21.3
^{244}**Cm**	15.5	14.9	13.7	12.5	11.4	10.3	9.3	8.3	7.5

3 Tomography

A tomographic method to determine the gamma activity distribution within a nuclear fuel assembly has been developed at the Department of Radiation Sciences (ISV). This method can be used in safeguard applications to verify the integrity of the fuel, as paper 2 describes.

Tomography means that the radioactivity distribution of the interior of a fuel assembly may be deduced from measurements of the gamma radiation emitted. The gamma emission from the fuel is measured using a well defined collimator geometry in about 50 angular positions and 50-100 lateral positions for each angular position around the fuel and from this information the gamma activity distribution within the fuel is reconstructed. The basic technique used here for reconstruction is single photon emission computed tomography.

The highly heterogenous material composition of the nuclear fuel neccesitates the use of specially developed tomographic algorithms, ref. [8]. The high-level radiation field around a fuel assembly implies that the collimator and detector system must be carefully designed in order to achieve a well defined direction of the beam of gamma radiation while measuring with relatively high count rate to achieve good counting statistics.

Thus, the development of an optimised measuring set-up has been of ut-

most importance. This work together with testing of various detector types and electronics have been carried out at the department of Radiation Sciences. For this work, Bismuth Germanate (BGO) scintillator detectors have been used. The work done by the author mainly involves detector simulations using Monte Carlo techniques and verifying measurements to determine characteristics of the detectors to be used in the tomographic equipment. Calculations has been performed in order to determine optimum shape of the scintillation crystals to have a large gamma detection efficiency and at the same time allow for a close spacing between detectors. An investigation on detection efficiency, peak-to-Compton ratio and of how the cross-talk between closely situated detectors depend on geometry was performed using Monte Carlo simulations.

Experimentally, high count-rate measurements on three nuclear fuel assemblies was performed using BGO detectors manufactured according to the calculated optimum shape.

This work is presented in papers 2 and 3.

4 Gamma-ray detector technology: HPGe with Compton suppression

In the nuclear industri, gamma radiation detection technology is constantly being developed in order to increase understanding about the behaviour and performance of nuclear fuel. Several studies have been performed to characterise and improve the performance of detector techniques.

Some fission products, e.g. krypton and xenon, are gaseous. The nuclear fuel is designed to contain all fission products produced in the fission process and most of the gaseous products are retained within the fuel matrix. However, when the tempereature increses above about 1000 degrees C, some gases may be released from the fuel matrix, ref. [9]. Using high resolution gamma-ray spectroscopy, this fission gas release within the nuclear fuel may be measured and a database with correlations between burnup of the fuel assemblies and measured fission gas release can be established. Especially the peak of 514 keV from the fission product ^{85}Kr has been used for such studies of the release into the fuel rod plenum. Due to the relatively small intensity of the Kr peak and the fact that its energy of 514 keV is close to the 511 keV positron annihilation energy, the use of high-resolution spectroscopy is necessary. To further increase the possiblity to detect the Kr peak, an anti-Compton suppression system may be used. The testing of such a system is described in paper 4.

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