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A nondestructive method for discriminating MOX fuel from LEU fuel for safeguards purposes

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Abstract

Plutonium-rich mixed oxide fuel (MOX) is increasingly used in thermal reactors. However, spent MOX fuel could be a potential source of nuclear weapons material and a safeguards issue is therefore to determine whether a spent nuclear fuel assembly is of MOX type or of LEU (Low Enriched Uranium) type.

In this paper, we present theoretical and experimental results of a study that aims to investigate the possibilities of using gamma-ray spectroscopy to determine whether a nuclear fuel assembly is of MOX or of LEU type.

Simulations with the computer code ORIGEN-ARP have been performed where LEU and MOX fuel types with varying enrichment and burnup as well as different irradiation histories have been modelled. The simulations indicate that the fuel type determination may be achieved by using the intensity ratio $^{134}Cs/^{154}Eu$.

An experimental study of MOX fuel of 14×14 PWR type and LEU fuel of both 15×15 and 17×17 type is also reported in this paper. The outcome of the experimental study support the conclusion that MOX fuel may be discriminated from LEU fuel by measuring the suggested isotopic ratio.

1. Introduction

MOX fuel was first used in a nuclear reactor in 1963 (World Nuclear Association, 2006) but came in general use first during the 1980s. Today MOX is used in some 30 reactors in Germany, France, Belgium and Switzerland. Japan is planning to use MOX fuel in one third of the Japanese reactors by 2010. Of the fuel used today in thermal reactors, MOX accounts for approximately 2% and the proportion is expected to increase to about 5% by 2010.

Ordinary LEU fuel, discharged from an LWR reactor, contains about one percent of plutonium by weight. Typically 2/3 of this is in the form of the fissile isotope ^{239}Pu. When making MOX fuel, this remaining plutonium content is extracted from the discharged fuel at a reprocessing plant. The plutonium is then mixed with depleted uranium, $PuO_2 + UO_2$, to form the mixed oxide fuel.

The increase in the use of MOX fuel may make the verification of spent fuel characteristics more extensive since there is also a safeguards interest to clarify whether a fuel assembly is of LEU or MOX type. Assuming that the intention is to use the isotopic inventory in the MOX fuel for weapons production, a one cycle MOX fuel with low burnup could be substituted with a multiple cycle LEU fuel with fairly high burnup in order to mislead authorities. This scenario has been recognized by the International Atomic Energy Agency (IAEA) (IAEA, 2003), and methods have been suggested, see e.g Lebrun et al. (2001).

Currently there are several, both destructive and nondestructive, quantitative methods addressing the problem with verifying spent fuel (IAEA, 2003a). Among the methods are high-resolution gamma-ray spectroscopy (HRGS) and active and passive neutron measurements, such as the Fork detector (FDET). In the present report we have

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studied the use of spectroscopic gamma-ray information at facilities where HRGS equipment is installed, typically large interim storages or likewise. All Swedish BWR nuclear power plants as well as the interim storage, CLAB, have mechanical equipment installed allowing to perform this type of measurements. The method is however not dependent on this type of installation, any kind of equipment capable of acquiring spectra with high resolution and that offers a stable measuring geometry will work, for instance the submergible detector system reported in Andersson and Grapengiesser (2002).

The technique presented in this paper has been studied and developed in Sweden at the Department of Nuclear and Particle Physics (former Department of Radiation Sciences) at Uppsala University in collaboration with the Swedish Nuclear Power Inspectorate (SKI) (Bäcklin et al., 1994). The work is a continuation of earlier efforts to apply spectroscopic gamma-ray measurements for fuel parameter verification and determination, see for instance (Berndt, 1988), whereas previous techniques have been focused on verifying burnup, cooling time and declared irradiation history (Willman et al., 2006).

2. Feasibility studies of spectroscopic gamma-ray measurements for discriminating MOX and LEU fuel

2.1. Isotope production in the fuel assemblies

In a fuel assembly, many different fission products are produced during reactor operation. After a cooling time of a few years, basically three principal gamma emitting isotopes dominate the gamma-ray spectrum, ^{137}Cs, ^{134}Cs and ^{154}Eu. The amount of fission products in a fuel assembly at a given time depends on the fuel parameters burnup, cooling time and initial isotopic composition. In addition, the irradiation history gives rise to measurable effects.

The basic idea motivating this study is the fact that the concentration of the fission product ^{154}Eu is different in irradiated MOX and LEU fuel, respectively. This is mainly due to the different fission yields in ^{235}U and the two plutonium isotopes, see Table 1.

In this work, a number of simulations have been performed with ORIGEN-ARP (Bowman and Leal, 2000) in order to determine how the differences in isotope buildup between LEU fuel and MOX fuel depend on various fuel parameter. The basic isotopic composition for the MOX fuel was taken from Thorne et al. (2002), and is shown in Table 2. The MOX fuel was modelled with a total plutonium content of 4.67%, which corresponds to a fissile plutonium content of 3%. For the modelling of the LEU fuel, 3% enrichment of ^{235}U was used as shown in Table 2.

Table 1
The thermal fission mass chain yields for ^{137}Cs, ^{134}Cs and ^{154}Eu as reported in England and Rider (1994)

Isotope	Thermal fission mass chain yield (nuclei per 100 fissions)			$T_{1/2}$ (years)
	^{235}U	^{239}Pu	^{241}Pu	
^{137}Cs	6.19	6.61	6.65	30.1
^{134}Cs	7.87	7.68	7.92	2.1
^{154}Eu	0.074	0.26	0.38	8.6

The half lives of the isotopes are also given.

Table 2
The basic plutonium and uranium isotopic vectors used in the studies of the two types of fuel

Isotope	Weight percent	
	MOX	LEU
^{238}Pu	2.5	–
^{239}Pu	54.7	–
^{240}Pu	26.1	–
^{241}Pu	9.5	–
^{242}Pu	7.2	–
^{234}U	0.00119	0.0267
^{235}U	0.25	3
^{236}U	–	0.0138
^{238}U	99.74881	96.9595

The isotopic vector for MOX was adapted from Thorne et al. (2002).

The irradiation histories used in the calculations were based on power cycles of 335 days with 30 days maintenance shutdown periods which approximate the irradiation histories of the experimentally measured PWR fuel. The irradiation histories studied comprised of one, three and five such power cycles.

The production of ^{137}Cs is mentioned here as it is a well known marker for burnup. It is however not explicitly used in this work.

2.1.1. Production of ^{134}Cs

The isotope ^{134}Cs, with a half life of 2.1 years, is created by neutron capture in the stable fission product ^{133}Cs, which is produced linearly with burnup. This process makes the intensity of ^{134}Cs, in principle, a quadratic function of burnup.

Furthermore, the enrichment of the fuel has an effect of the buildup of ^{134}Cs because a higher initial enrichment requires a lower neutron flux density to accomplish a given burnup. This effectively leads to a lower production of ^{134}Cs in high-enriched fuel assemblies.

Fig. 1 shows the buildup of ^{134}Cs as a function of burnup for MOX and LEU fuel, respectively. The content of ^{134}Cs in LEU fuel is slightly higher when compared to the MOX fuel. This is because the fission and absorption cross-sections of ^{239}Pu are larger as compared to those of ^{235}U. Accordingly, when the same power level is considered, the thermal neutron flux density is lower for the MOX fuel.

2.1.2. Production of ^{154}Eu

The production of ^{154}Eu involves both beta decay and neutron capture from five different mass chains (Berndt, 1988), ending with neutron capture in the stable isotope