CHARACTERIZATION OF SPENT FUEL ASSEMBLIES FOR STORAGE FACILITIES USING NON DESTRUCTIVE ASSAY

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Abstract

Many non destructive assay (NDA) techniques have been developed by the French Atomic Energy Commission (CEA) for spent fuel characterization and management. Passive and active neutron methods as well as gamma spectrometric methods have been carried out and applied to industrial devices like PYTHON™ and NAJA. Many existing NDA methods can be successfully applied to storage, but the most promising are the neutron methods combined with on line evolution codes. For dry storage applications, active neutron measurements require further R&D to achieve accurate results. Characterization data given by NDA instruments can now be linked to automatic fuel recognition. Both information can feed the storage management software in order to meet the storage operation requirements like: fissile mass inventory, operators declaration consistency or automatic selection of proper storage conditions.

1. INTRODUCTION

France, which is a major actor in nuclear industry, owns plants and facilities that cover the whole cycle of nuclear materials. Part of this cycle is: handling, reloading, transporting, storing and reprocessing of spent fuels. So, for a long time, the French CEA has studied technologies for non destructive measurement and characterization of spent fuel assemblies. Many R&D programmes have been undertaken in partnership with major French nuclear industry companies like EdF or COGEMA, the industrialization being performed by EURISYS MESURES[1]. Various characterization techniques have been developed for: safety criticality controls, core loading checking or Safeguards concerns. They have been implemented in various NDA instruments. In a first step, the physical methods involved in these NDA instruments are briefly described before their potential application for storage purposes is discussed. All known NDA techniques for spent fuel characterization use spontaneous or induced nuclear emissions. Gamma rays as well as neutron emissions are used [1].

2. PHYSICAL PRINCIPLES OF SPENT FUEL CHARACTERIZATION

2.1. Passive neutronic emission

Since the spontaneous neutron emission is linked to the burnup of the fuel with a power law, the burnup determination with passive neutron measurement is extremely accurate. The correlation law is:

$$BU = aNE^{b}\ , \qquad (1)$$

in which a is a constant slightly dependant on initial enrichment and b another constant close to 0.02 for usual irradiation histories. In addition, the constants (a,b) can be calculated using an evolution code taking into account initial enrichment and actual irradiation history of the assembly to

[1] The nuclear measurement company of the COGEMA group

be measured. As a result, relative error on neutron emission $^{\Delta NE}/_{NE}$ from the measurement leads to small burnup relative error:

$$\frac{\Delta BU}{BU} \cong 0.02 \frac{\Delta NE}{NE}, \quad (2)$$

2.2. Induced neutron emission

In nuclear fuel that contains fissile material, neutron emission can be induced by an external neutron flux. It could also be created by a high energy gamma intense flux (photo-fission) but, today, this technique has never been applied to spent fuel because of its cost and complexity. Neutron induced emission *INE* is linked to the multiplying factor k_{eff} by the relation:

$$INE = a \frac{Keff}{(1 - Keff)}, \quad (3)$$

Using pulsed neutron irradiation, the neutron detector measures on one hand the passive emission and on the other hand both induced and passive contributions. This NDA method is used for initial ^{235}U enrichment or for MOX characterization.

2.3. Total gamma emission

Gamma emission comes from fission products and activation products. After discharge of spent fuel assemblies from the reactor core, short lived fission and activation products are responsible for most of the total gamma emission which decreases very fast in the spent fuel. After several months, the total gamma emission *TGE* can be linked to the cooling time with the following correlation law:

$$CT = a(BU) \cdot \left(\frac{TGE}{BU}\right)^{b(BU)}, \quad (7)$$

$a_{(BU)}$ and $b_{(BU)}$ are quadratic functions of *BU*, determined by fitting of parametrical calculation results. Since the total gamma emission can be measured with simple and inexpensive detectors (e.g. ionisation chambers), the cooling time can be simply estimated.

Contrary to neutronic emission, gamma emissions can be collimated. This property is used to determine the relative irradiation profile along the assembly, in order to measure the burnup of the fuel extremities for safety criticality purpose. The average *BU* being known (using a neutron method), the total gamma profile is measured along the assembly with a collimated detector. The result of the scanning measurement is an array *TGE(z)*. The extremity burnup $EBU_{(0,z0)}$ is defined by the relation:

$$EBU_{(0,z0)} = BU \cdot \frac{z}{z_0} \cdot \int_0^{z_0} TGE(z) \cdot dz \Big/ \int_0^{z} TGE(z) \cdot dz \cdot F(CT), \quad (4)$$

This relation assumes that *TGE* is mainly composed of burnup proportionally produced gamma emitters. Actually, a correction factor *F(CT)* has to be used for short cooling times *CT*, because short lived fission products induce non proportionality. The following relation is convenient for PWR reactors:

$$F(CT) = 0.5 \cdot CT^{0.1}, \quad (6)$$

2.4. Gamma spectrometry

Many gamma emitters have interesting properties in order to characterize the spent fuel. Tables I and II, give an overview of several relevant isotope abundance and isotopic ratios to determine burnup and cooling time. Convenience of the different isotopes is displayed, with regard to the range of cooling times. Convenience of the gamma spectrometric methods is very dependent on the characteristics of the spent fuel. They have to be carefully selected to produce proper results. Anyway, because the slope of the correlation laws is always smaller for gamma emitters than for

neutron emission, burnup determination using passive neutron counting leads to more accurate results than gamma measurements.

TABLE I. OVERVIEW OF GAMMA SPECTROMETRIC BURNUP DETERMINATION

Isotope	**Correlation law**[2] :	**Cooling time**[3]		
	(example PWR 17x17 IE 3% CT 3 years)	**0 to 90 days**	**90 to 5,000 days**	**Over 5,000 days**
^{134}Cs	$a \cdot BU^2$ (a : 0)	+	-	0
^{137}Cs	$a \cdot BU$ (a : 3,000)	+	+	+
^{154}Eu	$a \cdot BU^b$ (a : 5, b : 2)	0	-	+
$^{134}Cs/^{137}Cs$	$a \cdot BU^b$ (a : 10^{-2}, b : 1)	++	+	0
$^{154}Eu/^{137}Cs$	$a \cdot BU^b$ (a : 10^{-3}, b : 1)	0	+	++

TABLE II. OVERVIEW OF GAMMA SPECTROMETRIC COOLING TIME DETERMINATION

Isotope	**Correlation law** : (example PWR 17x17 IE 3%)	**0 to 90 days**	**90 to 5000 days**	**Over 5000 days**
$^{144}Ce/^{137}Cs$	$^{144}Ce/^{137}Cs = a.exp^{b.CT}$ (a : 10, b : -0.002)	+	++	-
$^{106}Ru/^{137}Cs$	$(^{106}Ru/^{137}Cs)/TC^{0.5} = a.exp^{b.CT}$ (a : 1 , b : -0.02)	++	+	-

3. EVOLUTION OF THE FUEL UNDER AND AFTER IRRADIATION

The composition of the isotopes in the fuel changes during irradiation and cooling. Different nuclear reactions and decay lead to production and destruction. The evolution of the fuel components results from neutron fission captures, (n,2n) reactions and (α, β) radioactive decay. The following differential equation describes this complex process:

$$\begin{aligned}
&\frac{dN(t)}{dt}[A,Z] = \\
&\left[\Phi \ \sigma_{c(El,BU)} \ N(t)\right]_{[A-1,Z]} + \left[\Phi \ \sigma_{n,2n(El,BU)} \ N(t)\right]_{[A+1,Z]} \\
&+ \left[\lambda_{\beta+} \ N(t)\right]_{[A,Z+1]} + \left[\lambda_{\beta-} \ N(t)\right]_{[A,Z-1]} \\
&+ \left[\lambda_{\alpha} \ N(t)\right]_{[A+4,Z+2]} + \left[\lambda_{TI} \ N(t)\right]_{[A_{métastable},Z]} \\
&- \Phi\left[\left(\sigma_{c(El,BU)} + \sigma_{f(El,BU)} + \sigma_{n,2n(El,BU)}\right) \ N(t)\right]_{[A,Z]} \\
&- \left[\left(\lambda_{\beta+} + \lambda_{\beta-} + \lambda_{\alpha} + \lambda_{TI} + \lambda_{SF}\right) N(t)\right]_{[A,Z]}
\end{aligned} \quad , \qquad (8)$$

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2 In the correlation examples, activities are given in Curie/g, burnup in GW·d/t and cooling times in days

3 0 means « really not appropriate », - means « not appropriate »,
+ means « can be successfully used », ++ means « recommended »

In this relation, $N(t)_{[A,Z]}$ is the atomic abundance of the isotope with atomic number Z and mass number A. Φ is the neutron flux, σ_c, σ_f, $\sigma_{(n,2n)}$ are the cross sections respectively for capture, fission and (n,2n) reactions, λ_i the decay constants for β^+, β^-, α, spontaneous fission and isomeric transition reactions.

Contrary to decay constants, which are intrinsic values, neutron cross sections depend on irradiation conditions. In the relation (8), neutron cross sections are condensed into one energy group representative of the neutron spectrum and auto-protection effects. During fuel life these parameters change. As a result, for every kind of fuel, neutron cross sections have to be tabulated with regard to initial enrichment and burnup.

This important work has been carried out to provide the French reprocessing company COGEMA with a calculation tool CESAR .

[2]. This code is able to calculate the components and emissions of fuel taking into account their particular geometry, initial enrichment and irradiation histories. An on line version of this code has been developed for spent fuel NDA characterization purposes.

4. APPLICATIONS OF NDA METHODS

NDA methods have been applied by CEA in several plants and equipment. They have to be shared in two families depending whether they are applied under water or in air.

4.1. NDA in air

In air, gamma spectrometric methods are easier to carry out and give better results than under water. As an example, we can describe the spectrometric analysis of the spent fuel in the head end of the COGEMA La Hague reprocessing plant [1]. This device is composed of two HPGe detector with collimators. Burnup and cooling time are determined using respectively $^{134}Cs/^{137}Cs$ and $^{144}Ce/^{137}Cs$ ratio. Scanning the fuel between the detectors leads to a very accurate burnup profile measurement. Neutron passive counting combined with an on line evolution code, is also used and gives very accurate burnup determination (within 2% considering one single assembly).

It has also been shown [3], that neutron measurement in air combined with a correlation law can be used to determine the plutonium amounts of spent fuel using a relation like:

$$M_{Pu} = aNE^b \quad , \tag{9}$$

where M_{Pu} is the plutonium mass, NE the neutron emission rate and a is a function of the initial enrichment IE and cooling time CT in the following relation:

$$a = a_0 + (a_1 + a_2 CT)IE \quad , \tag{10}$$

The constants a_n and b depend only on the assembly type and since, $a_2 << a_1 << a_0$, the influence of the cooling time and initial enrichment remains very small. This very simple method gives accurate results and it has been shown that, considering several assemblies, the total plutonium amounts can be determined within 1% for PWR assemblies. No information on irradiation histories neither than on burnup are necessary. In addition, no on line code has to be used.

4.2. Underwater measurements

Various techniques have been used for under water measurements but neutron measurements are particularly convenient. Two major R&D projects have been carried out by CEA giving birth to the PYTHON ™ device [4] and NAJA device [5].

4.2.1 The PYTHON ™ device

The PYTHON device has been developed in collaboration between EDF and CEA. Its main objectives are to measure the average and extremity burnup for safety criticality purposes. The PYTHON device is a combination of:

- a passive neutron measurement;
- a collimated total gamma measurement;
- an on line evolution code.

Figure 1 shows a schematic view of the two measurement heads that operate on top of the storage racks in the NPP ponds. The figure is a graphic output of a MCNP model of the measurement heads [6]. It is used to optimize the head's design, to precalculate the measurements yields and to parametrically calculate the multiplication factor k_{eff} of the fuel taking into account boron concentration in the water and burnup.

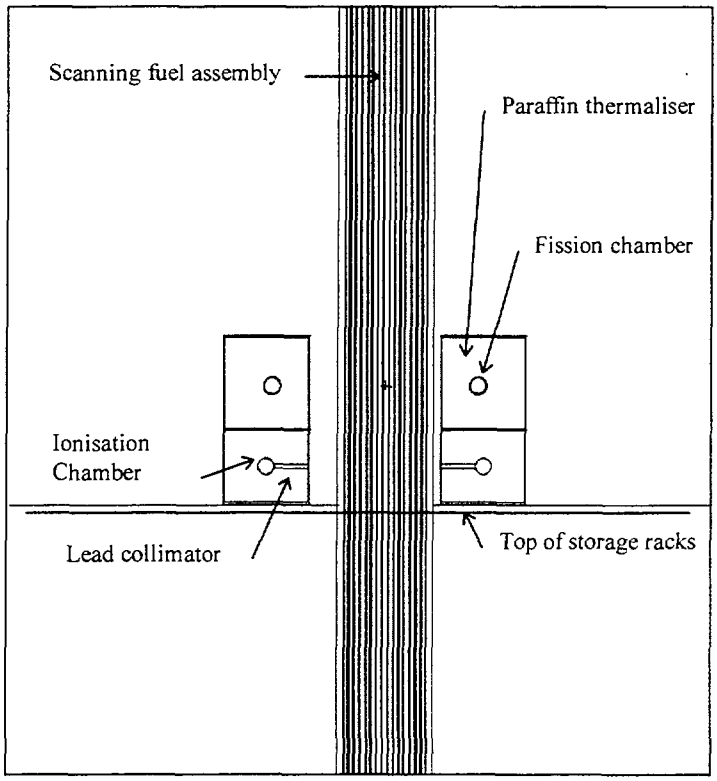

FIG. 1. The two detection heads of the PYTHON™ system

The neutronic yield is about 10^{-5} c/n·assembly^{-1}, so in order to achieve good statistical accuracy with low burnup, the detection heads are equipped with high efficiency fission chambers (1 c/n.cm^{-2}). The gamma detectors are simple ionization chambers with 10^{-9} A/Gy.h^{-1} efficiency.

Since the PYTHON device is intended to measure the average burnup, the neutron signal has to be representative of the entire fuel assembly. This means that contributions to the average signal have to originate from the whole fuel. FIG. 2 shows an example of the radial importance function of the fission chamber in the fuel section measured on a fuel mock-up in borated water [7]. Contrary to the gamma emissions that are absorbed when crossing fuel pins, the neutrons detected by the fission chamber originate from almost all the fuel section. In addition, since the two head signals are averaged, the sensitiveness to radial gradient for the burnup measurement is very low.

To take into account the neutron axial profile, fuel is scanned between the two heads and signals are averaged. However, despite neutron signals are acquired along the fuel, it is not possible to get from them a burnup profile. FIG. 3 shows the axial importance function of the detectors. It is clear

that almost a length of several tens of centimetres contributes significantly to the signal. As a result, the burnup profile has to be measured with the collimated total gamma detector and extremity burnup calculated using both gamma profile and average burnup using the relation 8.

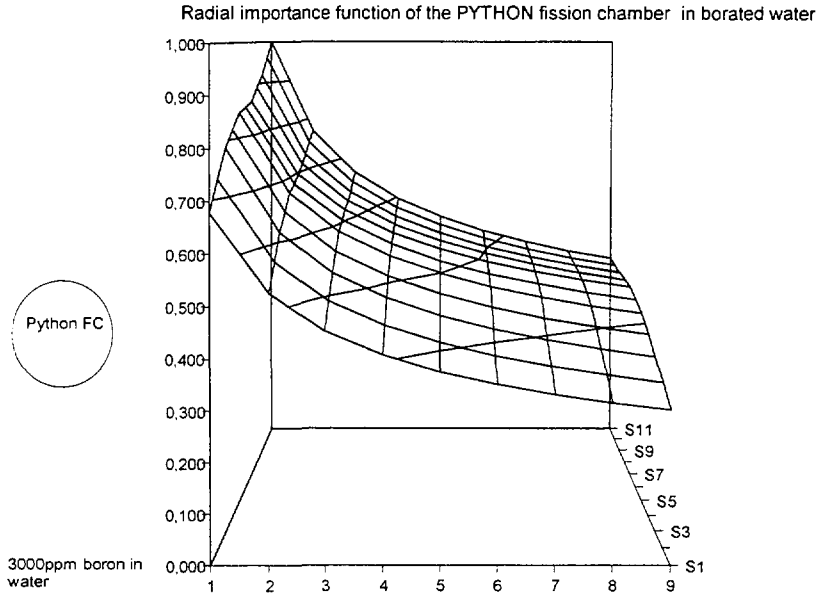

FIG. 2. Radial importance of Python fission chamber (9x11 pins mock-up)

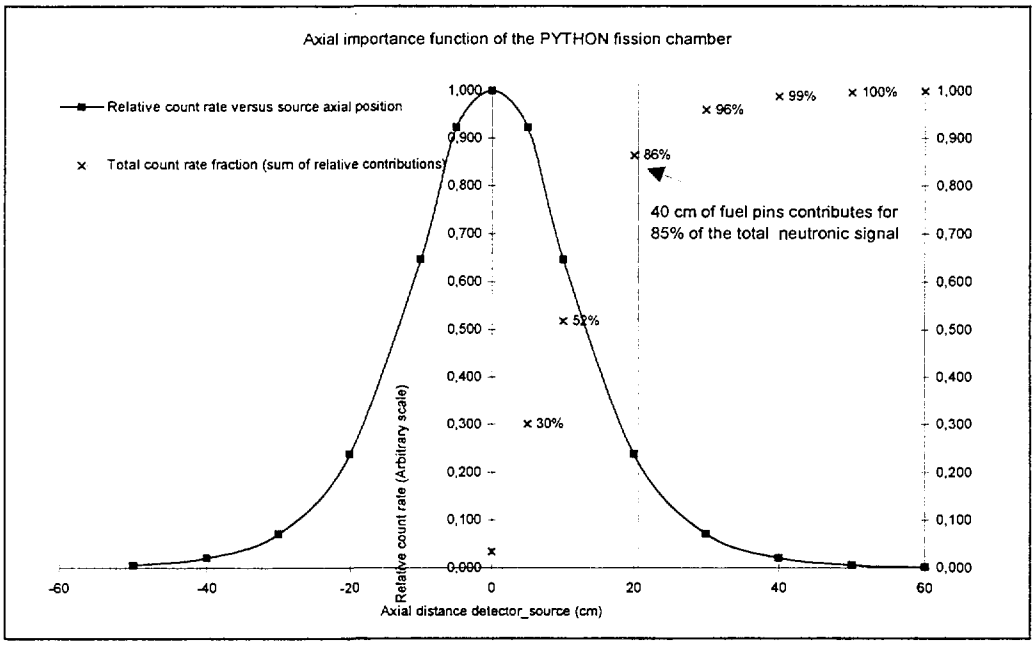

FIG. 3. Axial importance of Python fission chamber

For safety criticality purpose, the irradiation histories are supposed to be known as well as the initial components of the fuel. They are used as input data by the on-line evolution code that determines a correlation law (BU = f(NE)) for each fuel assembly. As a result, no standardization is required to determine the relevant correlation law to apply. Only a calibration is required to measure the detector yields. In order to avoid mistakes, the measured yields values are confirmed with MCNP calculations.

The PYTHON system has intensively been qualified using a prototype device with active mode capability at the Tricastin NPP and with industrial systems manufactured by EURISYS MESURES at the Gröhnde and Brökdorf NPPs. Table III summarizes the qualification range.

TABLE 3. PYTHON QUALIFICATION RANGE

	Tricastin	**Gröhnde**	**Brökdorf**
Number of assemblies	35	50	35
Fuel types	UOX PWR 17 x 17	UOX PWR 16 x 16	UOX PWR 16 x 16
Initial enrichments	1.8% to 3.7%	2.1% to 4%	1.8% to 3.7%
Cooling Time	80 days to 7.5 years	60 days to 6 years	500 days to 3.5 years
Reactivity (k_{eff})	0.4 to 0.7	Passive mode only	

The accuracy evaluated by a comparison with declared values for burnup and cooling time and with calculations for k_{eff} are as follows: on average burnup within 2%, on cooling time within 15% and on the multiplying factor k_{eff} within 3%.

At the moment another industrial system is delivered by EURISYS MESURES for the Gösgen NPP in Switzerland. Further R&D projects have been focused on extensions of the PYTHON capabilities in order to use burnup and reactivity measurements for core loading checks.

4.2.2. The NAJA device

The main objectives of the NAJA device consist in developing a measurement device which combines nuclear methods and video control in order to evaluate the physical characteristics of each fuel assembly (burnup, reactivity, initial enrichment, etc.) and to automatically validate the final core loading. Such a device would be placed on the passage of the fuel assembly between the storage pond and the reactor building. It should be useful for core loading conformity control and on-line core monitoring.

The NAJA device is able to automatically determine (with non-destructive measurements) for each assembly:

- the nature of the fuel element (fresh or irradiated, UOX or MOX);
- the presence and the kind of neutron absorber;
- the initial enrichment in ^{235}U for fresh UOX assembly;
- the identification number.

This information allows to characterize the fuel assemblies accurately and to be sure, without human factor hazard, of the core loading conformity.

The NAJA device has been optimized in order to take into account severe constraints (no influence on the loading or unloading schedule, no need for human interface, no impact on operation). Such optimization has led to the following conclusions:

- without human interaction, the device controls each assembly which goes to the reactor building (loading) or which goes to the storage building (unloading);
- the device is located on the passage of fuel elements on the pond building near the transfer tube;
- three nuclear methods are applied to cover the whole fuel assembly panel (active neutron interrogation, passive neutron counting, gamma spectrometry);
- an ultra-sonic probe is used to monitor the different parts of the fuel element (foot, beginning of the fissile length);
- a video system linked to Optical Character Recognition (OCR) software, leads to an automatic reading of the fuel assembly number.

In fact, two video systems are planned to be used: the first one is linked to the NAJA device in the storage pool and the second one is linked to the loading machine in the reactor building. These two video systems are necessary in order to be sure of the good appropriateness between the position X, Y of the fuel assembly in the core and its physical characteristics (UOX or MOX fuel, burnup, initial enrichment in ^{235}U, kind of absorbent etc.). Successful tests of the OCR software have been performed even for "black" (e.g. corroded) fuel assembly numbers.

One of the big interests of the NAJA device consists of the combination between the nuclear measurements, the ultra-sonic probe and the video system which allows to associate each fuel element placed in the core to a fuel identification number and its physical characteristics without any information coming from the operator. The combination between the ultra-sonic probe and the nuclear device leads us to assert the reproducibility and the reliability of the fissile column measurement.

The feasibility study of the device has been made using experimental results from the PYTHON device and theoretical calculations for its optimization. The panel of the fuel assembly characteristics which have been taken into account is large and representative of the French fuel cycle:

- average burnup of the spent fuel from 6,000 MW·d/tU to 48,000 MW·d/tU;
- cooling-time varying from 1 to 90 days;
- initial enrichment in ^{235}U for UOX assembly varying between 3 and 4 %;
- nature of neutron absorbent: pins containing silver - indium - cadmium and/or pins containing silver - indium - cadmium and boron carbide (B_4C).

The study indicates the good performances which should be obtained with the NAJA device (for example +/- 2 % as a global uncertainty at 2 standard deviations on the absolute average burnup evaluation or +/- 1 % (in relative) as uncertainty at 2 standard deviations on the initial or residual enrichment evaluation) without any influence on the loading or unloading timing. Detailed results of this feasibility study are given in reference [5].

Potential uses of the NAJA device derive directly from its main functions:

1. The core conformity control which allows us to increase the safety level of the plant significantly;
2. The absolute and accurate burnup measurements of the irradiated fuel assemblies which allow us to improve the global availability of the power plant and to gain some investment benefits.

5. APPLICATION OF NDA TO STORAGE FACILITIES

The devices described below have been developed to provide NDA capabilities applied to fuel management in NPP ponds. Nevertheless, the same or similar NDA techniques could be successfully applied to fuel storage. For storage purposes the concerns are:

- establish an inventory of entering materials (direct disposal for example);
- control the consistency between the entering fuels and operators declarations (e.g. partial defect test, burnup, plutonium amounts);
- select the proper storage conditions regards to the residual reactivity of the fuels;
- prepare input data for fuel evolution during storage;
- long-term checking of the fuel integrity.

Storage application of existing NDA techniques have to be divided in two cases whether the storage would be dry or wet. The wet storage conditions are similar to reactor ponds, so, all existing NDA techniques (including active methods) can be successfully applied.

In dry conditions, all the passive measurements are easier to perform. Passive neutron counting as well as spectrometric gamma scanning would give accurate results if associated with a convenient evolution code. But, at the moment no active method have been yet applied to spent fuels in dry

conditions (Cf. SAGOR Report [8]). If full characterization is necessary (including initial enrichment determination) no active technique is already available. In addition spent fuel could be placed in containers (steel bottles) that make the measurement more difficult.

The major challenge for storage application is to develop an active NDA in air. For active NDA, isotopic neutron sources, e.g. ^{252}Cf, are used for under water measurements because of technological difficulties to operate a neutron generator under water. In dry conditions, such a generator delivering activities over 10^{10}n/s could be easily used.

For storage operating, the OCR technique could be used to associate characterization of the fuel and its management in the storage. Using such a capability, storage can be designed to take into account the burnup credit in order to save space and costs.

6. CONCLUSION

Many NDA techniques have been developed for spent fuels characterization and management. The most promising for storage application are the neutron methods combined with on line evolution codes. All existing NDA methods but active one can be successfully applied to storage.

For dry storage applications, active neutron measurements require further R&D to achieve accurate results. Characterization data given by NDA instruments can now be linked to automatic fuel recognition. Both information can be fed into the storage management software.

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