Quantitative burnup determination based on isotopic relationships

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Abstract

A method for the determination of the burnup of irradiated fuel rods is proposed on the basis of the gamma spectrometric determination of the relationship between the activities of two isotopes: ^{134}Cs and ^{137}Cs, is described. The procedure is applied to experimental data obtained on commercial PWR-fuel rods, irradiated up to 100 GWd/[HM]. The method can be reliable applied up to burnups up to 70 GWd/tHM but, for burnups beyond 30 GWd/tHM, depends on the accurate knowledge of the irradiation history of the fuel rods.

***Keywords:* Burnup determination, isotopic relationships, ^{134}Cs, ^{137}Cs**

Introduction

The burn-up of nuclear fuel defines the energy produced per mass of fuel and is related to the inventory of fission products formed in the matrix of the fuel and to changes in neutron-physical and material properties during irradiation. For this reason, all properties of the fuel are usually reported as a function of this important parameter. Therefore, it is essential to have methods available that allow a reliable determination of this important indicator of the amount of energy produced during the irradiation.

The traditional method to determine the burn up is by measuring the content of an isotope that results from the fission process. The isotope ^{148}Nd has proven to be an ideal monitor due to its chemical and neutron physical properties [1]. The Nd isotopic composition can be determined by mass spectrometry after dissolution of the sample. The mass spectrometer has to be couple with ion chromatography to circumvent mass interferences [2]. Another method for the burn-up determination is based on the experimental determination of the sum of ^{145}Nd and ^{146}Nd, and has been proposed to be used for the determination of the burn-up on the basis of SIMS-measurements, where mass interferences cannot be avoided [3]. In this case, at very high burn-ups of UO_2 fuel and, especially, MOX fuel this method needs weighted yields for U and Pu to obtain an acceptable accuracy. All these methods are rather costly and time consuming [4].

To elude the above mentioned drawbacks, non-destructive methods have been continuously researched in the past. Among the non-destructive spectrometric methods, the burn-up determination based on the measurement of ^{137}Cs gamma spectrometry, supplies adequate results provided the gamma radiation detector is properly calibrated, using a suitable standard, and the self-attenuation effects of Cs and the effect of the geometrical factors are catered for [4,5].

Gamma spectrometry offers another alternative, based on isotopic relationships, which is independent of the measuring conditions and detection time. Among the possibilities suggested by the gamma spectrometry of spent fuel, the relationship between ^{134}Cs and ^{137}Cs offers a particularly interesting option. In fact, ^{134}Cs is formed mainly by beta-decay through the isobar:[1]

$$^{133}\mathrm{I} \rightarrow {}^{133}\mathrm{Xe} \rightarrow {}^{133}\mathrm{Cs}\ \text{(stable)}$$

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and subsequent neutron capture to yield ^{134}Cs. It is also formed as a direct fission product but with a low probability. On the other hand, ^{137}Cs formation in the reactor is almost only determined by the fission of U and Pu and behaves linearly as a function of the burn-up. The use of the ratio of these two isotopes would, in principle, permit the determination of the burn-up, independently from the particular conditions of the gamma measurement, which would affect in the same manner the measurement of both isotopes.

In the present paper, the determination of the burnup of fuel rods irradiated in commercial reactors will be discussed to show the advantages and limitation of the proposed method. Experimental data, coming from a large data-bank of gamma spectrometry performed on fuel rods irradiated in Light Water reactors over the last two decades will be presented in relation to the burnup determination by the isotopic relationship method.

1. Theoretical background

As previously mentioned the build up and decay of the fission products ^{134}Cs and ^{137}Cs follow different mechanisms. ^{137}Cs is mainly produced through the fission of the isotopes ^{235}U, ^{239}Pu, ^{240}Pu and ^{241}Pu. Since ^{137}Cs has a long half life time (30.07 years) and its cross section for neutron capture (n,γ) is very low (~ 0.223 barn for Maxwell Average neutron spectrum) it behaves almost linearly as a function of the burn-up (BU). For long irradiation times the decay can be accounted for in form of a correction factor. Thereafter, the amount of ^{137}Cs, N_{137Cs}, as a function of the BU can be written as [5]:

$$N_{^{137}Cs}\left(t_{irr.}\right) = BU \cdot \frac{\gamma_{^{137}Cs} \cdot N_{HM}}{t_{irr.} \cdot \lambda_{^{137}Cs}} \cdot \left(1 - e^{\left(-\lambda_{^{137}Cs} \cdot t_{irr.}\right)}\right)$$

$\gamma_{^{137}Cs}$: fission yield , $\bar{\sigma}_f$: average fission cross section,

$\bar{\phi}_n$: neutron flux , $\lambda_{^{137}Cs}$: decay constant of ^{137}Cs

N_{HM} : heavy metal atoms , $N_{^{137}Cs}$: ^{137}Cs atoms

The time dependent part of this equation can be considered as a correction factor for long irradiation times and takes into account that the radioactive decay of ^{137}Cs is not dependent of the burn-up but of the time of irradiation. However, if the burn-up is simplified by considering it in a linear relationship to the irradiation time (constant power history) the time can be eliminated:

$$BU = \frac{\bar{\sigma}_f \cdot \bar{\phi}_n \cdot N_{HM} \cdot t_{irr.}}{N_{HM}} = \bar{\sigma}_f \cdot \bar{\phi}_n \cdot t_{irr.}$$

$$N_{^{137}Cs}\left(BU\right) = \frac{\gamma_{^{137}Cs} \cdot \bar{\sigma}_f \cdot \bar{\phi}_n \cdot N_{HM}}{\lambda_{^{137}Cs}} \cdot \left(1 - e^{\left(-BU \cdot \frac{\lambda_{^{137}Cs}}{\bar{\sigma}_f \cdot \bar{\phi}_n}\right)}\right) \qquad \textbf{Eq. 1}$$

On the other hand, the build up and decay of ^{134}Cs is not so straightforward to describe as the ^{137}Cs generation. In fact, this isotope is practically not produced by fission (the cumulative fission yield amounts to about 10^{-7}) but through neutron capture (n,γ) of ^{133}Cs which is stable and has a cumulative fission yield of about 6.7% (for Maxwell average). In addition, the half life time of ^{134}Cs is relatively short (~2.07 a) and the cross section for neutron capture relatively high (~123.6 barns for Maxwell average). Thus, the mathematical description for the build up and decay of ^{133}Cs in a nuclear reactor can be written as follows:

$$\frac{dN_{^{133}Cs}}{dt} = \gamma_{^{133}Cs} \cdot \bar{\sigma}_f \cdot \bar{\phi}_n \cdot N_{HM} - \bar{\sigma}_{C,^{133}Cs} \cdot \bar{\phi}_n \cdot N_{^{133}Cs}$$

$$IC: \quad N_{^{133}Cs}\left(t_{irr.} = 0\right) = 0$$

$$Sol.: \quad N_{^{133}Cs}\left(t_{irr.}\right) = \frac{\gamma_{^{133}Cs} \cdot \bar{\sigma}_f \cdot N_{HM}}{\bar{\sigma}_{C,^{133}Cs}} \cdot \left(1 - e^{\left(-\bar{\sigma}_{C,^{133}Cs} \cdot \bar{\phi}_n \cdot t_{irr.}\right)}\right)$$

$\bar{\sigma}_f$: average fission cross section , $\bar{\sigma}_{C,^{133}Cs}$: average capture cross section ^{133}Cs

$\bar{\phi}_n$: average neutron flux , N_{HM} : heavy metal atoms

With this solution a simplified equation for the build up of ^{134}Cs can be established.

$$\frac{dN_{^{134}Cs}}{dt} = \bar{\sigma}_{C,^{133}Cs} \cdot \bar{\phi}_n \cdot N_{^{133}Cs} - \lambda_{^{134}Cs} \cdot N_{^{134}Cs} - \bar{\sigma}_{C,^{134}Cs} \cdot \bar{\phi}_n \cdot N_{^{134}Cs}$$

$$IC: \quad N_{^{134}Cs}\left(t_{irr.} = 0\right) = 0$$

$$N_{^{134}Cs}\left(t_{irr.}\right) = \frac{\gamma_{^{133}Cs} \cdot \bar{\sigma}_f \cdot \bar{\phi}_n \cdot N_{HM}}{\left(\lambda_{^{134}Cs} + \bar{\sigma}_{C,^{134}Cs} \cdot \bar{\phi}_n\right) \cdot \left(\bar{\sigma}_{C,^{133}Cs} \cdot \bar{\phi}_n - \left(\lambda_{^{134}Cs} + \bar{\sigma}_{C,^{134}Cs} \cdot \bar{\phi}_n\right)\right)}$$
$$\cdot \left(\bar{\sigma}_{C,^{133}Cs} \cdot \bar{\phi}_n + \left(\lambda_{^{134}Cs} + \bar{\sigma}_{C,^{134}Cs} \cdot \bar{\phi}_n\right) \cdot \left(e^{\left(-\bar{\sigma}_{C,^{133}Cs} \cdot \bar{\phi}_n \cdot t_{irr.}\right)} - 1 \right) \right.$$
$$\left. -\bar{\sigma}_{C,^{133}Cs} \cdot \bar{\phi}_n \cdot e^{\left(-\left(\lambda_{^{134}Cs} + \bar{\sigma}_{C,^{134}Cs} \cdot \bar{\phi}_n\right) \cdot t_{irr.}\right)} \right)$$

$\lambda_{^{134}Cs}$: decay constant of ^{134}Cs

$\bar{\sigma}_{C,^{134}Cs}$: average cross section of ^{134}Cs for absorption

If, as before, the simplifying assumption is made that the amount of fissions per second is constant (constant power history) during irradiation, the burn-up can be written as linear function of neutron flux, cross section for fission and time of irradiation and the time can be eliminated, Hence:

$$N_{^{134}Cs}(BU) = \frac{\gamma_{^{133}Cs} \cdot \bar{\sigma}_f \cdot \bar{\phi}_n \cdot N_{HM}}{\left(\lambda_{^{134}Cs} + \bar{\sigma}_{C,^{134}Cs} \cdot \bar{\phi}_n\right) \cdot \left(\bar{\sigma}_{C,^{133}Cs} \cdot \bar{\phi}_n - \left(\lambda_{^{134}Cs} + \bar{\sigma}_{C,^{134}Cs} \cdot \bar{\phi}_n\right)\right)}$$

$$\cdot \left(\bar{\sigma}_{C,^{133}Cs} \cdot \bar{\phi}_n + \left(\lambda_{^{134}Cs} + \bar{\sigma}_{C,^{134}Cs} \cdot \bar{\phi}_n\right) \cdot \left(e^{\left(-\frac{\bar{\sigma}_{C,^{133}Cs} \cdot BU}{\bar{\sigma}_f}\right)} - 1 \right) \right.$$

$$\left. - \bar{\sigma}_{C,^{133}Cs} \cdot \bar{\phi}_n \cdot e^{\left(-\left(\lambda_{^{134}Cs} + \bar{\sigma}_{C,^{134}Cs} \cdot \bar{\phi}_n\right) \cdot \frac{BU}{\bar{\sigma}_f \cdot \bar{\phi}_n}\right)} \right)$$

Eq. 2

This solution is rather complicated and - more important - the different variables are strongly dependent on the neutron energy. Therefore, equation 2 is not ideal for the determination of the real inventory of ^{134}Cs. In addition unlike ^{137}Cs the isotope ^{134}Cs behaves not linear to the burn-up and is, in principle, not an ideal monitor for the burn-up.
Nevertheless, some simplifications can be achieved by dividing Equation 2 by Equation 1, establishing the desired ratio of the activities ^{134}Cs/^{137}Cs as a function of the burn-up:

$$\frac{N_{^{134}Cs}}{N_{^{137}Cs}} = f(BU) \qquad \Rightarrow \frac{A_{^{134}Cs}}{A_{^{137}Cs}} = \frac{\lambda_{^{134}Cs}}{\lambda_{^{137}Cs}} \cdot f(BU)$$

$$\frac{A_{^{134}Cs}}{A_{^{137}Cs}} = \frac{\lambda_{^{134}Cs} \cdot \gamma_{^{133}Cs}}{\gamma_{^{137}Cs}}$$

$$\cdot \frac{\left(\bar{\sigma}_{C,^{133}Cs} \cdot \bar{\phi}_n + \left(\lambda_{^{134}Cs} + \bar{\sigma}_{C,^{134}Cs} \cdot \bar{\phi}_n \right) \cdot \left(e^{\left(-\frac{\bar{\sigma}_{C,^{133}Cs} \cdot BU}{\bar{\sigma}_f} \right)} - 1 \right) \right)}{\left(\lambda_{^{134}Cs} + \bar{\sigma}_{C,^{134}Cs} \cdot \bar{\phi}_n \right) \cdot \left(\bar{\sigma}_{C,^{133}Cs} \cdot \bar{\phi}_n - \left(\lambda_{^{134}Cs} + \bar{\sigma}_{C,^{134}Cs} \cdot \bar{\phi}_n \right) \right)}$$

$$-\bar{\sigma}_{C,^{133}Cs} \cdot \bar{\phi}_n \cdot e^{\left(-\left(\lambda_{^{134}Cs} + \bar{\sigma}_{C,^{134}Cs} \cdot \bar{\phi}_n \right) \cdot \frac{BU}{\bar{\sigma}_f \cdot \bar{\phi}_n} \right)}$$

$$\cdot \left(1 - e^{\left(-BU \cdot \frac{\lambda_{^{137}Cs}}{\bar{\sigma}_f \cdot \bar{\phi}_n} \right)} \right)$$

Eq. 3

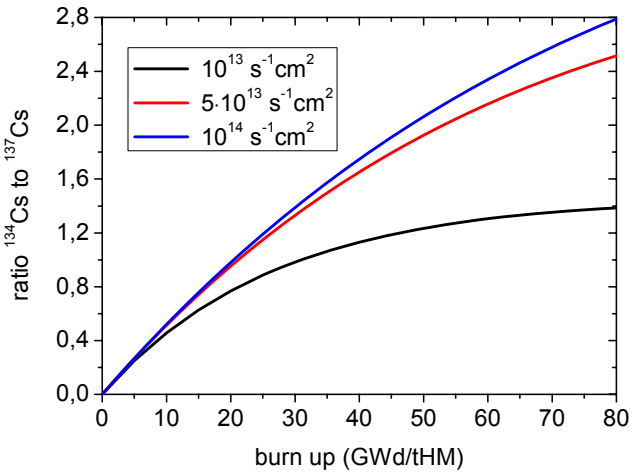

Figure 1: Ratio of ^{134}Cs to ^{137}Cs Activity for different Neutron Flux Densities

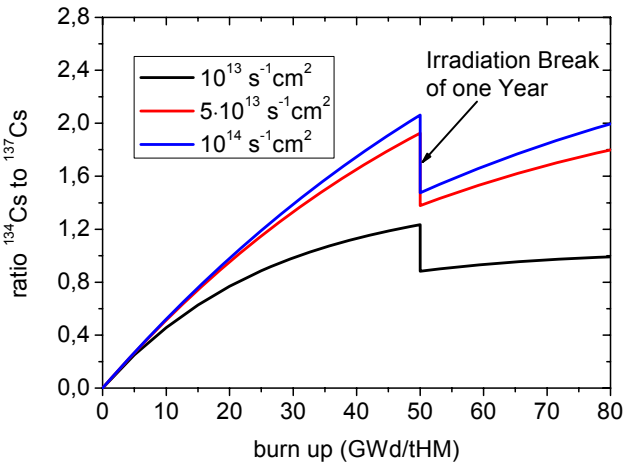

Figure 2: Ratio of ^{134}Cs to ^{137}Cs Activity with Irradiation Break

In Figure 1, the ratio of the activities ^{134}Cs/^{137}Cs calculated from Equation 3, is presented. The neutron flux density, which is inversely proportional to the irradiation time, was set as parameter.

Furthermore, the ratio will change with each shut down of the reactor or any interruption of the fuel rod irradiation, owing to the short half life time of ^{134}Cs. In Figure 2, the influence an irradiation break of one year after 50

GWd/tHM, is presented to illustrate the incidence of the short half-life of ^{134}Cs. This would suggest that, to determine the burn up using experimental data of the ratio between the two caesium isotopes, the knowledge about the exact irradiation history would be needed.

The previous mathematical exercise was undertaken to have some physical indication on the processes involved in the build up and decay of the two isotopes considered. An alternative and more detailed way to calculate the desired inventories for a given fuel rod, provided that irradiation history is known, is to use established inventory calculations codes like Origen® or Korigen®. These codes offer extended libraries of nuclear data, e.g. effective burn-up dependent cross sections, energy and isotope specific yields, reactor specific neutron spectra and radioactive decay data, hence, allowing more reliable calculations to be compared with experimentally measured values. Results of calculations using the Korigen®-code will be presented in § 3.

2. Experimental

During the last two decades, a large amount of fuel rods have been examined by gamma scanning in the hot cell installation of the Institute for Transuranium Elements, (ITU) covering a variety of LWRs. To perform the gamma spectrometry, a pure germanium detector was used in front of a collimator having a variable vertical window. The impulses detected by the Ge-detector are analysed by a computer programme to provide the activity of different nuclides. This analysis is performed in a discontinuous manner, through the integration of the counts during the necessary time to obtain spectrums which exhibit peaks with Gaussian distribution (typically, during 150 s covering fuel rod segments, 25 mm in length). A typical profile of ^{137}Cs, obtained using this procedure is shown in Fig. 3.

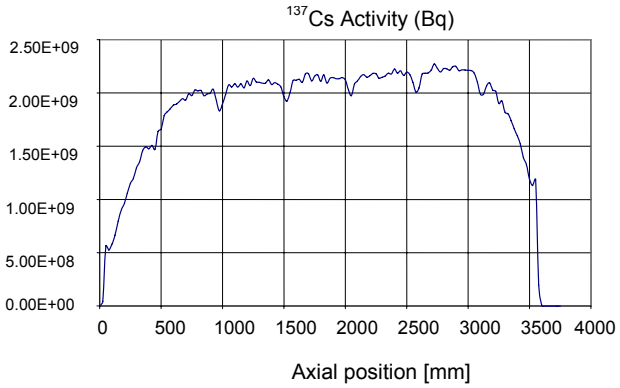

Figure 3: ^{137}Cs profile of a PWR fuel rod

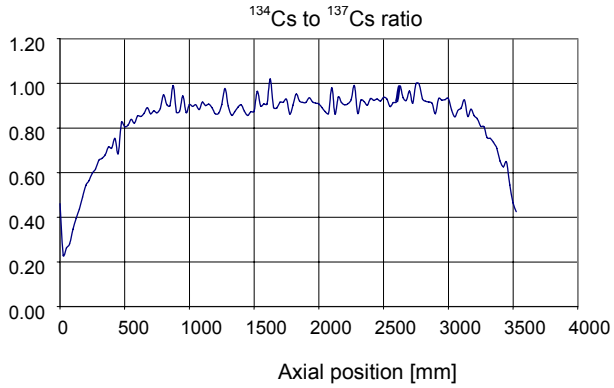

Figure 4: ^{134}Cs to ^{137}Cs ratio for a PWR fuel rod

As can be seen from Fig. 3, the ^{137}Cs profile has an increasing gradient from the outer portions of the fuel rod to the inner ones. The ^{137}Cs inventory is in linear relationship with the local burn-up and, thus, the profile reflects the local burn-up in the fuel rod. In Fig. 4 the ratio between the ^{134}Cs and ^{137}Cs activities over the fuel rod length is depicted. Evidently, also the activities ratio gives an indication of the local burn-up and follows, at least qualitatively, the same pattern illustrated in Fig. 3.

If the averaged burn-up (BU_A) of a fuel rod - ideally divided in "n" segments - is known, hence the measured ^{137}Cs profile can be used to determine the local burn-up (BU_L) of each segment "i" (notice that no quantitative statement is made concerning the burnup is made but only about the burnup distribution):

$$BU_L(i) = BU_A \cdot \frac{A_{^{137}Cs}(i) \cdot n}{\sum_{i=1}^{n} A_{^{137}Cs}(i)}$$

In Figure 5, the ratio of the $^{134}Cs/^{137}Cs$- activities as a function of the local burn-ups are shown for two PWR fuel rods. The first rod was irradiated up to 998 full power days (FPD) up to a burnup of 48.2 GWd/t[HM], and the second one for 2913 FPD up to a burnup of 97.7 GWd/t[HM]. The quantitative agreement will be discussed below in comparison with accurate code calculations.

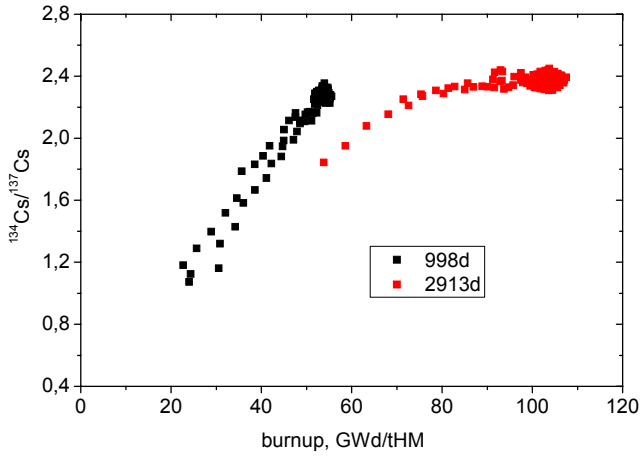

Figure 5: $^{134}Cs/^{137}Cs$ ratio vs burn-up for two PWR Fuel Rods

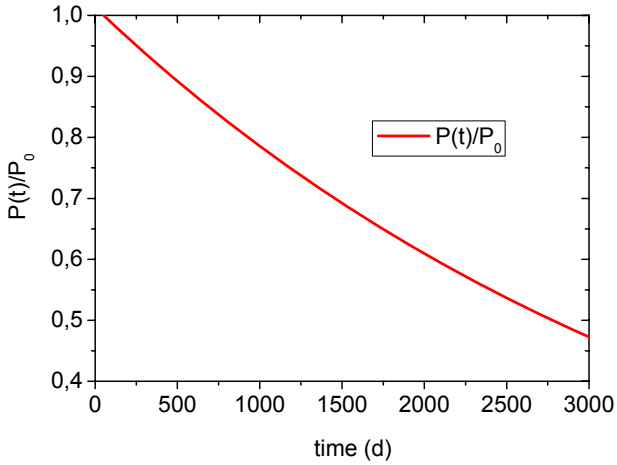

Figure 6: Postulated power history for burnup calculations

3. Code Calculations

The code calculations were performed using Korigen® (Version 2003), for fuel rods having an enrichment of 3.5%. The irradiation history was considered in 64 time steps of 51 days up to 3264 days, without irradiation breaks. A typical power history is presented in Figure 6 (P_0 is a generic starting value). This example reflects the real power history of a long time irradiated fuel rod up to a burnup of about 95 GWd/tHM. The used Libraries were "60PCU40 KFKBIB" for PWR (burn up dependent and averaged). The used libraries are only valid for low burnups (up to 60 GWd/tHM). In this sense, the present calculations have to be considered as an approximation. However, the error for the considered nuclides (^{134}Cs and ^{137}Cs) should be small.

Since the Korigen® output file delivers intermediate nuclide vectors for the programmed time steps it is possible to use several runs (with different power but same time steps) to produce “isochronous lines”. Along these time lines, the irradiation time is kept constant but the power history is different. In this way it is possible to display the expected range of burn-up and $^{134}Cs/^{137}Cs$ ratio of a particularly irradiated fuel rod. Fig. 7 shows such diagram for the case of PWR fuel.

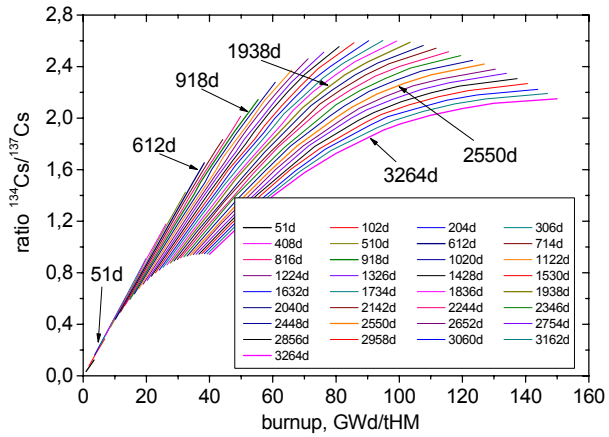

Figure 7: ^{134}Cs / ^{137}Cs ratio for different Korigen® runs (PWR)

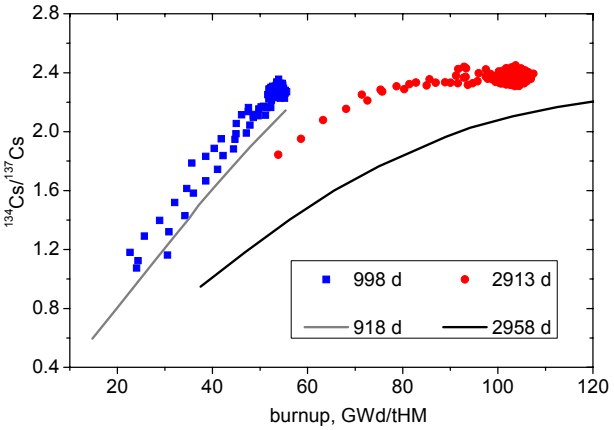

Figure 8: Comparison of code calculations with experimental results for PWR-fuel rods.

4. Results and discussion

In Fig. 8, the comparison between the experimental results reported in Fig. 5, are compared with code calculations, performed using the conditions described in § 3. It can be noticed that, for irradiation fuel rods up to three cycles (~ 1000 EFPD), the agreement is reasonable good whereas for longer irradiation times the code calculations provide a lower $^{134}Cs/^{137}Cs$ ratio as a function of burnup than the experimental values. In addition, it can be seen that, for low burnups, an almost linear relationship (having a steep slope) exists between burnup and the $^{134}Cs/^{137}Cs$ ratio. This makes the low burnup determination from this type of curves very well-defined since for a small variation in the burnup generates a large change in the isotope ratio. The diagram is, thus, very accurate for burnups up to 30 GWd/tHM. Between 30 GWd/tHM and 70 GWd/tHM a relatively precise knowledge of the time of irradiation becomes more important but, due to the still relatively significant gradient, it is still possible, in that range, to determine the burnup with fairly enough accuracy. Beyond this value both, in addition to the poor agreement between experimental and calculated values, the isotope ratio as a function of the burnup flattens and the gradient becomes too feeble to make a reliable statement about the burnup using this type of diagram.

Conclusions

A method based on the gamma spectrometric determination of the relationship of the activities of the isotopes ^{134}Cs and ^{137}Cs has been described. The method has been experimentally tested for PWR-fuel rods covering a burnup range from 20 to 110 GWd/tHM and the results compared with code calculations. The procedure can be reliable applied up to burnups of about 70 GWd/tHM. Beyond a burnup of 30 GWd/tHM, the knowledge of the irradiation history becomes more important. The method can be applied to test non-destructively the burnup of fuel rods irradiated in power reactors with sufficient accuracy provided the irradiation history is known.

References

1. ASTM-Standard Test Method, E 321-96, 1996
2. M. Betti, “Use of ion chromatography for the determination of fission products and actinides in nuclear applications”, Journal of Chromatography A 789 (1997) 369-379.
3. L. Degranges and B. Pasquet, «One Year Operation of the Shielded SIMS in the LECA Facility”, Proceedings of the Annual Meeting of the European Working Group “Hot Laboratories and Remote Handling”, Saclay, 2004
4. C. Devida, M. Betti, P.Peerani, W.Goll and E.H. Toscano, Proceedings of the Plenary Meeting of the European Hot Laboratories Research Capacities and Needs, 6th-8th September 2004, Halden, Norway.
5. D.Freis, R.Nasyrow and E.H.Toscano, Burnup Determination of High Temperature Reactors Spherical Fuel Elements by Gamma Spectrometry, HTR-2006, October 2006