

Evaluation of the irradiation-averaged fission yield for burnup determination in spent fuel assays

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Abstract. In order to derive the burnup of spent nuclear fuel from the concentration of selected fission products (typically the Nd isotopes and ¹³⁷Cs), their irradiation-averaged fission yields need to be known with sufficient accuracy, as they evolve with the changes in the actinide vector over the irradiation history. To obtain irradiation-averaged values, radiochemists often resort to robust generic methods – i.e., based on simple mathematical relations – that weight the fission yields according to the actinides contributing to fission, without performing core physics calculations. In order to assess the performance of those generic methods, a database of about 30 000 spent nuclear fuel inventories has been constructed from neutron transport and depletion simulations, covering a representative range of fuel enrichment, burnup, assembly designs and reactor types. When testing several existing methods for effective fission yield calculation, some inaccuracies were identified, originating from improper one-group cross-section parameters that do not accurately reflect resonance and self-shielding effects, and too crude approximations in the estimation of the actinide concentration evolution. Revised effective fission and absorption cross-section parameters are then proposed here, as a first improvement to the earlier burnup determination methods. As a second step, a novel method is proposed that reduces the error on their radiation-averaged fission yield values, and hence on burnup, while retaining a straightforward calculation scheme.

1 Introduction

Coupled neutron transport and depletion calculations are generally validated against well documented spent nuclear fuel (SNF) assay data. Those assays often report, in addition to the spent nuclear fuel inventory, the experimental sample burnup [1–12]. The experimental burnup offers a simple way to cross-check the burnup or the power history calculated by the plant operator. Two main methods for deriving the burnup from spent nuclear fuel assays can be distinguished [11]: a derivation based on the change in the heavy element isotopic ratio [11,13], and a derivation of the number of fissions that took place, as the product of the concentration of certain fission products in the spent nuclear fuel with their irradiation-averaged fission yields [11,14–19]. The burnup, expressed as fissions per initial heavy metal atoms (FIMA) is then given by:

$$BU = \frac{N_{BM}/\overline{Y}_{BM}}{N_{BM}/\overline{Y}_{BM} + \sum_{act.} N_{act.}} \quad (1)$$

with $\sum_{act.} N_{act.}$ the total concentration of actinides in the spent fuel, N_{BM} the concentration of a given burnup monitor isotope and \overline{Y}_{BM} its irradiation-averaged fission yield. One speaks of an irradiation-averaged fission yield because in the course of irradiation, the concentration of actinide isotopes, in particular the fissile ones, varies. Then, the distribution of fission events among those actinide isotopes evolves and, in turn, so does the distribution of fission products.

One makes here a further distinction between generic and robust methods that involve only simple mathematical operations, and methods that rely on lattice or core physics codes and the irradiation history to calculate the irradiation-averaged fission yield [10,20–27].

Several nuclides offer the possibility of calculating the burnup in a straightforward manner without recourse to lattice physics calculations. Such nuclides should ideally: (a) have almost identical yields for each of the dominant fissioning actinides; (b) not be present as impurities in fresh fuels and not be generated by activation; (c) have limited or no mobility themselves and not have mobile precursors; (d) have small neutron capture cross sections. These nuclides are called “burnup monitor nuclides”, or in short

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“burnup monitors”, and in the course of a radiochemical assay program they are used to evaluate the burnup of the sample being analysed. Such a burnup evaluation (sometimes named the “radiochemical burnup”) provides a burnup value that is independent from the one derived by the plant operator based on the reactor core power history and core physics data. In this work, we assess the robustness of earlier approaches to derive the sample burnup from the inventory of burnup monitor nuclides.

As part of the Ariane, Malibu, and REGAL Programs [28,29], several radiochemical assays have been performed at SCK CEN on very well characterized LWR fuel samples – in terms of design, fabrication, and irradiation data. While the average burnup value used in the quality-assurance procedure remained consistent with the burnup as evaluated by the plant operator, the burnup values associated with each individual burnup monitor showed significantly more scatter for the UO_2 fuel samples than what is typically observed for MOX fuel analyses. In order to shed light on this larger scatter of burnup values, it was decided to assess the procedures in place at SCK CEN and other laboratories.

Earlier burnup determination methods are first reviewed in Section 2. In Section 3, a database of simulated SNF inventories is constructed to assess those methods. It contains slightly less than 30 000 spent nuclear fuel inventories, obtained from depletion calculations; other relevant parameters were also retained for each simulated case, such as the number of fissions for each actinide, and the one-group, i.e., neutron spectrum-averaged, absorption and fission cross-sections (those will be further referred to as “effective” cross-sections). The simulated spent fuel inventories are used in Section 4 to assess the performance of earlier burnup determination methods, by comparing the burnup predictions from those methods with the burnup of the calculated cases. Strictly speaking, the comparison was conducted on the irradiation-averaged fission yield rather than the burnup, but those quantities largely scale with each other, as explained with equation (1).

Deficiencies and approximations in the cross-section parameters of earlier methods are then discussed in Section 5.1, and a novel set of cross-section parameters is then proposed for those methods. In an effort to further improve burnup determination from radiochemical assays, an improved methodology is finally proposed in Section 5.2. That method is first assessed and validated in Section 5.3 against the database of simulated cases developed in this work, and in a second stage, in Section 5.4, against a set of radiochemical assay results. It should be noted that the discussion conducted here is not restricted to radiochemical (dissolution-based) assays, but could be applicable to other techniques providing isotopic information on the fuel sample.

2 Principles of burnup determination

2.1 Burnup derived from heavy element isotopic ratios

Conceptually, the number of fissions taking place in a fuel sample is easily derived from the fresh and spent

fuel compositions. For fresh fuel, the composition is available from the fabrication data files. For irradiated fuel, the inventory of the dominant actinide isotopes (U, Pu) and part of the minor actinides (Am, Cm) needs to be analysed.

One early procedure to derive burnup from changes in heavy element isotopic is discussed in [11]. It was formalized in an ASTM standard (ASTM E244 [13]) but the standard was withdrawn as a full burnup determination method in 2001; yet part of it still remains applied through ASTM E321 [30], as discussed in Section 2.2.2. In this method, the burnup is derived solely from the heavy metal inventories of the fresh and irradiated sample. It also provides a method for estimating the contributions to fissions from ^{235}U , ^{239}Pu , ^{241}Pu and ^{238}U . The expressions involve only isotopic ratios and the initial inventory of ^{238}U . The method necessitates the evaluation of several isotopic ratios before and after irradiation, and accurate mass spectrometric measurements are needed for ^{235}U , ^{236}U , ^{238}U , ^{239}Pu , ^{240}Pu , ^{241}Pu and ^{242}Pu . The method is quite complex as it attempts to model the burnup of ^{235}U , ^{239}Pu and ^{241}Pu , while accounting for the capture fraction in the ^{238}U activation chain. It might be overly sensitive to the precision with which the inventory can be measured, in particular for ^{242}Pu .

- Regarding the fission contribution from ^{235}U , three approaches have been proposed [11]. The first one uses the difference in concentration of ^{235}U between fresh and irradiated fuel, corrected for the measured ^{236}U build-up. A second approach uses only the measured ^{235}U depletion and evaluates the ^{236}U build-up from the ^{235}U depletion and the ^{235}U capture-to-fission ratio. In the third approach, bearing in mind that the accuracy with which ^{236}U can be measured is better than the accuracy with which the decrease in ^{235}U concentration can be determined, measurement of the build-up of ^{236}U is preferred, with the ^{235}U fissions being derived from the capture-to-fission ratio.
- The number of fissions due to ^{239}Pu and ^{241}Pu are evaluated from the build-up of heavier isotopes through activation: the concentrations of $^{240,241,242}\text{Pu}$ and ^{241}Am account for the activation of ^{239}Pu , and the concentration of ^{242}Pu for activation of ^{241}Pu . Activations beyond ^{242}Pu are neglected. Again, the capture-to-fission ratios are used to determine the various fission contributions.
- Finally, the number of fissions due to ^{238}U is calculated from the reactor-specific fast fission factor, which is derived from core physics calculations.

A comprehensive description of the method is available in [11]. ASTM E244 is no longer used for absolute burnup analysis (as mentioned above the standard was withdrawn in 2001 [13]), but the method is still used in the standard ASTM E321 [30] to determine the irradiation-averaged fission yield. Test cases with simulated PWR UO_2 and MOX fuels have confirmed the inadequacy of the method for absolute burnup determination; the ASTM E321 variant of the method, is, however, further assessed in this work.

2.2 Burnup derived from monitor fission products

2.2.1 Concepts and practical limitations with “burnup monitor” fission products

The difference in actinide content between fresh and irradiated fuel would readily provide the burnup, expressed as a fraction of the initial inventory. However, as seen above, the measurement uncertainty is too high for such a method to be useable. Another approach is to consider the build-up of fission products as irradiation proceeds. This build-up is far from linear for most fission products because their concentrations evolve through a series of decays and may be affected by neutron capture. As the neutron-to-proton ratio in the fission fragments reflects that of the fissioning nucleus, the fission fragments typically feature an excess of neutrons and will decay rapidly to nuclei that are closer to the “stability valley” by β^- decay, followed by internal transition (emission of γ -ray) when the daughter nucleus is in an excited state. Consequently, decay series that remain isobaric¹ (i.e., for which the mass number does not vary) are usually observed. The individual nuclides have an independent (i.e., direct production) and cumulative fission yield. Given that most fission fragments produced in a specific isobaric chain have noticeably short half-lives, it is also useful to define the “chain fission yield” that applies to the entire isobaric decay chain. Furthermore, some of the nuclides in the decay chain have large neutron capture cross-sections; neutron capture events then interfere with these decay chains and redirect part of the decay series of atomic mass A to that of mass $A + 1$. The significance of this redirection depends on the relative importance of the reaction rate ($\sigma_c\phi$) compared to the decay constant (λ).

From the above considerations, it appears that only stable or long-lived nuclides, at the end of a decay series, and with small neutron capture cross sections will evolve in close-to-linear proportions with the burnup and hence could serve as useful burnup monitors. If a single nuclide in the decay chain presents an important neutron capture cross-section, one might combine the cumulative fission yield of two decay series to cancel out the branching effect. This approach is applied to the $^{143+144}\text{Nd}$ and $^{145+146}\text{Nd}$ pairs, given the non-negligible capture cross sections of both ^{143}Nd and ^{145}Nd . Similarly, in the case of medium-lived nuclides (half-lives comparable to the cooling time between the end of irradiation and the date of analysis), a nuclide and its daughter nuclide may be considered together. This is the case, for example, for ^{144}Ce (half-life: 285 d) which has often not fully decayed and needs to be accounted for in the inventory of the 144-mass series.

In addition to the above aspects, the dependence of the fission yield of each mass series on the nuclide initially undergoing fission should be considered. The irradiation-averaged fission yield changes as irradiation proceeds, as a result of the progressive change in the fuel composition due to consumption of ^{235}U and build-up of plutonium in UO_2 fuel, for example. Burnup monitors are best selected at a mass that shows limited dependence of the fission yield

on the actinide contributions to fissions. In this way, any uncertainty on the fissile actinide inventory results only in slight changes in the irradiation-averaged fission yield. The most suitable candidate nuclides have an atomic mass close to 100, or in the range 130–150. A more detailed selection of fission yield data [34], where one restricts the difference in fission yield to 5% for the two dominant contributors ^{235}U and ^{239}Pu (thermal spectrum fissions), and to 20% when including ^{241}Pu (thermal spectrum fissions) and ^{238}U (fission spectrum fissions), suggests that the 98, 99, 133, 134, 137, and 148 mass series are the best candidates (see Tab. 1).

The physical behaviour of the elements during irradiation and possible difficulties for their subsequent (radio-)chemical analyses further define the suitability (or not) of certain fission products as burnup monitors. When local burnup (pellet level or lower) determination is desired, macroscopic migration of the burnup monitor – or one of its predecessors – is to be avoided; this leads to fission gases (Kr, Xe) being rejected as monitors, and to treating volatile species with care. For example, in a fuel rod that is operated at elevated temperatures, a fraction of the Cs inventory migrates and precipitates at pellet–pellet interfaces. Other elements tend to form precipitates within the fuel matrix and may prove difficult to dissolve. It is then difficult to chemically recover their entire inventory; this is the case for the metallic elements such as Mo, Tc, Ru, Rh, Ag & Pd. For all these reasons, the choice of burnup monitor is often limited to lanthanides, notably ^{144}Ce and the neodymium isotopes [1,2,12,35]. Of the neodymium isotopes, ^{148}Nd is often used as reference nuclide for burnup analysis, in view of the small difference of fission yield between the two dominant fissile isotopes ^{235}U and ^{239}Pu . Still, there are good experimental reasons to evaluate the entire Nd-vector; for example, the $^{147}\text{Nd}(n,\gamma)^{148}\text{Nd}$ capture reaction is generally negligible for power reactor fuels, but may become significant for high-flux reactors. In order to use ^{148}Nd as a fission product monitor for such high flux reactor fuels, a correction factor needs to be calculated taking into account the neutron flux density in the fuel. Without this correction factor, using the ^{148}Nd concentration leads to an overestimation of the burnup. Data from the other Nd isotopes can help to identify when such situations arise, and can provide estimates of the burnup value if it is not possible to correct the ^{148}Nd for such capture by ^{147}Nd . In radiochemical assays, the preferred method for accurate and precise quantification of the inventory for many nuclides is isotope dilution mass spectrometry (IDMS) after chemical separation to avoid isobaric interferences. The measurement principle of IDMS is conceptually simple. It is based on two isotope ratio measurements of an analyte: one measurement on an as-separated aliquot, one on an aliquot to which a known amount of a spike is added. This spike contains the same element at a known concentration, but with a different, yet known, isotopic composition. After mixing the analyte with the spike, a blend is obtained with a different isotopic ratio to that of the as-separated aliquot. From the two isotopic ratio measurements and the masses of analyte and spike, the analyte concentration is derived with high accuracy and precision. Trivially, not

¹ A few fission products feature other decay mechanisms, such as neutron emission; in this case, a branching to the $A - 1$ mass series occurs.

only elemental concentrations are obtained, but also the entire nuclide vector of the analyte is quantified and can be exploited. Any discrepancies in the burnup derived from individual Nd isotopes (or isotope pairs when activation is to be accounted for) would merit further investigation since these could well be indicative of experimental problems e.g., mass discrimination or mass fractionation problems, isobaric interferences due to incomplete elemental separation.

Since it has a long half-life and a hard gamma emission line, ^{137}Cs is also used as burnup monitor, principally for non-destructive measurements. ^{137}Cs may also be considered as burnup monitor in spent fuel assays, providing that the inventory that possibly escapes from the fuel matrix (and generally precipitates at pellet–pellet interfaces) is properly collected. The burnup derivation from ^{137}Cs for leaking fuel rods is therefore excluded. Using γ -spectrometry in spent fuel assays, a completely different experimental route is followed. Again, any discrepancies between the burnup values derived from ^{137}Cs and from the Nd vector could indicate a problem on the measurement side (e.g., issue with the spiking for the Nd-vector measurement), which would then need further investigation. On the other hand, on well-measured samples, useful information may be derived from the Nd isotopic ratios in relation to the origin of fissions among the different fissile actinides.

2.2.2 Methods based on “burnup monitor” fission products

The procedure to derive burnup from the concentration of selected fission products has been formalised in ASTM E321 for burnup determination based on the ^{148}Nd concentration. A sufficiently accurate estimation of the irradiation-averaged fission yield for the selected burnup monitor fission products is necessary. Within ASTM E321, the ASTM E244 method is used to derive the relative contributions to fission of actinides and to weight the fission yield (see Sect. 2.1).

The ASTM E321 approach may be easily extended to other burnup monitors and/or to other methods to evaluate the irradiation-averaged fission yield. As stated earlier, a distinction can be made between robust methods that involve simple mathematical operations, and methods that rely on lattice or core physics codes. Methods based on a case-specific code evaluation to derive the irradiation-averaged fission yields have the advantage of providing accurate estimations of the absolute and relative fission contributions of the principal (fissile) actinides [10,20–27]. However, the independent nature of the derived burnup value can be questioned if it is later used to validate the same code. A further concern is the need to perform a simulation for each and every sample to be analysed, while a complete characterization of the fresh fuel and its irradiation history data are often not available. Therefore, such approaches will not be discussed further in this work.

A constant value for the irradiation-averaged fission yield is sometimes considered [1,23,36,37], but this generally results in a crude approximation, even for isotopes where the irradiation-averaged fission yield changes little

with the burnup. The ASTM E321 method for burnup determination from ^{148}Nd (cf. Sect. 2.1), then provides a more accurate procedure to calculate the irradiation-averaged fission yield. Other methods for evaluating that quantity have been proposed, in which the fission rates of the key actinides (^{235}U , ^{239}Pu , ^{241}Pu , and ^{238}U) are considered to be proportional to both their irradiation-average concentration and their effective (i.e., one-group, neutron spectrum-averaged) fission cross-section [14–18]. The irradiation-average distribution of fissions is then approximated by a weighted average of the beginning and end-of-life (BOL and EOL) repartitions, which therefore requires both fresh and spent fuel compositions:

$$\bar{Y}_i = \frac{1}{1+p} \left(\left[\frac{\sum_j N_j \sigma_{f,j} Y_j^i}{\sum_j N_j \sigma_{f,j}} \right]_{\text{BOL}} + p \left[\frac{\sum_j N_j \sigma_{f,j} Y_j^i}{\sum_j N_j \sigma_{f,j}} \right]_{\text{EOL}} \right) \quad (2)$$

with \bar{Y}_i the irradiation-averaged fission yield of mass series i ; N_j the molar concentration of the fissioning nuclide j ; $\sigma_{f,j}$ the one group-averaged fission cross-section of nuclide j , Y_j^i the chain fission yield of mass series i for the fissioning isotope j , and p is a weighting parameter. In the papers by de Regge [14] and Boden [17], the contribution from ^{238}U (fast fission) was neglected, such that only thermal fissions from ^{235}U , ^{239}Pu and ^{241}Pu were considered. This approach is currently applied at SCK CEN to most radiochemical assays of samples originating from thermal spectrum reactors. If the weighting factor p is taken as unity, the same weight is given to BOL and EOL contributions. The effective fission cross-sections were approximated by their thermal spectrum-averaged microscopic cross-sections for LWR fuel analyses [17]. This last approximation implicitly considers infinite-dilution and neglects resonance and shielding aspects affecting neutron transport. It may seem a crude approximation of the actual irradiation conditions, but it should be noted that only the ratios of cross-sections will play a role in equation (2). The use of microscopic cross-sections in the approach will be evaluated and discussed in Section 4.

The expression in equation (2) assumes a quasi-linear evolution of the relative fission distribution over the entire irradiation. This approximation is known to perform well for MOX fuels [17,21] in which the isotopic inventory of fissile isotopes evolves little, as well as for low burnup UO_2 fuels where the linear evolution in the concentration of the major fissile isotopes is reasonably well respected. However, this approximation fails in the case of UO_2 fuels with moderate to high burnup, for which a significant scatter among the different burnup monitors is observed. For the Nd-isotopes and for ^{137}Cs , the chain fission yields exhibit limited variation between the different fissile isotopes. It can then be demonstrated (cf. Appendix A) that the irradiation-averaged fission yield of ^{137}Cs or ^{148}Nd can be calculated accurately (deviation less than 2%) even if the fission contributions of ^{238}U , ^{239}Pu , and ^{241}Pu are only roughly known (with tolerances as large as 40%), where as for $^{145-146}\text{Nd}$ and $^{143-144,150}\text{Nd}$ these fission contributions must be estimated more precisely (10% and 5%, respectively).

Two variants of the above method have been reported by de Regge in 1977 and 1989 [14,16]. In his 1977

Table 1. Top part: chain fission yield (per 100 fissions) for mass series that show less than 7.5% difference between ^{235}U and ^{239}Pu (thermal spectrum fissions), and less than 30% when including ^{241}Pu (thermal spectrum fissions) and ^{238}U (fast spectrum fissions). Bottom part: same values for the other Nd isotopes used as burnup indicators at SCK CEN. Source of numerical data: [34].

Atomic mass series	^{235}U thermal spectrum	^{239}Pu thermal spectrum	^{241}Pu thermal spectrum	^{238}U fission spectrum	Difference ^{235}U - ^{239}Pu	Max difference w.r.t. ^{235}U	Comment
98	5.7903	5.830892	5.035685	5.944993	1%	16%	(a)
99	6.109164	6.211806	5.961885	6.168192	2%	4%	(a)
133	6.699915	7.016466	6.72919	6.762707	5%	5%	(b)
134	7.872644	7.676881	7.917365	7.609406	3%	4%	(b), (c)
137	6.188944	6.613842	6.650911	6.052506	7%	9%	(d)
148	1.673660	1.642120	1.932103	2.112485	2%	26%	
143	5.956198	4.413261	4.577993	4.622060	30%	32%	
144	5.499964	3.739785	4.227384	4.547939	38%	39%	
143+144	11.45616	8.153046	8.805377	9.169999	34%	35%	
145	3.933647	2.986310	3.262911	3.808983	27%	27%	
146	2.997113	2.458146	2.766456	3.445628	20%	34%	
145+146	6.93076	5.444456	6.029367	7.254611	24%	28%	
150	0.653325	0.967459	1.209439	1.273456	39%	60%	(e)

(a) ^{98}Mo or ^{99}Tc are difficult to dissolve; (b) the monoisotopic natural abundance of ^{133}Cs means that suitable Cs isotopic reference materials are hardly available, without which the calibrated isotopic ratio measurements (having the low uncertainties characteristic of IDMS) are not possible determine with a low uncertainty, it has also a non-negligible cross-section and forms ^{134}Cs upon neutron capture; (c) the decay chain along the mass series 134 is blocked by ^{134}Xe that escapes upon sample preparation and dissolution; (d) ^{137}Cs migration and in-pile decay should be properly accounted for; (e) mass 150 has a low fission yield.

publication, de Regge addressed, in addition to the method described above, a variant for fuels that feature both thermal and fast fissions. In this approach, the irradiation-averaged fission yield is evaluated first for thermal fissions and then separately for fast fissions, based on equation (2). The differences in ^{150}Nd and ^{148}Nd fission yield ratios in thermal and fast spectrum (cf. Tab. 1) are then exploited to derive the fraction of fast fissions, f :

$$\frac{N_{148}\text{Nd}}{N_{150}\text{Nd}} = \frac{(1-f)\overline{Y}_{\text{th}}^{148} + f\overline{Y}_{\text{fast}}^{148}}{(1-f)\overline{Y}_{\text{th}}^{150} + f\overline{Y}_{\text{fast}}^{150}} \quad (3)$$

The method offers an elegant way of deriving the fast fission contribution for fuels where the irradiation conditions are poorly known. The accuracy with which factor f is determined depends highly on the precision with which the $^{148}\text{Nd}/^{150}\text{Nd}$ ratio can be measured. Complications may also arise when ^{241}Pu makes a non-negligible contribution, since the $^{148}\text{Nd}/^{150}\text{Nd}$ ratios in fast fission of ^{238}U and thermal fission of ^{241}Pu are similar. In 1989, de Regge proposed a second variant for fast spectrum MOX fuel to better estimate the contributions from ^{239}Pu and ^{241}Pu at high burnup: one first derives the ratio of ^{239}Pu and ^{241}Pu relative contributions to fissions; the ratio of $^{148}\text{Nd}/^{150}\text{Nd}$ is then exploited to derive the relative contribution from ^{235}U , $F_{235\text{U}}$, while fissions originating from ^{238}U are neglected:

$$\left. \begin{aligned} \frac{F_{241\text{Pu}}}{F_{239\text{Pu}}} &= \frac{1}{1+p} \cdot \frac{\sigma_f, 241\text{Pu}}{\sigma_f, 239\text{Pu}} \cdot \left(\left[\frac{N_{241\text{Pu}}}{N_{239\text{Pu}}} \right]_{\text{BOL}} + p \left[\frac{N_{241\text{Pu}}}{N_{239\text{Pu}}} \right]_{\text{EOL}} \right) \\ \frac{N_{148}\text{Nd}}{N_{150}\text{Nd}} &= \frac{Y_{148}}{Y_{150}} \end{aligned} \right\} \quad (4)$$

The system is closed with equation (2) to evaluate the irradiation-averaged fission yield. The applicability of the

method to UO_2 fuel, where the ratio of ^{241}Pu to ^{239}Pu inventory at BOL is undetermined, requires an educated guess of that initial ratio. The methods proposed by De Regge [14,16] addressed primarily the needs of MOX and fast reactor fuels, while those of Boden [17] were intended for LWR MOX and UOX fuels. In order to distinguish between the three variants in this work, the methods based on equations (2)–(4) will be referred to as Boden 1992 [17], de Regge 1977 [14] and de Regge 1989 [16], respectively.

3 Construction of the assessment database

Computer simulations to predict the evolution of fuel composition during irradiation were performed for diverse fuel and assembly designs. Other parameters important for assessing the burnup determination method are also evaluated, such as the effective fission and absorption cross-sections, and the number of fissions due to each actinide isotope. Calculations were performed with the SCALE 6.2 suite for nuclear safety and design [38], developed by ORNL. Of particular interest were the depletion capabilities of the code, which are centred on the ORIGEN module, an activation/depletion/decay solver. The ORIGEN module tracks the nuclide inventory evolution through a transition matrix between all pairs of nuclides, whose coefficients are derived from a set of one-group cross-sections (re-evaluated at the different burnup steps) and nuclear decay data. The ORIGEN module can be used in the framework of coupled neutron transport/depletion sequences implemented in SCALE (e.g., Triton or Polaris sequences), but can also be used as a stand-alone module. It is then fed with an external source of effective cross-sections – or with a neutron flux spectrum via the

COUPLE module, which then generates the transition matrix for activation/depletion/decay in a format readable by ORIGEN. The SCALE suite also offers the possibility to use pre-built libraries, generated with the Triton sequence, and designed to facilitate depletion calculations for diverse assembly designs, by interpolation of major fabrication and irradiation parameters. This is achieved via the ARP module of SCALE.

The pre-built ARP libraries available in SCALE were constructed from simulations with the latest code version, using the Triton module and the ENDF/B-VII.1-based 252-group cross section libraries. These pre-built libraries cover diverse types of reactors and assembly designs, including MOX fuel assemblies:

- BWR UO₂ & MOX: 22 assembly designs, from 7 × 7 to 10 × 10 lattices.
- PWR UO₂ & MOX: 18 assembly designs, from 14 × 14 to 18 × 18 lattices.
- VVER: 5 assembly designs.
- Candu: 3 assembly designs.
- RBMK: 1 assembly design.
- Magnox: 1 assembly design.
- AGR: 1 assembly design.
- IRT (MTR plate fuel): 2 assembly designs.

For each of the assembly designs, the ARP module offers interpolation capabilities with respect to the initial enrichment – for MOX fuel, this also includes the isotopic vector –, void fraction (only for BWR assemblies) and burnup, totalling 1470 *enrichment* | *void fraction* pairs and 28227 *enrichment* | *void fraction* | *burnup* combinations.

The pre-built ARP libraries within SCALE were utilised to evaluate the nuclide inventory, and the relative contributions to fission were derived from ORIGEN calculations. In addition, effective cross-section values were extracted, for each burnup step, from these libraries through the XSECLIST utility program, available within SCALE. The composition, fission and cross-section data obtained for the 28227 *enrichment* | *void fraction* | *burnup* combinations considered in this study will be further referred to as the assessment database.

4 Evaluation of burnup methods with the assessment database

Burnup determination from radiochemical analyses requires evaluation of the relative fission fraction of all four major fissioning isotopes: ²³⁵U, ²³⁸U, ²³⁹Pu and ²⁴¹Pu. The 1470 pre-built ARP libraries were evaluated, as a function of burnup, for:

- the effective (one-group) fission and absorption cross-sections,
- the concentration of the actinides,
- the relative fission contribution of the principal (fissile) actinides.

The data are interpreted later (see Sect. 5.1) to derive generic correlations, which can then be implemented in radiochemical assay procedures to determine sample burnup.

The effective fission and absorption cross-sections were extracted at different burnups from the pre-built ARP libraries using the XSECLIST utility program. These cross-sections are typically normalised to the thermal flux rather than total flux [38]. The calculated effective cross-sections differ considerably from microscopic cross-sections – i.e., in the infinite dilution limit – reported in nuclear data tables, due to rod self-shielding and lattice shielding effects. This indicates a need to use advanced modelling in burnup determination procedures. In addition, the ratio of these cross-sections is not invariant: e.g., the ratio of ²³⁹Pu to ²⁴¹Pu effective fission cross-section varies between 0.6 and 1.1 (see Fig. 1, left), with a majority of cases in the 0.85–1.0 range. In comparison, the ratio of microscopic cross-sections of ²³⁹Pu and ²⁴¹Pu in the thermal energy range is close to 0.74 [39]. The difference is even more pronounced for the ratios of ²³⁹Pu or ²⁴¹Pu to ²³⁵U (as illustrated in Fig. 1, right, for the ²³⁹Pu – ²³⁵U pair) where the ratios of the effective fission cross-sections significantly deviate from the ratio of microscopic fission cross-sections, which amount to 1.28 and 1.74 for ²³⁹Pu-to-²³⁵U and ²⁴¹Pu-to-²³⁵U, respectively. This suggests that irradiation-averaged fission yield calculations cannot be derived reliably from microscopic fission cross sections because of resonance and self-shielding effects.

The effective cross-sections, the isotopic inventory, and the relative fission contribution of the principal (fissile) actinides were evaluated for the complete set of ARP *enrichment* | *void fraction* | *burnup* combinations. Applying the four methods presented in Section 2.2.2 (ASTM E321, de Regge 1977, de Regge 1989, Boden 1992) to that assessment database, it becomes possible to investigate their relative performances in detail. Of interest is the ability of each method to calculate, in a robust manner, the *relative* contributions to fissions and the irradiation-averaged fission yields of the different burnup monitors. The direct derivation of burnup from the actinide inventory alone, in ASTM E244, has not been retained in view of the deficiencies identified in the method. As an integral part of the assessment of all these methods, the irradiation-averaged fission yield of several burnup monitors, evaluated from the calculated actinide BOL and EOL inventory, was compared to the “true” irradiation-averaged fission yield, derived directly from the distribution of fissions generated by the ORIGEN simulation. Three situations were considered, each corresponding to a column in Figure 2, namely the application of the different methods to the BOL and EOL inventories of:

- the assessment database, used as a set of very precise, yet fictive, assays from which to derive the burnup, while using the originally proposed set of cross-sections in the different burnup determination methods.
- a perturbed database and the original set of cross-sections for the burnup determination methods. Here the experimental scatter of real assays is simulated through data perturbation, assuming a normal distribution, centred on the calculated results and with 1% standard deviation (relative). The objective is to assess the sensitivity of the methods to data perturbation.

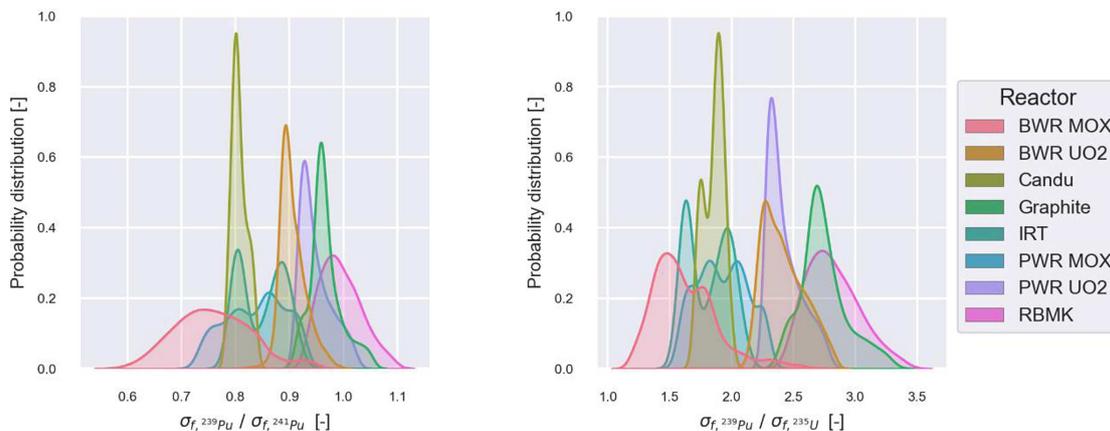


Fig. 1. Distribution of the effective fission cross-section ratio of (left) ^{239}Pu to ^{241}Pu , and (right) ^{235}U to ^{239}Pu for all cases of the assessment database. A distribution plot according to the reactor type was deemed sufficient, in view of a limited dependence of those ratios with assembly design, enrichment, water density or burnup.

- the assessment database, while using the effective (one group) cross-section set derived from ORIGEN for each individual case (referred to as “case-specific” cross-sections), rather than the parameter set of the original methods.

Several observations can be made from Figure 2. Firstly, the more reliable performance of mass series 148, compared to mass series 146, as burnup monitors is evident; given that for all methods a limited spread (scatter of results) and a limited bias (systematic deviation), of less than 2.5%, is observed. The performance of the methods was also assessed for other mass series, which are not illustrated here for the sake of conciseness. While mass series 137 performs equivalently well as mass series 148, the other mass series considered (143–145 and 150) show significantly larger biases and spread, similarly to those observed for mass series 146. The ASTM E321 method seems to perform slightly better than Boden 1992 and de Regge 1989 methods for mass series 143–146 and 150, probably because it also accounts for the contribution to fissions originating from ^{238}U . A further observation is that MOX cases tend to perform better with the Boden 1992 and de Regge 1989 methods, which is most probably related to the large errors in the irradiation-averaged fission yield calculations of the ASTM E321 method (inherited from ASTM E244), as discussed in Section 2.1.

The perturbation of the assessment database, depicted in the second column, has a limited effect on the performance of the methods. The last column, where case-specific effective cross-sections are used, shows the smallest spread and bias. When using case-specific effective cross-sections, only the intrinsic limitations of the methods themselves remain as source of the spread. It should be noted that the case-specific cross-section parameter set includes a fission cross-section value for ^{238}U ; those fissions are then accounted for in the Boden 1992 [17] and de Regge 1989 [16] methods. In the next section, we describe the derivation of a generic set of cross-sections to improve the predictions of the existing methods.

5 Improvements to generic burnup calculation methods

5.1 Improvement of effective cross-section sets for existing methods

The performance of burnup evaluation methods that use selected burnup monitors depends critically on the derivation of the irradiation-averaged fission yield of the selected burnup monitors. In turn, that derivation requires a good estimation of the lifetime average fissile isotope concentration, the effective fission cross section of each of the fissile isotopes, as well as proper nuclear data for fission yield (their accuracy is not addressed in this work).

The superior performance of the various burnup determination methods when case-specific effective cross-sections are used prompts us to propose a generic set of parameters that would perform better than the sets originally proposed in each of the existing methods (e.g., the microscopic thermal fission cross sections for the Boden 1992 method). Relationships between effective cross-sections were therefore evaluated. Those suggest an important level of correlation for actinide effective cross-sections, as already observed for example with the limited variations of the $\sigma_{f,^{239}\text{Pu}}/\sigma_{f,^{235}\text{U}}$ cross-section ratio (illustrated in Fig. 1). These relationships may be further exploited, considering that, ultimately, only the ratios of effective fission cross-sections matter in the irradiation-averaged fission yield evaluation, and not their absolute values. For the methods based on burnup monitors, only effective fission cross-sections are involved. Figure 3 also shows that the contribution from ^{238}U fissions cannot be neglected in most cases, and hence is included in the derivation scheme. The proposed set of parameters is given in the first part of Table 2.

The heavy element isotopic ratios method [11], still in use in ASTM E321 [30] involves additional effective cross-section ratios, such as the capture-to-fission ratio for various isotopes, and the relative contribution of the ^{238}U fast fissions, f_{238} (cf. Fig. 1). Reactor-specific

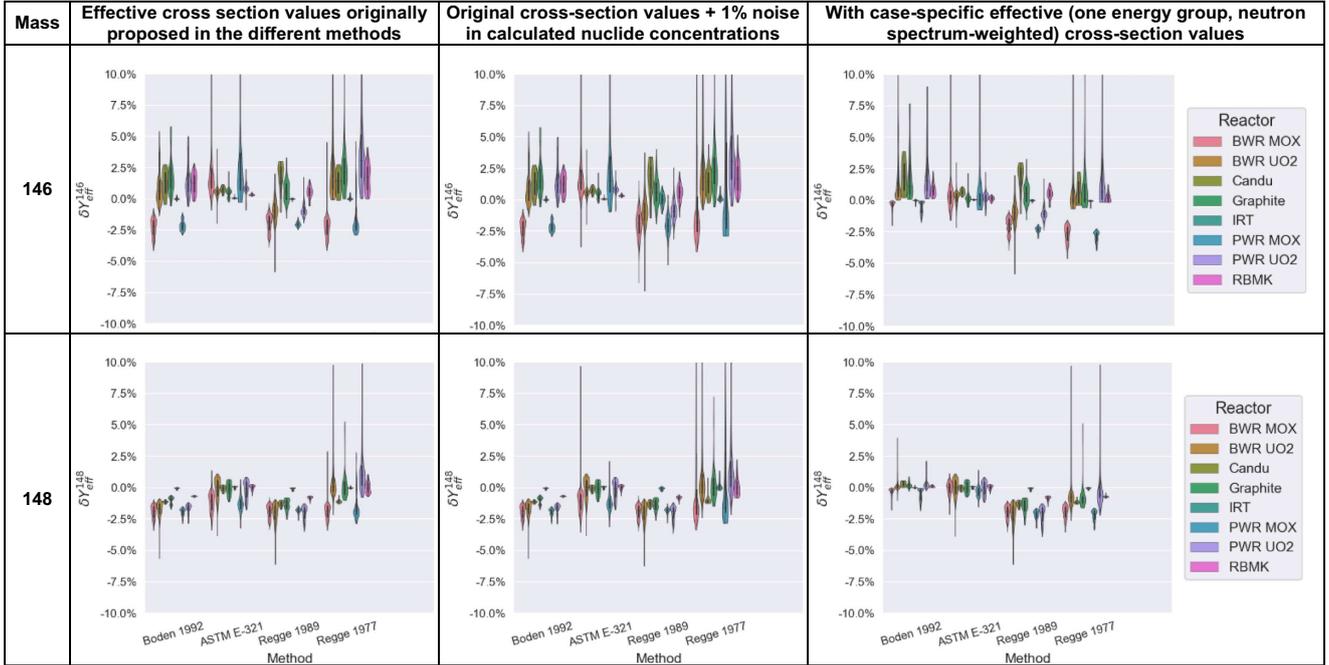


Fig. 2. Ratio of the irradiation-averaged fission yields predicted by the different burnup determination methods (from the calculated fuel inventories) to the simulation-based irradiation-averaged fission yields for mass series 146 and 148. The comparison provides an integral assessment of the distribution of fissions predicted by the different burnup determination methods. In the first column (“original”), the set of cross-section parameters as proposed in the individual articles [13,14,16,17] is used. The second column illustrates the sensitivity to experimental scatter, which is simulated by means of noise to the nuclide concentrations in the assessment database, assuming a normal distribution (1% standard deviation) around the calculated value. The last column illustrates the performance of the different methods when the case-specific effective cross-section values are used to calculate the irradiation-averaged fission yields.

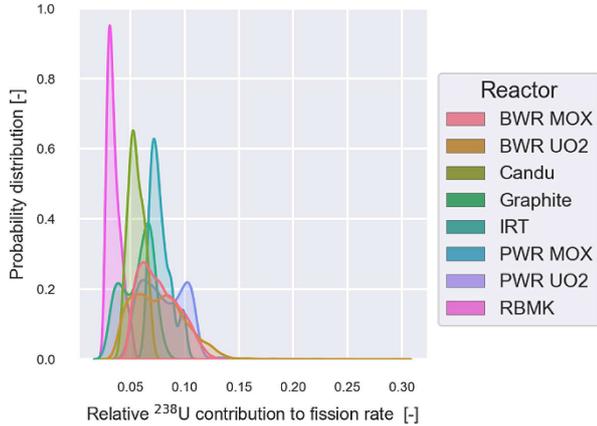


Fig. 3. Distribution of the ^{238}U relative contribution to fissions for all cases from the assessment database. A non-negligible amount of fissions originate from ^{238}U , as the low cross-section is somewhat balanced by the large ^{238}U content in low-enriched uranium fuels.

values for those parameters were also derived and are reported in the second part of Table 2. In light of the limitations pointed out in Section 2.1, the ^{242}Pu inventory in this parameter set is corrected for activations beyond that required by the original method (for this nuclide).

5.2 Improvements to the heavy element atom percent fission determination

5.2.1 Fission contribution for ^{235}U

The contribution of ^{235}U to fissions is very well dealt with by the heavy element isotopic ratios method [11], where the total number of fissions attributed to ^{235}U is derived from the depletion of ^{235}U or, alternatively, from the build-up of ^{236}U . This latter relationship was selected to be part of the improved burnup determination method proposed here:

$$F_{235\text{U}} = (N_{235\text{U},\text{BOL}} - N_{235\text{U},\text{EOL}}) - (N_{236\text{U},\text{EOL}} \cdot f_c - N_{236\text{U},\text{BOL}}) \quad (5a)$$

$$F_{235\text{U}} = (N_{235\text{U},\text{BOL}} - N_{235\text{U},\text{EOL}}) \cdot \frac{\sigma_f^{235\text{U}}}{\sigma_a^{235\text{U}}} \quad (5b)$$

$$F_{235\text{U}} = (N_{236\text{U},\text{EOL}} \cdot f_c - N_{236\text{U},\text{BOL}}) \cdot \frac{\sigma_f^{235\text{U}}}{\sigma_a^{235\text{U}}} \quad (5c)$$

with f_c is a correction factor for ^{236}U depletion [13]:

$$f_c = \frac{\sigma_a^{235\text{U}} - \sigma_a^{236\text{U}}}{\sigma_a^{235\text{U}}} \times \frac{1 - \frac{N_{235\text{U},\text{EOL}}}{N_{235\text{U},\text{BOL}}}}{\exp\left(\frac{\sigma_a^{236\text{U}}}{\sigma_a^{235\text{U}}} \cdot \log \frac{N_{235\text{U},\text{EOL}}}{N_{235\text{U},\text{BOL}}}\right) - \frac{N_{235\text{U},\text{EOL}}}{N_{235\text{U},\text{BOL}}}} \quad (6)$$

Table 2. Reactor-specific parameters for burnup monitors based methods: (a) effective fission cross-section ratios used in irradiation-averaged fission yield evaluation [14,16–18]; (b) ^{238}U contribution to fission and effective absorption cross sections used in the method proposed in Section 5.2; (c) effective absorption-to-fission cross-section ratios for use in the ASTM E244 and E321 methods [13,30].

	MOX	UO ₂				U alloy
	PWR/BWR <10% Pu	PWR/BWR <6% enr.	Magnox/AGR <5% enr.	RBMK <3% enr.	Candu Nat. U	MTR HEU (20–90% enr.)
(a) $\sigma_{f,239\text{Pu}}/\sigma_{f,235\text{U}}$	1.8	2.4	2.8	←(*)	1.8	←
$\sigma_{f,241\text{Pu}}/\sigma_{f,235\text{U}}$	2.2	2.6	2.8	←	2.1	←
$\sigma_{f,238\text{U}}/\sigma_{f,235\text{U}}$	62×10^{-4}	23×10^{-4}	7.9×10^{-4}	←	4.3×10^{-4}	←
(b) $f_{238} = \frac{F_{238\text{U}}}{F_{\text{tot}}}$	7.80%	7.80%	5.60%	3.60%	5.20%	0.50%
$\sigma_{a,235\text{U}}/\sigma_{f,235\text{U}}$	1.3	1.22	1.2	←	1.18	1.2
$\sigma_{a,236\text{U}}/\sigma_{f,235\text{U}}$	0.41	0.16	0.073	←	0.035	0.073
(c) $\sigma_{a,239\text{Pu}}/\sigma_{f,235\text{U}}$	1.59	←	←	←	←	←
$\sigma_{a,241\text{Pu}}/\sigma_{f,235\text{U}}$	1.49	←	←	←	←	←

(*)←: same value as in previous column.

As mentioned above (see Sect. 2.1), either formulation may be chosen to derive the fission contribution from ^{235}U . Generally, the formulation of equation (5c) will yield the most accurate results, except when working with reprocessed uranium, for which the fresh fuel may contain a significant initial amount of ^{236}U . Generic values for effective absorption to fission cross section ratios are given in Table 2 (strictly, Tab. 2 gives effective absorption to fission ratios).

5.2.2 Fission contribution of ^{238}U

As for the heavy element isotopic ratios method, the contribution of ^{238}U by fast fission is not derived from isotopic analysis but estimated from the reactor-specific fast fission factor f_{238} . Figure 3 indeed shows that that parameter varies little for each reactor and fuel type.

$$F_{238\text{U}} = f_{238} \cdot F_{\text{tot}} \quad (7)$$

5.2.3 Fission contribution of ^{239}Pu and ^{241}Pu

The derivation of Pu isotopes contribution to fissions in the heavy element isotopic ratios method is not retained in view of the deficiencies formerly discussed. In de Regge [13,16,30], the ratio of contributions from ^{239}Pu and ^{241}Pu is used. However, for UO₂ fuel, the method has a singular value at zero burnup, and its original formulation assumed a linear evolution with burnup of the ^{239}Pu and ^{241}Pu fission contributions, which is also not the case for UO₂ fuel. In order to improve on these shortcomings, we have assessed the effective fission cross-section ratio and the irradiation-averaged $^{241}\text{Pu}/^{239}\text{Pu}$ concentration ratio from the calculated dataset:

$$\frac{F_{241\text{Pu}}}{F_{239\text{Pu}}} = \frac{\sigma_{f,241\text{Pu}}}{\sigma_{f,239\text{Pu}}} \left\langle \frac{N_{241\text{Pu}}}{N_{239\text{Pu}}} \right\rangle \quad (8)$$

Values for the ratio of ^{241}Pu and ^{239}Pu effective fission cross sections are readily derived from the parameter set in Table 2. The evolution of the relative inventory

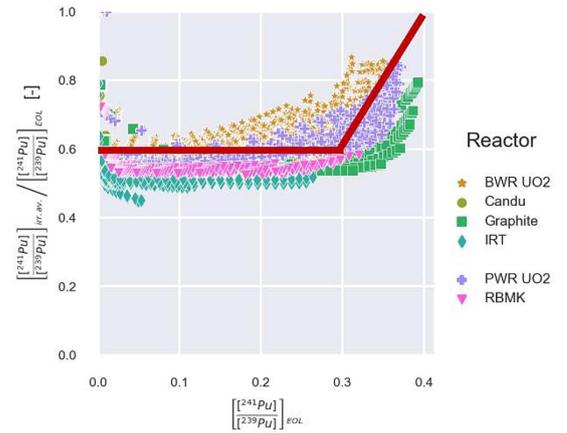


Fig. 4. Ratio of the irradiation-average ^{241}Pu -to- ^{239}Pu concentration ratio for all cases of the assessment database, with its EOL value for uranium-based fuels, plotted as a function of the EOL concentration ratio. The red line is the approximation proposed in this work (Eq. (8)).

of ^{241}Pu compared to ^{239}Pu was investigated for the case of UO₂ and MOX fuels separately. The effective (irradiation-averaged) $^{241}\text{Pu}/^{239}\text{Pu}$ concentration ratio has been plotted in Figure 4 as a fraction of its EOL value. The proportionality value ranges from 0.55 to 0.67 before saturation of ^{241}Pu occurs and climbs progressively towards a value of unity at higher burnups, when saturation of ^{241}Pu and ^{239}Pu is achieved throughout the majority of the irradiation period. This ratio mostly affects the ^{241}Pu contribution to fissions, which, in most cases, is a small contributor to fissions compared to ^{239}Pu ². A simple and robust relationship is then sufficient to determine the irradiation-averaged fission yields of burnup monitors. In the case of uranium-based fuels, therefore, we propose assigning a constant proportionality value of 0.6 when the

² The ^{241}Pu -to- ^{239}Pu cross-section ratio is about 1, and the irradiation-average inventory ratio remains below 0.3.

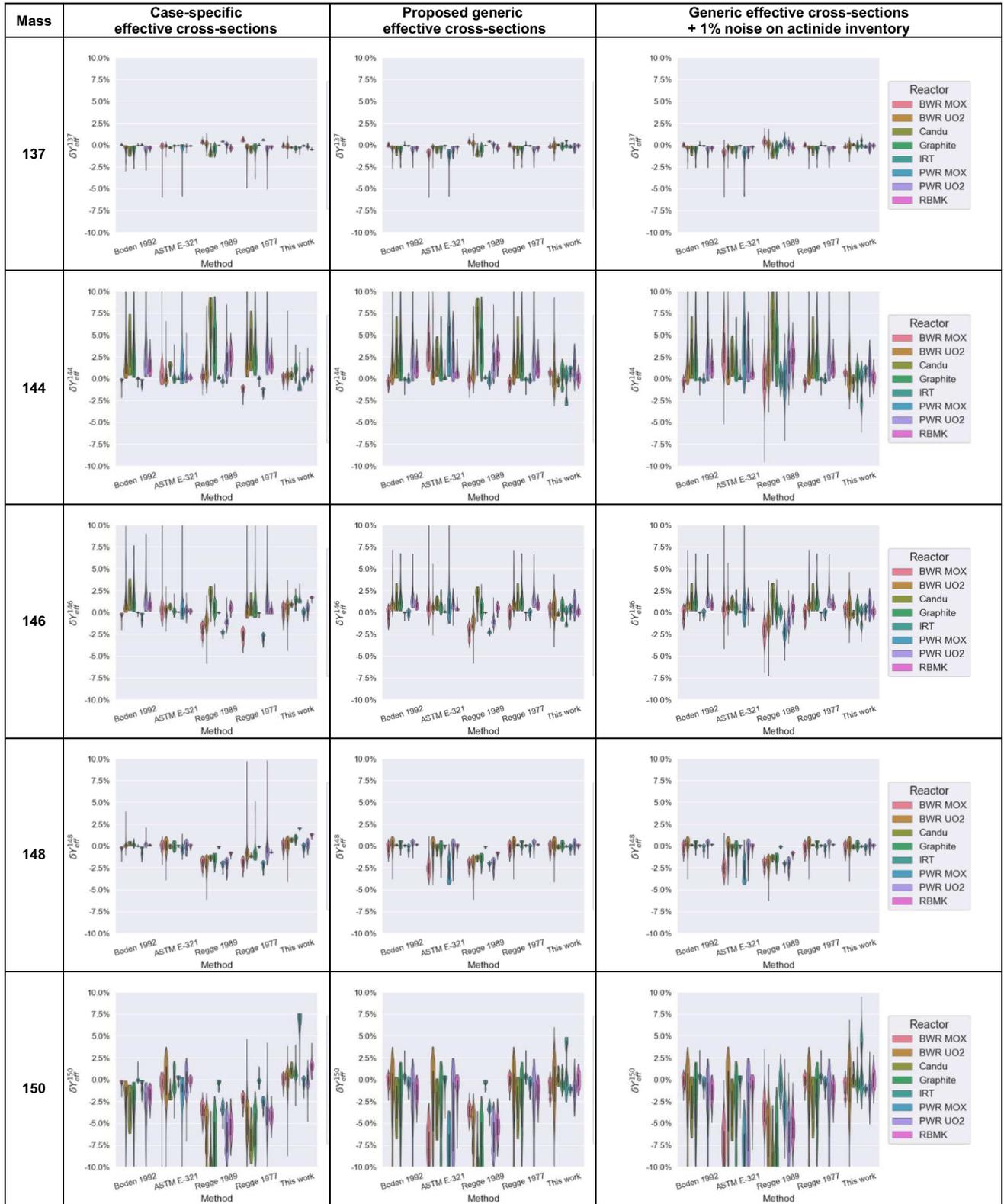


Fig. 5. Ratio of the irradiation-averaged fission yield values estimated by the different burnup determination methods, to the simulation-based irradiation-averaged fission yield values. The comparison offers an integral assessment of the distribution of fissions predicted by the different burnup determination methods, including the one proposed in this work. In the first column, the ORIGEN (calculated, case-specific) effective cross-section values are used. The second column illustrates the performance of the various methods, with the set of generic effective cross-sections as proposed in Table 2. The last column illustrates the sensitivity of the methods to scatter, which is simulated as 1% noise in the actinide inventory, assuming a normal distribution.

Table 3. Comparison of the burnup predictions with the Boden 1992 method [17] and the method proposed in this work, for several RCA assays in SCK CEN laboratories. Various reactor designs and fuel types were investigated, including gadolinium-doped fuel.

Reactor	Fuel	BU	Boden 1992 [17]					This work					
			BU(i)/BU - 1					BU(i)/BU - 1					
			¹³⁷ Cs	¹⁴⁸ Nd	¹⁵⁰ Nd	¹⁴³ Nd + ¹⁴⁴ Nd	¹⁴⁵ Nd + ¹⁴⁶ Nd	¹³⁷ Cs	¹⁴⁸ Nd	¹⁵⁰ Nd	¹⁴³ Nd d + ¹⁴⁴ Nd	¹⁴⁵ Nd + ¹⁴⁶ Nd	
BWR	UO ₂	5.78	-7%	2%	7%	-2%	0%	-1%	-6%	2%	4%	0%	
	UO ₂	5.81	3%	0%	5%	-5%	-2%	-1%	3%	0%	0%	-2%	
	UO ₂	6.21	6%	-1%	0%	-4%	-1%	1%	8%	-2%	-1%	-3%	
	UO ₂ -PuO ₂	3.78	-4%	1%	0%	1%	2%	0%	-3%	0%	-1%	2%	
	UO ₂ -PuO ₂	4.00	-3%	1%	-2%	2%	3%	0%	-2%	0%	-3%	3%	
	UO ₂ -PuO ₂	5.12	-3%	1%	-1%	1%	1%	-1%	-1%	1%	-2%	2%	
	UO ₂ -PuO ₂	5.42	1%	0%	-1%	0%	1%	-1%	3%	-1%	-2%	0%	
	UO ₂ -PuO ₂	6.20	-5%	1%	0%	2%	3%	0%	-3%	0%	-1%	2%	
MTR	U(Mo)	10.68	-4%	6%	0%	0%	-1%	2%	-5%	6%	-8%	5%	
	UO ₂	4.84	2%	0%	5%	-5%	-3%	-1%	3%	0%	0%	-1%	
PWR	UO ₂	5.34	-3%	2%	5%	-3%	-1%	0%	-2%	1%	-1%	1%	
	UO ₂	5.35	-4%	3%	7%	-4%	-1%	0%	-4%	2%	1%	0%	
	UO ₂	5.69	-2%	2%	5%	-4%	-1%	-1%	-1%	1%	-1%	0%	
	UO ₂	6.09	-3%	2%	7%	-5%	-2%	0%	-3%	2%	1%	0%	
	UO ₂	7.16	3%	0%	4%	-5%	-2%	2%	4%	0%	-2%	-2%	
	UO ₂	7.32	2%	1%	3%	-4%	-1%	4%	3%	0%	-3%	0%	
	UO ₂ -Gd ₂ O ₃	1.44	-8%	3%	17%	-8%	-4%	2%	-7%	3%	6%	-1%	
	UO ₂ -Gd ₂ O ₃	1.44	-7%	3%	16%	-8%	-4%	2%	-6%	3%	4%	-1%	
	UO ₂ -PuO ₂	4.56	-4%	1%	1%	0%	2%	0%	-2%	1%	0%	1%	
	UO ₂ -PuO ₂	5.68	-6%	1%	1%	1%	3%	0%	-5%	1%	0%	2%	
	PWR(*)	UO ₂ -PuO ₂	1.06	1%	-1%	1%	0%	-1%	-2%	3%	-2%	0%	1%
		UO ₂ -PuO ₂	3.78	-1%	0%	0%	0%	1%	-1%	0%	0%	0%	1%
		UO ₂ -PuO ₂	5.66	3%	-1%	-3%	0%	0%	-1%	5%	-2%	-3%	1%
		UO ₂ -PuO ₂	5.66	3%	-1%	-3%	0%	0%	-1%	5%	-2%	-3%	1%

(*): PWR-representative conditions in non-commercial (research) reactors.

²⁴¹Pu/²³⁹Pu concentration ratio at EOL is less than 0.3, and a linear relationship beyond this:

$$\left\langle \frac{N_{241\text{Pu}}}{N_{239\text{Pu}}} \right\rangle = \begin{cases} 0.6 \cdot \frac{N_{241\text{PuEOL}}}{N_{239\text{PuEOL}}} & \frac{N_{241\text{PuEOL}}}{N_{239\text{PuEOL}}} < 0.3 \\ 4 \cdot \frac{N_{241\text{PuEOL}}}{N_{239\text{PuEOL}}} - 0.6 & \frac{N_{241\text{PuEOL}}}{N_{239\text{PuEOL}}} \geq 0.3 \end{cases} \quad (9)$$

In MOX fuels, the evolution of plutonium isotope concentrations is small. The average of BOL and EOL concentrations then performs satisfactorily, as seen for the Boden 1992 method [17]:

$$\left\langle \frac{N_{241\text{Pu}}}{N_{239\text{Pu}}} \right\rangle = \frac{1}{2} \left(\frac{N_{241\text{PuBOL}}}{N_{239\text{PuBOL}}} + \frac{N_{241\text{PuEOL}}}{N_{239\text{PuEOL}}} \right) \quad (10)$$

5.2.4 Closure relation for the fission distribution

The system needs to be closed by an additional relationship, such as the expression of the total number of fissions. The derivation of that quantity from the depletion of actinides has the advantage of not correlating the irradiation-averaged fission yields with the inventory of a burnup monitor. However, it involves the difference of two large numbers, which is prone to significant numerical error propagation. A relative error of 1% on the ²³⁸U concentration would propagate to 20% to 50% error on the predicted total amount of fissions at typical LWR burnups. The relation between the number of fissions and the concentration of a burnup monitor is then preferred, as those typically have zero concentrations at BOL and the uncertainty on the total amount of fissions then remains of

the same order of magnitude as that of the burnup monitor concentration. Similar considerations led us to prefer evaluating the number of ²³⁵U fissions from the ²³⁶U build-up for enriched natural uranium fuels (no ²³⁶U in the fresh material) or for reprocessed uranium fuel with low ²³⁶U content as compared to the EOL inventory. Any of the burnup monitors may be selected for this purpose, but since the fission yield of ¹⁴⁸Nd has the least variation with fission isotope concentration, we selected ¹⁴⁸Nd for our closure equation. In that case, the closure equation reads as:

$$\sum_j Y_j^{148\text{Nd}} F_j = N_{148\text{Nd}} \quad (11)$$

with $Y_j^{148\text{Nd}}$ the fission yield of ¹⁴⁸Nd for the fission of isotope j ($j = ^{235}\text{U}, ^{239}\text{Pu}, ^{241}\text{Pu}, ^{238}\text{U}$). The proposed method consists of solving the set of equations (5a)–(5c), (7), (8) and (11). Adding the expression for the total number of fissions, it may be re-written as a set of 5 linear equations with the unknown $F_{235\text{U}}, F_{238\text{U}}, F_{239\text{Pu}}, F_{241\text{Pu}}, F_{\text{tot}}$; which can be solved quite easily. Taking as an example the case where the contribution from ²³⁵U is derived from ²³⁶U build-up in equations (5a)–(5c), this yields equation (12):

$$\begin{cases} F_{235\text{U}} + F_{238\text{U}} + F_{239\text{Pu}} + F_{241\text{Pu}} - F_{\text{tot}} = 0 \\ F_{235\text{U}} = \left(N_{236\text{U,EOL}} \cdot f_c - N_{236\text{U,BOL}} \right) \cdot \frac{\sigma_f^{235\text{U}}}{\sigma_a^{235\text{U}}} \\ F_{238\text{U}} - f_{238} \cdot F_{\text{tot}} = 0 \\ F_{241\text{Pu}} - \frac{\sigma_f^{241\text{Pu}}}{\sigma_f^{239\text{Pu}}} \left\langle \frac{N_{241\text{Pu}}}{N_{239\text{Pu}}} \right\rangle F_{239\text{Pu}} = 0 \\ Y_{235\text{U}}^{148\text{Nd}} F_{235\text{U}} + Y_{238\text{U}}^{148\text{Nd}} F_{238\text{U}} + Y_{239\text{Pu}}^{148\text{Nd}} F_{239\text{Pu}} + Y_{241\text{Pu}}^{148\text{Nd}} F_{241\text{Pu}} = N_{148\text{Nd}} \end{cases} \quad (12)$$

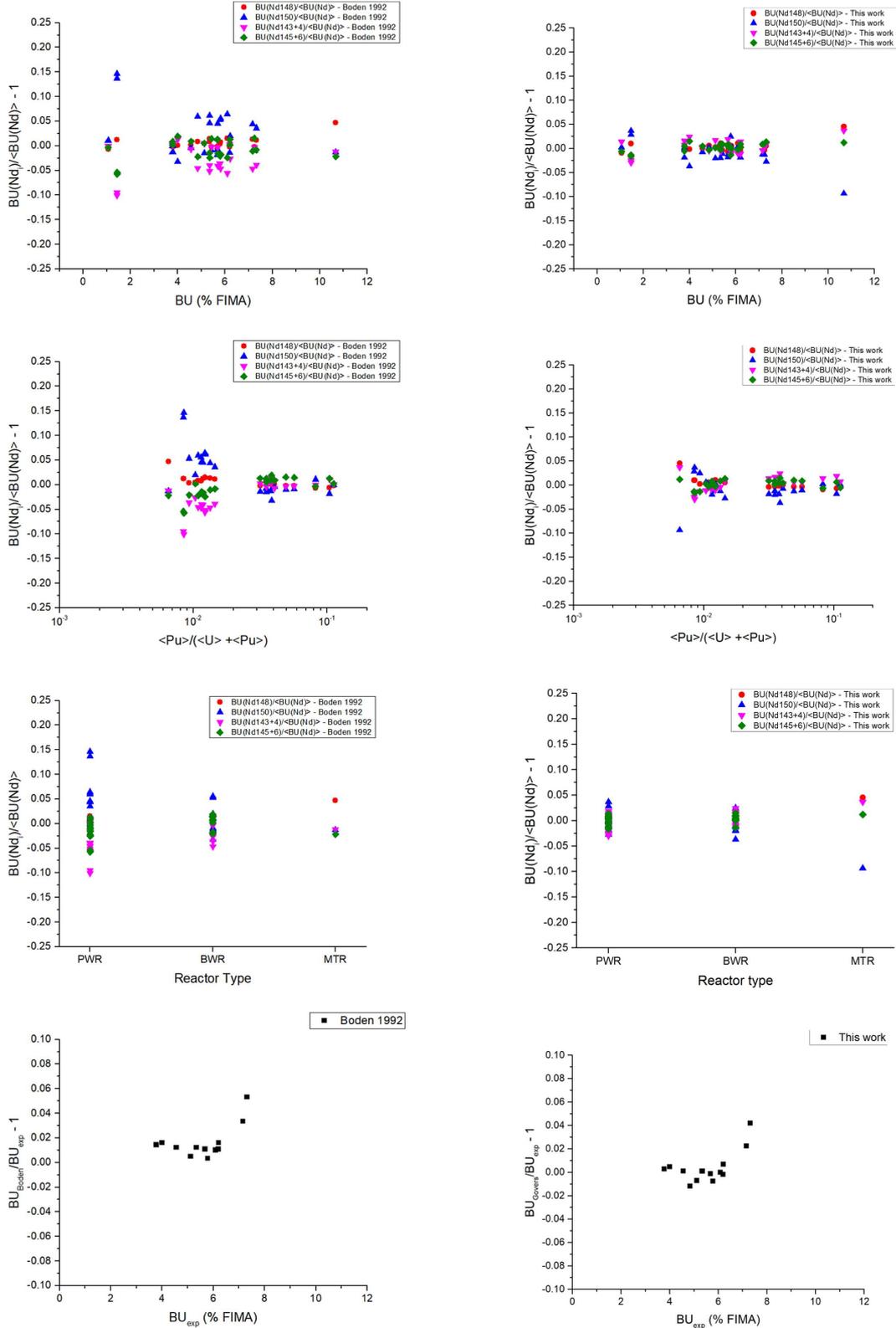


Fig. 6. Burnup derived from different Nd isotopes (^{148}Nd , ^{150}Nd , $^{143}\text{Nd}+^{144}\text{Nd}$, and $^{145}\text{Nd}+^{146}\text{Nd}$) according to the approach proposed by Boden et al. [17] (left column) and in this work (right column). The same set of data is expressed as a function of burnup (top row), Pu content (second row) and reactor type (third row). The ratio of average burnup to reported burnup is presented in the bottom row. Here, only data for which the burnup was assessed by cross check analyses are withheld.

with f_c and $\left\langle \frac{N_{241\text{Pu}}}{N_{239\text{Pu}}} \right\rangle$ defined in equations (6) and (9) (or (10) for MOX fuels), respectively.

5.3 Assessment of the proposed changes

The performance of the derived set of effective cross-sections, when applied to the existing methods, is illustrated in Figure 5; the graphs also reveal the performance of the new method proposed here. For the purpose of clarity, ^{143}Nd data are not shown, but exhibit a trend similar to that of ^{144}Nd ; the same for ^{145}Nd , which exhibit a behaviour similar to that of ^{146}Nd . The comparison illustrates on the left the optimal approach, where case-specific effective cross-sections are used for all methods; in the middle, the performance of all methods with the proposed generic set of effective cross-sections (Tab. 2), and on the right, the same evaluation for a perturbed inventory database, where 1% noise is introduced to simulate experimental scatter.

Figure 5 indicates that the proposed set of cross-section parameters performs for all methods about as well as the case-specific values. Most calculation methods, however, still have an appreciable scatter in the irradiation-averaged fission yield derived from nuclides ^{144}Nd , ^{150}Nd , and ^{143}Nd (the latter not being represented in Fig. 5). The method proposed in Section 5.2 is the only one with narrow spread on all isotopes: less than 1% for ^{137}Cs and ^{148}Nd , and in most cases below 5% for $^{143-146}\text{Nd}$ and ^{150}Nd . It also shows almost no bias compared to calculated cases. We therefore suggest adopting this approach for future burnup determinations in radiochemical assays, as it offers relative simplicity combined with an elevated level of predictability.

5.4 Application to earlier radiochemical assays

The performance of the proposed method compared to that currently in use at SCK CEN (Boden 1992, [17]) has been evaluated using a series of radiochemical assays conducted in the laboratory, covering UOX, MOX and U(Mo) fuels irradiated in PWR, BWR and MTR reactors [21,28,29,40]. The comparison is detailed in Table 3 and illustrated in Figure 6. It shows that the novel method significantly reduces the scatter between burnup monitor isotopes, in particular for the UOX fuels. When assessed on samples for which independent cross-checks exist for the burnup (Fig. 6, bottom row), the proposed method shows no significant bias, while a bias of about 2% is observed for the Boden 1992 method.

The scatter of burnup values for individual burnup monitors is reduced substantially for most burnup monitors, except for ^{148}Nd where it may slightly increase for several “real” cases. It should, however, be noted that the deviation of the ^{148}Nd -burnup had the smallest deviation from the average burnup from all burnup monitors, so that all-in-all a better performance is observed, especially for low burnup PWR gadolinium-doped fuel samples. This demonstrates the superior performance of the proposed

method even for fuel types that are, strictly speaking outside the domain considered for constructing the assessment database. The improvement is smaller in the case of MOX fuels, for which the Boden 1992 method already performs well; this is in line with smaller uncertainties on contributors to fissions in MOX fuels.

6 Conclusions

A new method has been proposed in this work for the calculation of the irradiation-averaged fission contribution of the fissile and fertile nuclides relevant to most spent nuclear fuel burnup assays. Together with a physically-informed set of effective (one-group, neutron spectrum-averaged) cross-sections, it enables one to readily calculate the effective irradiation-averaged fission yield of selected fission products. The method is demonstrated in the present paper for nuclides of general interest for burnup determination, but may also be extended to estimate the irradiation-averaged cumulative fission yield of other fission product decay series.

When comparing the newly proposed method with existing methods, it was observed that the scatter and bias between burnup values derived from individual nuclides could be reduced significantly. The good performance of the method has been assessed both on a set of simulated spent fuel inventories and on real experimental cases, covering a wide range of reactor types, fuel enrichment and burnup.

The present method is not intended to replace or to outperform core physics calculations, but rather to enable derivation of the burnup of spent nuclear fuel from radiochemical assay data when it is either not possible or not desirable to perform dedicated core physics calculations. By resorting to a rapid and robust method, laboratories may, for example, perform a rapid consistency check of the experimentally determined nuclide concentrations without having to resort to more elaborate calculation efforts.

Conflict of interests

The authors declare that they have no competing interests to report.

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Appendix A Accuracy of the irradiation-averaged fission yield

This manuscript will further address the efficiency of different burnup determination methods based on burnup monitor inventories. In the present section, one will seek to quantify aspects related to the accuracy of the determination of the irradiation-averaged fission yield. All methods presented in Section 2.2 ultimately express the irradiation-averaged fission yield as a weighted sum of relative contributions to fission from the various actinides (f_j):

$$\bar{Y}_i = \sum_j f_j Y_j^i \quad (\text{A.1})$$

with $\sum_j f_j = 1$. The uncertainty on the irradiation-averaged fission yield, resulting from uncertainties on the relative contributions to fission, can be evaluated as follows. Let us consider an evaluated distribution of actinide contributions to fissions, (\tilde{f}_j), that deviates from the true distribution as: $\tilde{f}_j = f_j(1 + \delta f_j)$ (with δf_j the relative difference of contribution to fission from nuclide j). The relation $\sum_j \tilde{f}_j = 1$ also holds. The irradiation-averaged fission yield for the evaluated case gives:

$$\tilde{Y}_i = \bar{Y}_i(1 + \delta \bar{Y}_i) = \sum_j \tilde{f}_j Y_j^i = \sum_j f_j (1 + \delta f_j) Y_j^i$$

The relative difference in the irradiation-averaged fission yield for burnup monitor i is given by:

$$\delta \bar{Y}_i = \frac{\sum_j f_j \delta f_j Y_j^i}{\sum_j f_j Y_j^i}$$

If one isolates one of the contributions, arbitrarily taken as $j = 0$, one obtains:

$$\delta \bar{Y}_i = \frac{f_0 \delta f_0 Y_0^i + \sum_{j \neq 0} f_j \delta f_j Y_j^i}{f_0 Y_0^i + \sum_{j \neq 0} f_j Y_j^i} = \frac{f_0 \delta f_0 + \sum_{j \neq 0} f_j \delta f_j \cdot Y_j^i / Y_0^i}{f_0 + \sum_{j \neq 0} f_j \cdot Y_j^i / Y_0^i} \quad (\text{A.2})$$

Considering the relations $\sum_j f_j = 1$ and $\sum_j \tilde{f}_j = 1$ ($\rightarrow \sum_j \delta f_j = 0$), and introducing $\Delta Y_j^i = (Y_j^i / Y_0^i - 1)$, one finally gets:

$$\delta \bar{Y}_i = \frac{\sum_{j \neq 0} \delta f_j \cdot f_j \cdot (Y_j^i / Y_0^i - 1)}{1 + \sum_{j \neq 0} f_j \cdot (Y_j^i / Y_0^i - 1)} = \frac{\sum_{j \neq 0} \delta f_j \cdot f_j \cdot \Delta Y_j^i}{1 + \sum_{j \neq 0} f_j \cdot \Delta Y_j^i} \quad (\text{A.3})$$

- The relation reflects the intuitive reasoning that uncertainties on the irradiation-averaged fission yield diminish when the relative fission distribution is accurately known ($\delta f_j \rightarrow 0$).
- The weight of actinide j in the number of fissions is reduced ($f_j \rightarrow 0$).
- The fission yields are similar for dominant contributors ($Y_j^i / Y_0^i \rightarrow 1$, or $\Delta Y_j^i \rightarrow 0$).

For LWR UOX cases, the contributions to fission, f_j , from ^{239}Pu over the entire irradiation is typically below 60%, and

Table A.1. Accuracy of the irradiation-averaged fission yield for different deviations (40%, 10% and 5%) on the relative contributions to fissions of ^{239}Pu , ^{238}U and ^{241}Pu .

Mass series	$\delta \bar{Y}_i (\delta f_j < 40\%)$	$\delta \bar{Y}_i (\delta f_j < 10\%)$	$\delta \bar{Y}_i (\delta f_j < 5\%)$
98	0.5%	0.1%	0.1%
99	0.3%	0.1%	0.0%
133	1.2%	0.3%	0.1%
134	0.8%	0.2%	0.1%
137	1.9%	0.5%	0.2%
148	1.9%	0.5%	0.2%
143	11.5%	2.9%	1.4%
144	13.5%	3.4%	1.7%
145	8.5%	2.1%	1.1%
146	4.3%	1.1%	0.5%
150	14.3%	3.6%	1.8%

those of ^{238}U and ^{241}Pu below 15%. Using equation (6), an evaluation of the accuracy of the irradiation-averaged fission yields for different burnup monitors is reported in Table A.1; it reflects that an accuracy of better than 2% on the irradiation-averaged fission yield is obtained when the deviation on the contribution to fissions, δf_j , is less than 40% for each of the fissioning isotopes for mass series 137 and 148, when the deviation is less than 10% for mass series 145 and 146, and when the deviation is less than 5% for the mass series 143, 144 and 150.

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