DOI: 10.15669/pnst.8.381

Progress in Nuclear Science and Technology
Volume 8 (2025) pp. 381-386

ARTICLE

Development of an Inductive Method using Ordinary Least Squares Regression Technique in Nuclear Forensics

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Nuclear forensics involves examining nuclear materials and other radioactive substances, seized out of regulatory control, to determine their origin. In the case of spent nuclear fuel, the goal is to identify its initial composition (enrichment, plutonium content), reactor type, and the conditions of its irradiation (burnup, cooling time), based on the isotopic composition of the analyzed sample. In order to solve the inverse problem, several methods can be employed. CEA has developed a range of methods for that purpose, one of them being the "inductive method". Using a numerical model solving the problem of depletion under irradiation and during cooling, this method consists in determining the expected target (^{235}U enrichment, plutonium content, reactor type, burnup and cooling time) based on marker isotopes. We have identified relevant marker isotopes and established differentiation criteria able to differentiate between light water-moderated reactors. Our method allows predicting the expected targets with an ordinary least squares regression technique in the case of pressurized water reactors and boiling water reactors.

KEYWORDS: nuclear forensics, spent nuclear fuel, enrichment, burnup, isotopic composition, inverse methods

I. Introduction

In support of investigations, nuclear forensics uses various methods to interpret the analytical result of a seized nuclear material sample. The aim is to determine the target parameters (irradiation conditions and initial fuel composition) using its signature: the isotopic composition of the sample and the presence of marker isotopes.

Historically, one method often put forward is the inductive method.[1,2)] This method is based on solving the inverse problem of irradiated fuel depletion, and assumes that for an analyzed signature, one and only one evolutionary pathway exists. A numerical model, based on the physical reactions that occur during the irradiation process in the reactor and during cooling, is then used to reproduce the depletion of the isotopic composition undergone by the fuel. Finally, the inverse problem is solved by iteratively tracing the fuel's initial composition and irradiation characteristics.

II. The Development of the Inductive Method at CEA

The inductive approach has been studied at CEA since 2019. It uses the ordinary least squares method (*Méthode Inverse par Moindres Carrés Ordinaire* (MIMCO) in French). This method has been first described by Legendre.[3)] For instance, the initial enrichment, e_5, and the burnup, BU, are the two unknown quantities, forming a target vector X:

$$X = \begin{pmatrix} e_5 \\ BU \end{pmatrix} \quad (1)$$

The marker isotopes that will allow to determine the unknown quantities are Y_1 and Y_2 forming a marker vector Y:

$$Y = \begin{pmatrix} Y_1 \\ Y_2 \end{pmatrix} \quad (2)$$

The derivative matrix S is defined as:

$$S = \begin{pmatrix} \frac{\partial Y_1\ calculated}{\partial e_5} & \frac{\partial Y_1\ calculated}{\partial BU} \\ \frac{\partial Y_2\ calculated}{\partial e_5} & \frac{\partial Y_2\ calculated}{\partial BU} \end{pmatrix} \quad (3)$$

If the problem is non-linear, the solution is obtained iteratively with the following formula:

$$X_{n+1} = X_n + (S_n^{\mathrm{T}}.S_n)^{-1} \cdot S_n \cdot (Y_{\mathrm{measured}} - Y_{\mathrm{calculated}}(X_n)) \quad (4)$$

with X_n the estimation of the target vector at the iteration n, $Y_{\mathrm{calculated}}(X_n)$ the maker vector and S_n the derivative matrix calculated with a depletion code using the quantities of the target vector at the iteration n, *i. e.* X_n. Y_{measured} is a marker vector made with measured values of the marker isotopes Y. We perform the depletion calculations with the CESAR code.[4)]

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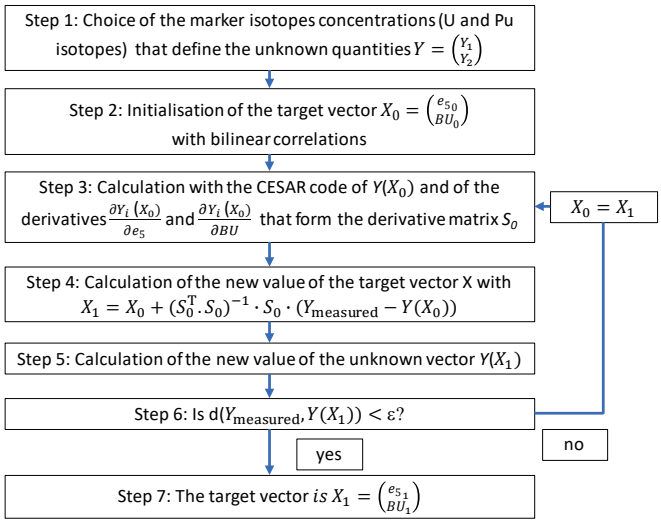

Fig. 1 Flow chart of the MIMCO method

The **Fig. 1** illustrates the inductive method with a flow chart. The different operations are the following:

- Step 1: Choice of the marker isotopes concentrations (U and Pu isotopes) that define the unknown quantities as ratios of isotope concentration. These unknown concentration ratios are grouped in a vector, *e. g.* with two ratios, $Y = \begin{pmatrix} Y_1 \\ Y_2 \end{pmatrix}$,
- • Step 2: One initializes the target quantities we wish to determines, *e. g.* the initial enrichment in ^{235}U, e_5, and the burnup, BU, thanks to bilinear correlations (see section III), make the initial target vector $X_0 = \begin{pmatrix} e_{5_0} \\ BU_0 \end{pmatrix}$,
- • Step 3: One calculates the values of the isotope concentration ratios with the CESAR depletion code using the initial target values to obtain $Y(X_0)$ and the derivative of the $Y_i(X_0)$ quantities relatively to the target quantities, $\frac{\partial Y_i(X_0)}{\partial e_5}$ and $\frac{\partial Y_i(X_0)}{\partial BU}$, allowing to form the derivative matrix S_0 given by (3),
- • Step 4: One uses the Legendre formula (4) to obtain the new value of the target vector X_1,
- • Step 5: One calculates of the new value of the unknown vector $Y(X_1)$,
- Step 6: One compares the distance between this new vector $Y(X_1)$ and the vector made with measurements Y_{measured}, $d(Y_{\mathrm{measured}}, Y(X_1))$. If this distance is lower than a chosen small value ε, the algorithm stops and one obtains the desired target vector as $X_1 = \begin{pmatrix} e_{5_1} \\ BU_1 \end{pmatrix}$. Otherwise, one goes back to step 3 with X_0 taking the value of X_1.

The depletion calculations are performed with the CESAR code using the values of the target as input parameters. The inverse method uses the ordinary least squares method MIMCO to determine iteratively the unknown values of the target vector.

In our early work, the prediction of target parameters was based on the use of a wide range of marker isotopes, including ^{148}Nd (marker of burnup) and ^{137}Cs (marker of cooling time), as well as ^{146}Nd, ^{241}Am, ^{155}Gd and ^{156}Gd in order to discriminate between moderate and light water reactors. The major disadvantage of using such isotopes in a forensic investigation is that obtaining precise concentrations by mass spectrometric analysis (*e. g.* TIMS or MC-ICP-MS) can take several weeks or even months. A further constraint lies in the formalism of the sample analysis results: these are provided as ratios of isotopes of the elements considered, rather than as absolute atomic concentrations. The latest version of our work uses Uranium and Plutonium isotope ratios only.

III. Bilinear Correlations for Inverse Method Initialization

Initial ^{235}U enrichment (e_5) and burnup (BU) are two key indicators for identifying the reactor in which an analyzed sample has been irradiated. In the inverse method, the initial estimate of these two targets is used as a reference for the first CESAR depletion calculation, which initiates the first MIMCO iterative step. If the initial estimates are too far from the true value, the MIMCO might not converge. To avoid to have to test several sets of initial conditions, it is then useful to have an initial estimate that is already close to the true value and that what will be presented below with bilinear correlations. Note however that MIMCO is quite a robust method: it converges in 2-4 iterations and the results doesn't depend of the initial guess. It is not really surprising, considering that MIMCO should converge in one step in case of a truly linear problem and that almost what we have with initial ^{235}U enrichment and burnup since they could be approximated with bilinear correlations.

Target parameters are generally linked to the depletion of marker isotopic elements by a coupled depletion and nonlinear transport equation. Multilinear relationships between key isotopic concentration ratios, initial enrichment and burning rate were recently established.[5] These relationships were tested with experimental data from the SFCompo 2.0[6] database and depletion calculations.

It was shown[5,7] that in many cases, a bilinear regression is sufficient to represent the experimental data set with sufficient accuracy. The nuclides of interest are uranium and plutonium isotopes. The ^{236}U/^{238}U ratio is proving to be a key marker for estimating initial enrichment and burnup. The ^{235}U/^{238}U, ^{240}Pu/^{236}U and ^{240}Pu/^{238}U ratios are effective for determining initial enrichment, and the ^{242}Pu/^{238}U, ^{242}Pu/^{240}Pu and ^{242}Pu/^{239}Pu ratios for burnup.

For example, the first bilinear correlation can be used to estimate the initial ^{235}U enrichment of a fuel irradiated in a PWR, based on the measurement of two isotopic ratios: ^{235}U/^{238}U and ^{236}U/^{238}U. The formula for calculating enrichment or burnup is as follows:

$$e_5\ (\%)\ or\ BU\ \left(\frac{GW_d}{ithm}\right) = c_0 + c_1.\frac{E_1}{E_2} + c_2.\frac{E_3}{E_4} \qquad (5)$$

E_i is the concentration of an isotope *i*. c_i are the correlation coefficients.

The study presented above was repeated by Chen *et al.*[7] and extended to boiling water reactors (BWRs), VVER reactors (440 and 1000), advanced gas-cooled reactors (AGRs) and high-power pressure tube reactors (HPRTs).

Based solely on computational data and extrapolating the simplifying assumptions made by Chen *et al.*[7], we have established bilinear correlations for new power reactor types (SFR, CANDU, *etc.*) or research reactors (see **Table 1** relatively to initial ^{235}U enrichment and **Table 2** relatively to burnup for PWR and BWR values). As data on irradiated samples from PWR and BWR reactors and CESAR libraries are available, we have established bilinear correlations on both experimental and calculated data. Comparison of the correlations obtained enables us to assess the quality of the computational method, and these predictions serve as an entry point for MIMCO. By measuring the ratios of interest used in the bilinear correlations, initial enrichment and burnup values can be estimated. There are as many estimates of these two targets as there are reactor types for which bilinear correlations have been established.

Table 1 Bilinear initial enrichment correlations, based on values calculated by the CESAR code

Reactor type	$\frac{E_1}{E_2}$	$\frac{E_3}{E_4}$	c_0	c_1	c_2	R^2	w_1	w_2
PWR	$\frac{^{235}U}{^{238}U}$	$\frac{^{236}U}{^{238}U}$	0.498	77.06	492.9	0.969	0.505	0.495
BWR	$\frac{^{235}U}{^{238}U}$	$\frac{^{236}U}{^{238}U}$	0.165	89.93	541.2	0.992	0.466	0.534

R^2 is the coefficient of determination, while w_i give the relative contribution of the two isotopic ratios to the linearity of the model.[7]

Table 2 Bilinear burnup correlations, based on values calculated by the CESAR code

Reactor type	$\frac{E_1}{E_2}$	$\frac{E_3}{E_4}$	c_0	c_1	c_2	R^2	w_1	w_2
PWR	$\frac{^{236}U}{^{238}U}$	$\frac{^{242}Pu}{^{238}U}$	0.680	4,811	21,403	0.999	0.268	0.732
BWR	$\frac{^{236}U}{^{238}U}$	$\frac{^{242}Pu}{^{238}U}$	3.228	8,658	3,555	0.764	0.987	0.013

Note that the relatively small value of R^2 for *BU* BWR correlation in Table 2 is due to the fact that the CESAR code provide calculation for 10 void fractions between 0.009 and 0.65 while the void fractions in SFCompo 2.0 vary between 0.12 and 0.68. More generally, a different combination of isotopes ratios could give a different correlation with different coefficient of determination R^2 and thus a different estimation results for *BU*.

IV. Deduction of the Reactor Type by Prediction of Target Parameters

We have developed a code that takes into account the results of irradiated sample analysis in the form of uranium and plutonium isotope ratios (up to 11 ratios). Via a pre-selection module, it proposes the probable reactor type in which the sample was irradiated. By compiling the coefficients of the bilinear correlations of a number of reactor types, the script calculates the initial estimate of the target parameters (enrichment and burnup) for each of them, then compares the values with the validity domains of the respective CESAR libraries. Lastly, the script indicates which reactor types are considered potential (if the targets are within the validity range) or unlikely (if not). As input, the user always selects a set of markers (*i.e.* ratios of U and Pu isotopes concentrations) on which the inverse method will be applied. The MIMCO can then be performed, at the user's discretion, on the reactor types deemed probable.

Assigning a reactor type may be straightforward in cases where the sample corresponds to an atypical plant in terms of initial enrichment, burnup, neutron energy spectrum or fuel type. However, for certain types of fuel, these characteristics are similar and attribution can be tricky. This is particularly the case for PWR and BWR reactors using UOx fuel, both of which are moderated with light water.

In this case, as the user does not know *a priori* how to allocate a fuel sample to a particular light-water moderated type, he will run the MIMCO with both PWR and BWR. Allocation will be made *a posteriori* by comparing the results obtained with the two reactor types.

The proposed method consists in studying the calculated deviation (C/M ratio) of the uranium and plutonium isotope ratios provided by the analysis of the irradiated sample, by comparing the convergence quality of the different U and Pu ratios obtained with the two MIMCOs run in parallel. Both methods can converge, but one path may converge faster and more accurately if the U and Pu isotopic vectors correspond to a particular reactor type.

In order to compare the quality of convergence of the various calculated ratios to the measured ones, we have chosen to analyze the standard deviation of the calculations-measurement ratios. The standard deviation characterizes the variability of the calculation-measurement ratios around their mean value, and the variability can be expected to be minimal for the reactor type corresponding to that of the irradiated sample. An example of the dispersion of the C/M ratios is provided in **Fig. 2** below for a self-generated PWR case. A self-generated PWR case is a numerical test case simulated with a depletion code. In the case where the code used for simulating the evolution of the spent fuel is the same that is used for the inverse method, one speaks of "*inverse-crime*". It could be seen on the figure that the minimal dispersion of these ratios corresponds effectively to the PWR case. In practice, we will consider the standard deviation of the distribution of $\frac{C}{M}$ values and look for the minimal value of $\sigma\left(\frac{C}{M}\right)$. This is what has been done in the following section dealing with experimental cases.

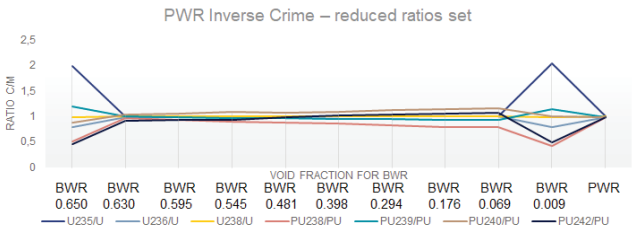

Fig. 2 C/M ratios for determining the reactor type

V. Validation by Predicting Target Parameters on Experimental Cases

We have tested our code with two PWR and two self-generated BWR cases. The final predictions logically correspond to the channel assigned by applying the differentiation criterion seen above.

In order to verify the applicability of the methodology to an unknown irradiated fuel sample, the light water reactor type differentiation method needs to be tested on experimental cases. To this end, two PWR and two BWR reactor cases from the SFCompo 2.0 irradiated fuel database were tested.

The approach described above was applied, and the results of target parameter predictions for the four test cases are presented in **Table 3**, **Table 4**, **Table 5** and **Table 6**. The first column is the reactor type, PWR or BWR. For BWR, the void fraction is given. There are 10 different values of the void fraction, as provided by the CESAR code. The second column gives the standard deviation of the ratio of calculated value versus measured value, $\sigma\left(\frac{C}{M}\right)$, for the set of selected ratios of the uranium and plutonium isotope concentrations. The minimal value we are looking for is highlighted in green in this second column and it then gives the estimated reactor type as well as the values of e_5 and BU. The third column is the predicted initial enrichment in ^{235}U, e_5. The fourth column is the BU. The fifth column is the difference between the calculated and measured values of e_5 in percent. The last column is difference between the calculated and measured values of BU in percent.

For the Three Mile Island case (Table 3), the minimal value of $\sigma\left(\frac{C}{M}\right)$ is 0.0336 corresponding to the PWR simulation.

Table 3 PWR test case – Three Mile Island

Reactor type	$\sigma\left(\frac{C}{M}\right)$	e_5 (%)	BU MWd/ithm	ϵ_{e_5} (%)	ϵ_{BU} (%)
BWR 0.650	0.3591	3.92	83,224	-2.1	81.3
BWR 0.630	0.0500	3.97	54,262	-1.0	18.2
BWR 0.595	0.0500	3.99	43,055	-0.5	-6.2
BWR 0.545	0.0613	4.03	40,615	0.4	-11.5
BWR 0.481	0.0542	4.04	39,702	0.9	-13.5
BWR 0.398	0.0637	4.08	38,974	1.7	-15.1
BWR 0.294	0.0784	4.12	38,323	2.7	-16.5
BWR 0.176	0.0946	4.16	38,278	3.7	-16.6
BWR 0.069	0.1007	4.19	45,716	4.4	-0.4
BWR 0.009	0.3786	4.17	88,742	4.1	93.3
PWR	**0.0336**	**4.11**	**44,362**	**2.5**	**-3.4**

Indeed, the Three Mile Island reactor is a PWR. Our prediction is then than this "unknown" sample correspond to a fuel sample irradiated in a PWR with e_5 = 4.11 % and BU = 44362 MWd/ithm. The measured values of e_5 and BU are 4.01 % and 45,900 MWd/ithm, corresponding to a discrepancy of 2.5 % and -3.4 % in value with the predicted value. For practical purpose of nuclear forensics, these discrepancies are very satisfactory. These discrepancies exist due the imperfection of the simulation tool on the one hand, and on the other hand from the fact that only a selection of few ratios of isotopes concentrations have been used for the inversion method.

For the second PWR test case, Calvert Cliffs, the results are given in Table 4.

Table 4 PWR test case – Calvert Cliffs

Reactor type	$\sigma\left(\frac{C}{M}\right)$	e_5 (%)	BU MWd/ithm	ϵ_{e_5} (%)	ϵ_{BU} (%)
BWR 0.650	0.7374	2.36	161,103	-3.7	332.3
BWR 0.630	0.0399	2.41	51,421	-1.9	38.0
BWR 0.595	0.0497	2.42	40,429	-1.5	8.5
BWR 0.545	0.0549	2.43	37,883	-0.9	1.6
BWR 0.481	0.0649	2.45	36,443	-0.2	-2.2
BWR 0.398	0.0799	2.47	35,057	0.7	-5.9
BWR 0.294	0.0997	2.49	33,599	1.5	-9.8
BWR 0.176	0.1179	2.51	32,532	2.4	-12.7
BWR 0.069	0.1227	2.53	39,163	3.0	5.1
BWR 0.009	0.6697	2.51	82,793	2.2	122.1
PWR	**0.0314**	**2.49**	**40,777**	**1.5**	**9.4**

The minimal value of $\sigma\left(\frac{C}{M}\right)$ is 0.0314 corresponding rightly to the PWR simulation. The predicted value of e_5 and BU are 2.49 % and 40,777 MWd/ithm to be compared to the measured values of 2.45 % and 37,270 MWd/ithm. This corresponds to a discrepancy of 1.5 % and 9.4 % in value with the predicted value. Again, for practical purpose of nuclear forensics, these discrepancies are satisfactory. *A posteriori*, one could point out that there are smaller discrepancies with some BWR calculations. For instance, if one considers the quadratic sum of ϵ_{e_5} and ϵ_{BU} given in %, the PWR case gives 90.61 and the BWR with a void fraction of 0.481 gives 4.88. However, the correct attribution of reactor type is effectively PWR, proving the efficiency of the proposed method. The use of the smallest dispersion of the $\frac{C}{M}$ allows to find the case that provides the best global coherence between measured values and calculated values.

For the first BWR test case, Fukushima-Daini-1, the results are given in Table 5.

Table 5 BWR test case – Fukushima-Daini-1

Reactor type	$\sigma\left(\frac{C}{M}\right)$	e_5 (%)	BU MWd/ithm	ϵ_{e_5} (%)	ϵ_{BU} (%)
BWR 0.650	0.5399	2.93	155,601	-2.2	337.6
BWR 0.630	0.0782	2.98	47,794	-0.7	34.4
BWR 0.595	0.0620	2.99	38,197	-0.3	7.4
BWR 0.545	0.0507	3.01	35,920	0.4	1.0
BWR 0.481	0.0416	3.03	34,683	1.1	-2.5
BWR 0.398	**0.0380**	**3.06**	**33,506**	**1.9**	**-5.8**
BWR 0.294	0.0451	3.09	32,317	2.9	-9.1
BWR 0.176	0.0590	3.11	31,511	3.8	-11.4
BWR 0.069	0.0592	3.14	38,201	4.5	7.4
BWR 0.009	0.5268	3.12	84,360	3.9	138.2
PWR	0.0773	3.08	38,648	2.8	8.7

The MIMCO predicts a BWR irradiation with a void fraction of 0.398. The discrepancies with measured values (e_5 = 3.00 %, BU = 35,560 MWd/ithm) are for e_5 and BU 1.9 % and -5.8 % respectively, which is satisfactory. Experimentally, the void fraction varied during the irradiation between 12 and 27 %. It is thus difficult to comment about the void fraction value of 0.398 given by the MIMCO.

For the second BWR test case, Fukushima-Daini-2, the results are given in Table 6.

Table 6 BWR test case - Fukushima-Daini-2 (void fraction = 0.11)

Reactor type	$\sigma\left(\frac{C}{M}\right)$	e_5 (%)	BU MWd/ithm	ϵ_{e_5} (%)	ϵ_{BU} (%)
BWR 0.650	0.5011	3.58	86,640	-8.4	104.6
BWR 0.630	0.1492	3.64	60,886	-6.9	43.8
BWR 0.595	0.1315	3.66	47,651	-6.4	12.5
BWR 0.545	0.1159	3.70	44,814	-5.5	5.8
BWR 0.481	0.1045	3.71	43,716	-5.1	3.2
BWR 0.398	0.0857	3.74	42,888	-4.3	1.3
BWR 0.294	0.0646	3.78	42,112	-3.4	-0.6
BWR 0.176	0.0474	3.81	42,064	-2.5	-0.7
BWR 0.069	**0.0436**	**3.84**	**49,387**	**-1.8**	**16.6**
BWR 0.009	0.5081	3.82	90,097	-2.4	117.5
PWR	0.1864	3.78	48,956	-3.3	15.6

The MIMCO predicts a BWR irradiation. The discrepancies with measured values (e_5 = 3.91 %, BU = 42,350 MWd/ithm) are for e_5 and BU -1.8 % and 16.6 % respectively, which is satisfactory for e_5 and relatively satisfactory for BU. Experimentally, the void fraction is stable during the irradiation at 0.11. As the CESAR depletion code gives simulation results for 10 fixed void fractions, the MIMCO result should be one of these 10 values. In this Fukushima-Daini-2 case, the minimal value of 0.0436 is obtained for $\sigma\left(\frac{C}{M}\right)$. In fact, the void fraction measured value of 0.11 lies in between the value of 0.176 and 0.069. By noting that e_5 = 3.81 % and BU = 42,064 MWd/ithm for the simulated void fraction of 0.176, one could reasonably assume that a minimal value of $\sigma\left(\frac{C}{M}\right)$ exists between the void fraction of 0.176 ($\sigma\left(\frac{C}{M}\right)$ = 0.0474) and the void fraction of 0.069 ($\sigma\left(\frac{C}{M}\right)$ = 0.0436). In the case of an irradiation with a constant void fraction, the MIMCO not only predicts the correct reactor type but also provides a very good estimation of the void fraction.

Overall, the results obtained for the various test cases are very good, with the irradiation parameters of both PWRs accurately predicted. The reactor type is correctly found, meaning that we are able to distinguish between PWRs and BWRs in case of LWRs. The predicted enrichment does not exceed a relative error of 2.5%, while the burnup prediction is close to 9.4%. Concerning the two BWR cases, the predicted enrichment does not exceed a relative error of 1.9%, while the burnup prediction is below 17%. These results are similar to those obtained with our previous version of the inverse method, which used more marker isotopes. The computational costs of both versions are quite similar (several minutes thanks to the MIMCO and the speed of the CESAR code[7] which is the fast industrial code derived from the reference code, the DARWIN2.3 package for fuel cycle applications.[8] The isotope concentrations or ratios of isotopes concentration obtained by mass spectrometry are the input data for both versions of the inverse method. Since uranium and plutonium concentration ratios with a good accuracy are obtain more rapidly and more easily (few weeks compared to several weeks) than those of fission products, the conclusions of the forensics analysis for an unknown spent nuclear fuel could be given more rapidly with this new inverse method.

VI. Conclusion

The inductive method, studied at CEA as part of nuclear forensics since 2019, is based on solving the inverse problem of irradiated fuel depletion, and assumes that for an analyzed signature, one and only one evolutionary pathway exists. A numerical model is then used to reproduce the depletion of the fuel's isotopic composition. Finally, the inverse problem is solved using an iterative process based on the CESAR code, to trace the initial composition of the fuel and the characteristics of its irradiation using the ordinary least squares method. Compiling the coefficients of the bilinear correlations of several reactor types, a script calculates the initial estimate of the target parameters (enrichment and burnup) for each of them, and always selects a set of markers on which the inverse method will be applied. The method gives good results, allowing the recovery of reactor types from self-generated and real experimental cases in cases of LWRs

The validation of this method could be extended to other reactor types (*e. g.* reactor with low or no enriched fuel, neutron fast spectrum reactor, *etc.*). One could also consider to use this method to predict other physical parameter relevant for nuclear forensics, *i. e.* cooling time or averaged irradiation power.

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