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Specific Application of Burnup Credit for MOX PWR Fuels in the Rotary Dissolver

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In prospect of a Mixed OXide spent fuels processing in the rotary dissolver in COGEMA/La Hague plant, it is interesting to quantify the criticality-safety margins from the burnup credit. Using the current production computer codes and considering a minimal fuel irradiation of 3 200 megawatt-day per ton, this paper shows the impact of burnup credit on industrial parameters such as the permissible concentration in the dissolution solution or the permissible oxide mass in the rotary dissolver. Moreover, the burnup credit is broken down into five sequences in order to quantify the contribution of fissile nuclides decrease and of minor actinides and fission products formation.

The implementation of the burnup credit in the criticality-safety analysis of the rotary dissolver may lead to workable industrial conditions for the particular MOX fuel studied. It can eventually be noticed that minor actinides contribution is negligible and that considering only the six major fission products is sufficient, owing to the weak fuel irradiation contemplated.

KEYWORDS: Burnup credit, spent MOX fuel, rotary dissolver, CRISTAL

1. Introduction

The current criticality-safety analysis regarding the processing of UO_2 spent fuels in the rotary dissolver in COGEMA/La Hague plant, France, only considers the decrease in fissile nuclides due to fuel irradiation in reactor (actinide-only burnup credit).

In the perspective of a Mixed OXide (MOX) spent fuels application, COGEMA and SGN have studied criticality-safety conditions of the rotary dissolver using the entire burnup credit (BUC) method for a particular MOX PWR fuel application. This burnup credit method takes into account:

- the decrease in fissile nuclides and the presence of fission products (FPs) and minor actinides (mAs) due to the fuel irradiation in reactor,
- the decrease in fissile nuclides due to the spent fuel cooling after irradiation.

In this study the BUC is broken down into five sequences thus highlighting their relative importance. These sequences consider the following media:

1. fresh MOX fuel,
2. MOX fuel aged,
3. spent MOX fuel without FPs,
4. spent MOX fuel with 6 FPs,
5. spent MOX fuel with 15 FPs.

There are not yet qualified burnup measurements implemented for MOX spent fuels, but the measurement method already exists. So, only with a qualitative burnup measurement, the MOX fuel irradiation can be guaranteed and consequently we are allowed to take into account a minimum burnup of 3 200 megawatt-day per ton. This value is representative of the 50-least-irradiated-centimeters of assembly rods irradiated during a single irradiation cycle.

Criticality studies about the rotary dissolver are peculiar because of the reference fissile material - an heterogeneous system of oxide pellets in dissolution into a nitrate aqueous solution which is fissile and moderator in the same time. The purpose of these studies is to determine upper safety limits for fissile concentration in the nitrate solution and for oxide mass in each bucket of the rotary dissolver.

The study described in this paper is conducted with the french computer codes currently used for production studies. Restricting hypotheses on fuel burnup and fuel inventory are made. The aim is to prove that using a minimal BUC for the criticality-safety analysis of the rotary dissolver may lead to workable industrial conditions for the particular MOX PWR fuel studied.

2. Reference Fissile Materials

2.1 Main Hypotheses

The reference fissile material in the buckets of the rotary dissolver is an array of MOX pellets (UO_2+PuO_2) moderated by the dissolution solution. The dissolution solution can be:

- An uranium VI and plutonium III nitrate aqueous solution – $UO_2(NO_3)_2 + Pu(NO_3)_3$ – in which the isotopic composition in uranium and plutonium is identical to the pellets' one. The free nitric acidity of the solution is put equal to zero so as to minimize neutron absorption. The fissile concentration in the nitrate solution is maximized up to the upper safety limit in infinite system.
- An aqueous nitric acid solution – HNO_3 – which is the process starting solution. In that case, the nitric acidity of the solution is estimated at the lowest value of 3 N.

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In this study, it was established that the most reactive configuration of the rotary dissolver is always obtained with the uranium and plutonium nitrate solution. Thus, the only results presented in this paper are obtained with the nitrate solution.

Considering that pellets are in dissolution, their radius can be lower than the nominal value. So the pellets' radius is set in order to maximize the dissolver reactivity. Furthermore, the moderator-to-fuel ratio (defined as the ratio between nitrate solution volume and MOX pellets' volume) is optimized. Finally, the pellets' clad is not considered. The initial fissile isotopic composition of the MOX pellets (thus for the nitric solution) is given in the Table 1.

Table 1 initial mass isotopic composition of the studied MOX fuel

^{235}U / U	0.72 %
^{238}U / U	99.28 %
^{239}Pu / Pu	67.69 %
^{240}Pu / Pu	21.02 %
^{241}Pu / Pu	8.62 %
^{242}Pu / Pu	2.67 %
Pu / (U+Pu)	4.05 %

2.2 Burnup Credit Hypotheses

The burnup credit is broken down into the five following sequences:

1. fresh MOX fuel (reference sequence),
2. MOX fuel aged,
3. spent MOX fuel without FPs,
4. spent MOX fuel with 6 FPs,
5. spent MOX fuel with 15 FPs.

For each sequence, nuclides considered in the calculations are given in the Table 2. Major actinides (MAs: U and Pu isotopes and ^{241}Am), minor actinides (mAs) and fission products (FPs) taken into account are those identified for MOX burnup credit calculations.[1)] The six FPs of the sequence 4 are the six most important in term of reactivity worth among the fifteen usual FPs.[1)]

The fuel burnup is reduced to the minimal value of 3 200 megawatt-day per ton – MWd/t. This value is representative of the least irradiated chopped rods (due to the axial profile burnup of fuel assemblies) and needs no previous burnup measurement to be guaranteed (only an irradiation confirmation). This hypothesis about the fuel burnup is very conservative for standard MOX fuels with average fuel assembly burnup between 20 and 40 GWd/t. As the fuel burnup is minimized, the impact of the irradiation conditions simulated in the depletion calculation on nuclides balance is attenuated.

The MOX fuel studied here is supposed to be 15 years old (irradiation and cooling time).

For sequences 2 to 5, the nitrate solution is an uranium VI, plutonium III and americium III nitrate aqueous solution – $UO_2(NO_3)_2 + Pu(NO_3)_3 + {}^{241}Am(NO_3)_3$ – with free FPs for sequence 4 and 5.

Note: For the second sequence, the fuel inventory is not computed by depletion codes but is established from ^{241}Pu half-life which is 14.35 years.

Table 2 nuclides considered in the calculations for each BUC sequence: (a) are nuclides which balance is altered compared with the previous sequence, (b) are nuclides added compared with the previous sequence. Nuclides with the same balance as the previous sequence are not repeated.

sequence		nuclides altered (a) and added (b)
1. initial sequence		U: 235, 238 Pu: 239, 240, 241, 242
2. ^{241}Pu decay	a	Pu: 241
	b	^{241}Am
3. irradiation	a	U: 235, 238 Pu: 239, 240, 241, 242, ^{241}Am
	b	U: 234, 236 Pu: 238 mAs: ^{242m}Am ^{243}Am ^{243}Cm ^{244}Cm ^{245}Cm
4. 6 FPs	b	^{103}Rh, ^{133}Cs, ^{143}Nd, ^{149}Sm, ^{151}Sm, ^{155}Gd
5. 15 FPs	b	^{95}Mo, ^{99}Tc, ^{101}Ru, ^{109}Ag, ^{145}Nd, ^{147}Sm, ^{150}Sm, ^{152}Sm, ^{153}Eu

3. Rotary Dissolver Modeling

The rotary dissolver is made up of a fixed element – the tank – and of a rotary element – the wheel – which comprises twelve buckets. The wheel is partly submerged in the dissolution solution. Each bucket is about 110 liters volume. During the process, only four consecutive buckets contain fuel rods in dissolution. The others are empty or out of the dissolution solution (then the dissolution process is completed).

The rotary dissolver model on the Fig.1 is built on two buckets and a reflection to simulate two other buckets submerged in the dissolution solution. The buckets are equally filled with the array of MOX pellets moderated by the dissolution solution. The filling level depends on the fissile mass per bucket and on the moderator-to-fuel ratio. A reflection by the dissolution solution surrounds the buckets and a reflection by water surrounds the dissolver tank.

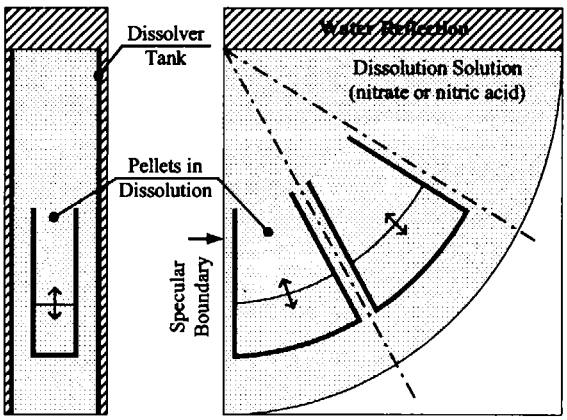

Fig.1 Schema of the rotary dissolver modeling

4. Method

4.1 Computer Codes Used

4.1.1 Depletion Code

The fuel inventory after irradiation is computed by the depletion code CESAR 4.36.[2] This code needs a dedicated macroscopic cross-sections library which shall be created for any given MOX fuel and is thereafter built by the APOLLO1.33 and APOGENE 3.2 computer codes. APOLLO, associated with the "CEA86" cross-section library, computes the neutronic parameters of fissile materials on a two-dimensional mixed rod array. APOGEN then builds the CESAR library from APOLLO results.

This computer codes linking is the current way of depletion calculation for production studies used at SGN.

Two fuel assembly designs are processed. Preliminary calculations show that the impact of the design on fuel inventory is negligible because of the low fuel burnup studied.

4.1.2 Criticality Calculations

The Calculations of the neutron multiplication factor (k_{eff}) are performed with the standard way of the criticality-safety package CRISTAL V0 (CIGALES, APOLLO2 and MORET 4 codes).[3), 4)]

CIGALES 2.0 computes the atomic densities of the fissile mixtures with the density laws. For nitric solutions, an "isopiestic" density law is used.[5)] Then, CIGALES generates the calculation file for APOLLO2 code.

APOLLO2 associated with the "CEA93-V4" cross-section library computes the neutronic parameters – material buckling B_m^2 and k_∞ – of the reference fissile materials and generates, for all fissile and non-fissile materials, 172-group macroscopic cross-sections usable by MORET code. The CEA93 library is based on JEF2.2.[6)]

The MORET 4 code uses cross-sections from APOLLO2 to compute k_{eff} in a three-dimensional geometry through a Monte-Carlo method.

4.2 Calculation Sequence

The preliminary stage of the calculation sequence is the determination of the MOX fuel inventory after an irradiation of 3 200 MWd/t and a cooling time making the MOX fuel 15 years old. Then, the material balance for the 15 FPs used and for absorbing mAs (^{243}Am and ^{244}Cm) is reduced by 30 %. The material balance for fissile mAs (^{242m}Am, ^{243}Cm and ^{245}Cm) is increased by 30 %. These conservative hypotheses, which reduce the ratio non-fissile / fissile nuclides in the reference fissile materials used, are ventured so as to allow a margin for the implemented calculation scheme.

Then, for each of the five BUC sequence described in section 2.2, the calculation scheme is as displayed in the following subsections.

4.2.1 Study of the Nitrate Solution in Infinite System

The first stage of the calculation scheme is the study of the nitrate solution in infinite system. This stage is represented in the Fig.2.

The atomic composition of the nitrate solution excluding FPs and mAs (thus only considering major actinides MAs: U and Pu isotopes and ^{241}Am) is computed by the CIGALES code with an isopiestic density law. CIGALES requirements include the relative composition of U, Pu and ^{241}Am and their initial concentration in the solution. Then, mAs atomic concentrations are added using the mAs / MAs atomic ratio from the depletion calculation. The FPs atomic concentrations are likewise added using the FPs / MAs atomic ratio. In that case, the MAs concentrations provided by CIGALES are not altered.

The infinite multiplication factor (k_∞) of the nitrate solution is computed by APOLLO2 code and is then compared with the safety criterion: $k_\infty \leq 0.93$. Then, this whole stage is repeated with other fissile concentrations in order to find the upper safety concentration by interpolation.

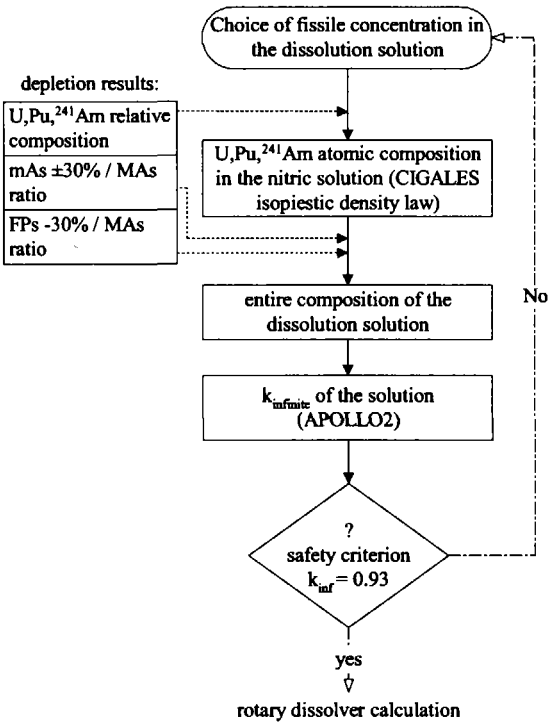

Fig.2 Calculation diagram of the nitrate solution's study

4.2.2 Rotary Dissolver Calculation

The second stage of the calculation sequence is the rotary dissolver calculation. This stage aims at determining:

- the permissible fissile mass in each bucket of the rotary dissolver (mass control mode – MCM),
- the k_{eff} of the rotary dissolver in a geometry control mode – GCM (buckets are full of MOX pellets with an optimized moderator-to-fuel ratio).

In these calculations, the fissile concentration in the nitrate solution is the upper safety concentration from the previous stage rounded up to the superior value. Its composition is computed as presented in the Fig.2. Moreover, the MOX pellets' composition from the depletion calculation is maximized up to a 10.5 density.

For a given fissile mass in the buckets (mass control mode – MCM), the pellets' radius is chosen in order to maximize the dissolver reactivity and the moderator-to-fuel ratio is optimized the same way. The filling level of the buckets is fitted for each value of the fissile mass and of the moderator-to-fuel ratio. The usual permissive criterion for the rotary dissolver in mass control mode is $k_{eff} + 3\sigma \leq 0.97$ (where σ is the standard deviation). The permissible and critical masses are determined by interpolation.

For the geometry control mode – GCM (buckets full of MOX pellets), the pellets' radius is set in order to maximize the dissolver reactivity and the moderator-to-fuel ratio is optimized the same way. Then, each value of the moderator-to-fuel ratio corresponds to a fissile mass per bucket. This calculation is performed only when the fissile mass per bucket in a mass control mode and the optimized moderator-to-fuel ratio associated lead to a filling level near full. The transition from a MCM to a GCM usually happens for an oxide mass per bucket around 170 kg.

5. Results and Analysis

5.1 Infinite System

5.1.1 Nitrate Solution

For each BUC sequence, the upper safety fissile concentration and the critical fissile concentration in the nitrate solution are given in the Fig.3.

It can be noticed that the burnup credit for the dissolution solution is rather significant (from 234 to 309 g/l permissible and from 287 to 400 g/l critical). The main credit comes from the second and the third sequence. Since the FPs are taken into account, the margin is only about 10 g/l for the permissible concentration, due to the restricting hypotheses ventured about irradiation and FPs balance. The concentration credit between the sequences 4 and 5 is lower than 1 g/l, the whole FPs credit then comes from the 6 FPs.

In the case of the spent UO_2 fuels processing, the nominal fissile concentration in the nitrate solution is around 200 g/l and the safety calculations are made with a restricting maximal concentration of 400 g/l. In our case, this concentration is not permissible, even with the entire burnup credit (sequence 5), but it is just critical. Even so the MOX processing is still conceivable because of a permissive concentration around 300 g/l.

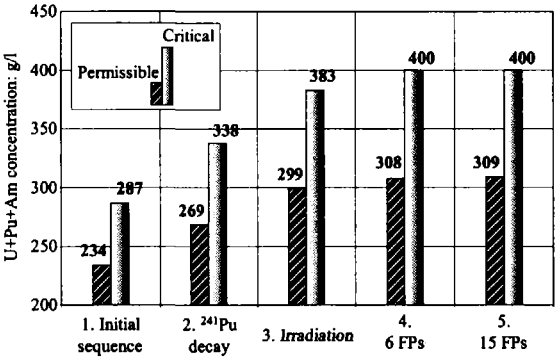

Fig.3 Permissible and critical fissile concentration in the nitrate solution for each BUC sequence.

5.1.2 MOX Pellets in Dissolution

The burnup credit for an infinite system of MOX pellets in dissolution should first be considered, prior to the examination of the rotary dissolver calculation results. The maximum k_∞ (whatever the pellets' radius and the moderator-to-fuel ratio are) of each BUC sequence is computed by the APOLLO2 code. The results on the Table 3 show that the entire burnup credit reaches almost 8 600 pcm. More than half of credit comes from the second sequence (because of the 15 year-old fuel) and only 16 % comes from FPs, of which the main 6 contribute nearly 90 %.

Table 3 Infinite system of MOX pellets in dissolution: cumulated credit and sequence share of each of the five BUC sequences. The maximal value of k_∞ is always reached for a 0.4 cm pellets' radius and a 5.0 moderator-to-fuel ratio.

sequence	k_∞ max.	cumulated credit (pcm)	contribution (%)
1. initial sequence	1.33617	-	-
2. ^{241}Pu decay	1.27085	4 890	57
3. irradiation	1.24063	7 150	26
4. 6 FPs	1.22281	8 480	16
5. 15 FPs	1.22135	8 590	1

5.1.3 Minor Actinides Contribution

Some calculations in infinite system for the dissolution solution and the pellets in dissolution have been made in order to quantify the mAs contribution in the sequences 3 to 5.

With regard to the dissolution solution, the permissible and critical concentrations are not modified if the mAs are not taken into account, which means that the concentration credit of mAs is lower than 1 g/l for the nitrate solution.

Considering the array of pellets in dissolution, the difference in k_∞ does not exceed 20 pcm whether the mAs are taken into account or not (Table 4) and the difference in B_m^2 is around 0.08 %. The reactivity

worth is greatly included in the uncertainty in the value of k_{eff} for the rotary dissolver application.

The mAs contribution to the burnup credit is negligible because of the very low fuel burnup considered and their consequently weak balance in the fuel inventory. Thus, the mAs have not been taken into account for the rotary dissolver application (section 5.2).

Table 4 Infinite system of MOX pellets in dissolution: comparison between a model with minor actinides (a) and a model without minor actinides (b). The maximal value of k_∞ is always reached for a 0.4 cm pellets' radius and a 5.0 moderator-to-fuel ratio.

sequence	k_∞ max.		$\Delta k_\infty/k_\infty$
	(a)	(b)	pcm
3. irradiation	1.24063	1.24088	-20
4. 6 FPs	1.22281	1.22304	-19
5. 15 FPs	1.22135	1.22156	-17

5.2 Rotary Dissolver Application

The application of the burnup credit to the rotary dissolver geometry will be considered in this last section, so as to convert the infinite credit in pcm into a concrete parameter such as the permissible oxide mass in dissolver buckets.

The dissolver calculations' results in a MCM and in a GCM are represented by the curves of the Fig.4. The values of the permissible and critical masses interpolated from these results are given in the Table 5. For these calculations, the uncertainty σ in the value of k_{eff} is less than or equal to 200 pcm.

For the initial sequence (fresh MOX fuel), the permissible oxide mass per bucket is 41 kg and an added mass of 21 kg of oxide per bucket suffices to lead to critical conditions for the dissolver. Such a constraint does not allow a workable industrial application because one fuel assembly should be processed in 15 or more buckets. Then, the answer could be to poison the dissolution solution, with gadolinium for instance.

In the following BUC sequences, the mass credit is quite remarkable. Just from the second sequence, the permissible mass is twice the first sequence's one and the dissolver is sub-critical in GCM. Moreover, when taking into account FPs (sequences 4 and 5) the maximal $k_{eff}+3\sigma$ value in GCM (whatever the oxide mass per bucket is) is slightly lower than the current acceptable criterion. So, the reactivity worth between the sequences 3 and 4 is not sizeable but it allows the transition between a MCM and a GCM in this case study.

It can be noticed that the credit between the sequences 4 and 5 is negligible seeing that it is of the same order as calculations uncertainties. The cross-sections calculation time, by APOLLO2, for pellets in dissolution is quite higher for the sequence 5 than for the sequence 4 because of the number of nuclides processed. Considering the number of calculations needed for one value of k_{eff} (about four pellets' radius and eight moderator-to-fuel ratio for each radius), the sequence 4 study is sufficient.

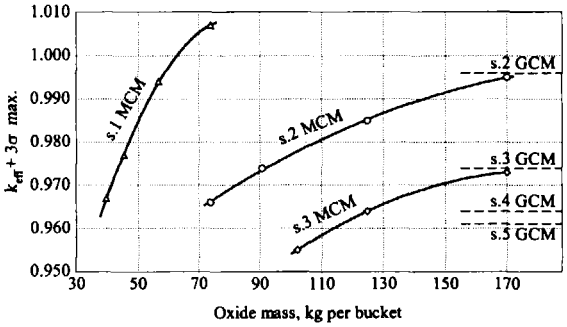

Fig.4 maximal $k_{eff}+3\sigma$ values (whatever pellets' radius and moderator-to-fuel ratio are) for MOX pellets in dissolution into the rotary dissolver. "s.*i*" is relative to the BUC sequence number *i*. MCM and GCM respectively mean "mass control mode" and "geometry control mode".

Table 5 Permissible and critical oxide mass per bucket for a mass control mode – MCM – and maximal value of $(k_{eff}+3\sigma)$ for the dissolver in a geometry control mode – GCM (whatever the oxide mass per bucket is).

sequence	MCM		GCM
	permissible mass, kg	critical mass, kg	$k_{eff}+3\sigma$ max.
1. initial sequence	41	62	>> 1
2. ^{241}Pu decay	82	n/a	0.996
3. irradiation	147	n/a	0.974
4. 6 FPs	n/a	n/a	0.964
5. 15 FPs	n/a	n/a	0.961

6. Conclusion

From a criticality-safety view with the current approach of safety analysis, the industrial processing, in the rotary dissolver in COGEMA/La Hague plant, of the particular spent MOX fuel studied can not be realized without poisoning the solution dissolution. The permissible concentration in the solution is only equal to 234 g/l and the permissible oxide mass per bucket is only equal to 41 kg.

This study points out that the use of the burnup credit method allows a 8 600 pcm margin benefit. These credit corresponds to a permissible fissile concentration in the solution equal to 309 g/l and allows a geometry control mode for the dissolver. These conditions permit to envisage a processing

without poisoning (as it is currently done for UO_2 fuels) or at least permit to benefit by safety margin in a poisoned process. These results are all the more worthwhile because of the restricting hypotheses made about fuel irradiation (minimized to 3 200 MWd/t), FPs balance (minimized by 30 %) and mAs balance (minimized by 30 % for non-fissile mA nuclides and increased by 30 % for fissile mA nuclides).

Considering these restricting hypotheses, it is also shown that taking into account minor actinides is negligible regarding their impact on reactivity. Finally, the breakdown into five sequences of the burnup credit shows that the use of the fifteen FPs is not more advantageous than using only the six more important FPs.

7. References

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