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ANALYTICAL OUT-OF-PILE AND IN-PILE EXPERIMENTS ON GADOLINIA BEARING FUELS

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New fuel management schemes in PWRs can be achieved through the use of burnable poisons like gadolinia bearing fuel rods. However, the introduction of such a design has required a qualification program, which has been performed in collaboration between CEA, FRAGEMA and/or FRAMATOME by specialized teams in CEA facilities. The main scoops of this program concern :
the fabrication process ; the out of pile physical properties determination ; the in pile thermomechanical behaviour and fission product release ; the neutronic studies in view to validate the Computed Gd efficiency and the LBP depletion calculation schemes and to analyse and assess various schemes of core calculations.

1. Manufacturing processes

The design requirements (homogeneous Gd distribution, no pure Gd_2O_3 agglomerates, stoechiometry - O/M = 2, homogeneous structure) have been considered in the investigation of an elaboration process, mainly during the mixing of oxides powders and their sintering.

The processes involve fine powders of UO_2 (dry route elaborated) and Gd_2O_3 (grain diameter : 3 μm) and mixing conditions allowing to break the agglomerated rough materials. Thus, two mixing processes are able to give the required final homogeneity :

- wet ball milling to elaborate samples for out-of-pile and in-pile testing studies
- special dry mixing able to break the agglomerates, giving results very similar to the previous ones, and choosen by FRAGEMA for industrial application because of its very easy utilization

During the sintering, the partial pressure of oxygen is adjusted by means of a $H2/H_2O$ mixture, in order to obtain the required stoechiometry of the samples.

2. Physical properties

The samples used for measurements were (U-Gd)O_2 solid solutions containing 4, 8 or 12 wt % Gd_2O_3. They were prepared by wet ball milling and sintered at 1700°C and their stoechiometry has been controlled.

2.1. Specific heat

In the range 300 - 1000 K, for 8 wt% Gd_2O_3, we got :

$$C_p\ (J.kg^{-1}.K^{-1}) = 337.7 - 1.823\ 10^{-2} T - 0.997\ 10^{7}\ T^{-2}$$

The obtained values are greater by 9% at 500 K and 7% at 1000 K than the UO_2 values.

2.2. Melting point

In the range 0-12 wt% Gd_2O_3, the liquidus and solidus curves are very closed. The melting point of gadolineated oxides was not found to be lowered as compared to the UO_2 one. We found $T_m = 3115 \pm 30$ K

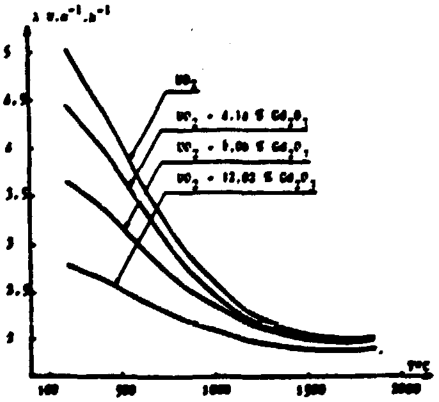
2.3. Thermal conductivity

Measurements were performed either through thermal diffusivity (200 to 1800°C - 0, 4, 8, 12 wt% Gd_2O_3) or radial heat flow (200 to 1100°C, 0 and 8 wt% Gd_2O_3). The two methods gave quite similar results. The lowering of thermal conductivity due to Gd (fig. 1 and table 1) appears to be less important than found by Fukushima et al /1/

TEMP. (°C)	WEIGHT % Gd_2O_3		
	4,14	8,06	12,02
200	0,85	0,73	0,55
400	0,91	0,78	0,61
600	0,93	0,81	0,67
800	0,95	0,86	0,74
1000	0,96	0,90	0,81
1200	0,96	0,95	0,86
1400	0,97	0,96	0,90
1600	0,99	0,97	0,93
1800	0,99	0,97	0,93
O/M	2,001	2,001	1,991

(UGd)O_2 versus UO_2 thermal conductivity as a fonction of temperature

Table I



Thermal conductivity of UO_2 and (UGd)O_2 as a function of temperature

Fig. 1

3. Analysis of the thermomechanical behaviour under irradiation

3.1. Analytical Experiments GDGRIF 1 and GDGRIF 2

This analytical irradiation program is conducted in the "SILOE"Pool reactor in GRENOBLE in order to analyse the behaviour of Gd poisoned UO_2 in PWRs. It includes two irradiation experiments GDGRIF 1 and GDGRIF 2 with respectively 5 and 8 wt% Gd_2O_3. The parameters monitored during irradiation are :

- the power generated by the fuel
- centerline temperature
- stable and radioactive fission gas release

Trends in these parameters are directly compared with those encountered in a UO_2 fuel rod of the same U 235 enrichment (respectively 1.5 and 1.8% for GDGRIF 1 and GDGRIF 2) which is simultaneously irradiated in the same loop. Thus, each experiment includes the irradiation of two superposed instrumented short length rods in a pressurized water loop named "GRIFFON" /2/.

The GDGRIF 1 irradiation, which was performed under an incident neutron flux of $\simeq 1.10^{14} n.cm^{-2}.s^{-1}$ during $\simeq$ 3.3 Siloe cycles, was deliberately stopped just before the burn out of Gd 155 and Gd 157 ; over that period, the fission power generated in the rod varied from 6 up to 20.5 $kW.m^{-1}$ and the overall power level (including heating) from 9.5 to 24 $kW.m^{-1}$. The GDGRIF 2 irradiation lasted 7.5 cycles under the same flux, leading to the complete burnout of active Gd within about 6 cycles ; the fission power inside the rod varied from 5.5 to 29.9 $kW.m^{-1}$ (overall power from 9.1 to 33.5 $kW.m^{-1}$). The GDGRIF 1 post irradiation examinations are completed ; they are under way for GDGRIF 2. Here are the main results (more details in /2/).

- in pile thermal conductivity of poisoned UO_2 fuel.
  With 5 wt% Gd_2O_3, the ratio $R = \lambda(UGdO_2) / \lambda UO_2$ varies from 0.77 to 0.92 while the average fuel temperature T varies from 470 to 723°C. With 8 wt% Gd_2O_3, this ratio varies from 0.58 ($T = 500°C$) to 1 ($T=1000°C$). These results are in fairly good agreement with out-of-pile results (cf § 2), considering the precision of both methods ; further, they show that for usual in pile irradiation conditions (T_C 950 to 1000°C), the centerline temperature of Gd rods is very close to the one of pure UO_2 fuel rods at the same power level

- fission gas release : fission gases were sampled twice a cycle by He sweeping in both experiments ; the results are quite identical with those obtained in pure UO_2 fuel rods irradiated in the same conditions /3/.

3.2. Overall experience through post irradiation examination on CAP and BR3 fuel rods /2/

Irradiation of UO_2 Gd_2O_3 test rods was initiated by CEA and FRAGEMA about ten years ago in the experimental PWRs CAP in Cadarache (France) and BR3 in Mol (Belgium). Fuel rod geometry was 17 x 17 and gadolinia content was 3 wt%. Two gadolinia dispersion forms were used :

- homogeneously mixed oxide,
- scattered particles

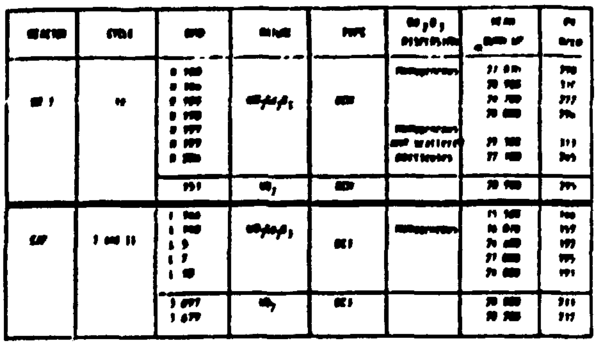
And two sintering processes were implemented : Normal Double Cycle (NDC) and Inverse Double Cycle (IDC) (See TABLE II).

The irradiation conditions in the CAP reactor were similar to those in the 17 x 17 commercial reactors : linear heat generation rate up to 20 kW/m (25 kW/m local). A first group of rods was irradiated up to 15-16 GWd/tU (local burnup of 25 GWd/tU) and a second group up to 25-29 GWd/tU (local burnup of 37 GWd/tU).

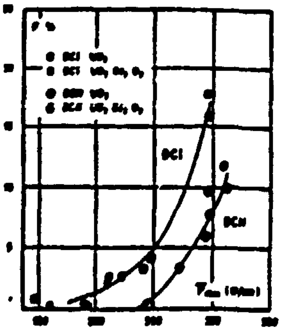
In the BR3 reactor the conditions were more severe : linear heat generation rate up to 31 kW/m (37 kW/m local) and burnup ranging from 22 to 29 GWd/tU (local burnup to 39 GWd/tU).

Post irradiation hot cell examinations were then performed. The main results and conclusions of these investigations are presented below.

Concerning the fuel stack length variation ; the hydrostatic density evolution ; the aspect and composition of the fuel cladding contact zones ; the internal oxidation layer ; the fission gas release (fig.2) and the solid fission product distribution, no significant difference was observed between UO_2 Gd_2O_3 and pure UO_2 fuels in the range of irradiation parameters for the two types of Gd fuel studied.



PIE of Gd Poisoned Rods
(3 wt % Gd_2O_3)
Table II



Fission gas release

Fig. 2

For the Cs 137 behaviour we notice a greater retention in the poisoned fuel than in "pure" UO_2.

Concerning the microstructure and for similar conditions, the Gd fuel appears colder than pure UO_2 despite its presumably lower thermal conductivity and the UO_2 Gd_2O_3 grain size is smaller than that of unpoisoned fuel after irradiation.

As for the post irradiation gadolinium distribution : on the microdispersed product, whether obtained by the NDC or IDC process, and whether irradiated at normal power or at high power, no radial gadolinium redistribution was observed during examinations by the Castaing X-microprobe. As for the macromasses, although the fuel was subjected to high power levels, the gadolinea spheres retained their integrity. A gadolinium diffusion zone around the spheres was simply observed in the hot central zone of the fuel, extending beyond the width of the zone after fabrication (from 30/50 to 60/80 microns).

4. Neutronics studies

Several experiments have been carried out in the experimental reactors MINERVE and EOLE at CADARACHE and MELUSINE in GRENOBLE.
An exhaustive study has been performed by sample oscillation in MINERVE /4/ in order to measure the variation on poison worth according to different parameters :

- Gadolinium content and dispersion (homogeneous dispersion or Gd_2O_3 particle sizes from 10 μm up to 400 μm in UO_2-Gd_2O_3 and Al_2O_3 Gd_2O_3 samples) ; nature of the support (UO_2, Al_2O_3) ; U235 enrichment ($UO_2Gd_2O_3$ samples with increasing 235 U enrichment).

The CAMELEON critical experiment performed in EOLE /5, 6, 7/ gave extensive information about the flux level in absorbers, BOL efficiency of Gd and BOL power distributions in a poisoned fuel assembly. Different realistic designs of Gd poisoned fuel assembly have also been studied in CAMELEON.

The GEDEON irradiations in MELUSINE /8,9/ were conceived in order to check Gd depletion and power peak calculations during fuel burn up. The GEDEON I experiment loaded with a 5 wt % $Gd_2O_3/Gd_2O_3+UO_2$ oxide has been irradiated up to 9 GWd/t.

The GEDEON II, loaded with 8 wt % Gd_2O_3 was started up in May 1985 and will last about 2 years to reach a burn up of 13 GWd/T.
The interpretation of these various experiments allowed us to separate calculation pattern qualification and Gd cross-section improvements.

Fundamental experiments, such as Gd samples oscillations and the CRISTO III experiment /10/ (soluble Gd worth in LWR lattices), enabled us to validate Gd nuclear data within ± 1 % and ± 5 % uncertainty, respectively for thermal cross-sections and resonance integral.

The CAMELEON and the GEDEON experiments enabled us to improve our Gd depletion pattern down to + 1% on odd Gd isotopes. Sensitivity studies /11/ determined the corresponding uncertainty on the computed reactivity worth of $UO_2Gd_2O_3$ during all the cycle : + 2 % of the total Gd initial poisoning. Thus, cycle length calculations, related to Gd poisoned PWR cores, were checked within 1 % accuracy. On the other hand, the computed radial power maps are in agreement with gamma spectrometry distribution /12/ ; the potential power peak in the UO_2-Gd_2O_3, arising at the end of the first cycle, was checked within + 3 % /11/.

5. Analysis of various schemes of core calculations

Fuel management studies have shown the interest for fresh assemblies containing up to 16 burnable poison-rods of UO_2-Gd_2O_3. These rods may be located inside the fresh assembly or at the periphery. The peripheral situation is interesting for macroscopic power map in the core but leads to complexity to calculate the cross section of the different cells because of the surroundings of the burnable poison which are various : fresh or burned fuels at different irradiations. For the sake of simplicity, studies have been carried out with a four assemblies pattern where 2 fresh assemblies contain poison rods and the other two have reached a 11 GWd/t burn up (fig.3). Several types of transport calculation have been considered with APOLLO and followed by diffusion calculation in the pattern. Transport calculations include :

- usual multicell calculation with an average neutron exchange between cells on each type of assemblies (ROTH)
- more sophisticated multicell calculation with neutron exchange side by side, and no cylindrisation of cell on each type of assemblies or directly on the pattern (EURYDICE option and ROTH4)
- cell calculation with extra-region

In the case of an isolated poisoned assembly, the multicell calculations are not adequate for symmetry conditions on poison rods ; so it is necessary to permute the two peripheral rows of fuel rods to improve the results.

The full core analysis is presented in reference /13/.

The following table shows the main results concerning comparison on reactivity and power peaking.

Type of transport calculation	Detailed multicell EURYDICE on the pattern Ref. calculation	EURYDICE on the pattern limited description of flux zone	Multicell [illegible] on each assembly		MULTICELL EURYDICE [illegible] on each assembly with permutation	CELL - EXTRA-REGION
			without permutation	with permutation		
Reactivity pcm	0	- 31	- 336	- 203	- 128	+ 427
$\frac{\Delta P}{P}$ max % Gd rods	0	- 3,3	+ 8,5	+ 0,6	- 0,6	+ 2,6
$\frac{\Delta P}{P}$ max % rods of fuel poison assembly	0	- 1,7	- 4,3	- 8	- 3	- 4,3
$\frac{\Delta P}{P}$ max % assembly without Gd rods	0	- 3,2	+ 1,6	- 2,5	- 2	+ 5,1

Table 3

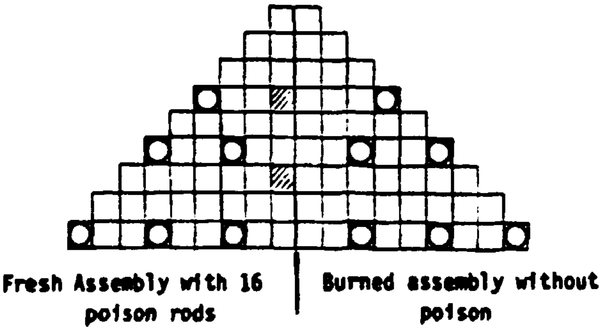


FIGURE 3

Conclusions

All the above studies show that :

- The introduction of Gd_2O_3 as a burnable poison do not affect significantly the overall behaviour of the PWRs' fuel under irradiation (at least up to 8 wt% Gd_2O_3)

- Neutronics studies and analysis of various schemes of core calculations with the French computer codes system allow to predict the neutronic state of poisoned Gd Cores in different configurations and its evolution.

- A proved fabrication procedure is available, and is used for industrial production. At the end of 1986, FRAGEMA will have built, for domestic and foreign market 212 Gd_2O_3 bearing fuel assemblies (i.e. 4.0 metric tons of UO_2-Gd_2O_3 pellets) ; 156 of them are now under irradiation with perfect behaviour.

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