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TITRE (en français) : ERBIUM : POISON ALTERNATIF ? ADDITIF DE STABILISATION ? QUEL DEVENIR ?

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RESUME (en français) : L'erbium a été proposé très tôt comme poison alternatif au gadolinium. L'intérêt potentiel de ce poison par rapport au gadolinium est qu'il présente une section efficace d'absorption dans le domaine thermique relativement faible (^{167}Er) et une intégrale de résonance non négligeable qui conduisent à une cinétique de consommation relativement lente plutôt adaptée aux cycles longs voire très longs.
Les modes d'empoisonnement spécifiques à l'erbium montrent en général une pénalité résiduelle supérieure à celle du gadolinium. Ces pénalités proviennent de résultats de calculs et de bibliothèques sans qu'il y ait confirmation formelle par des résultats expérimentaux.
Une revue des connaissances acquises sur Er depuis les années 1960 jusqu'à la période actuelle est présentée.
Les nouvelles potentialités pour le contrôle d'une part et pour la stabilisation de certains composés comme la zircone stabilisée par Er pour les combustibles en solution solide d'autre part conduisent à la mise en place d'expériences de validation et de qualification mais aussi génèrent de nouvelles possibilités d'empoisonnement par la mise au point d'alliages de Zr/Er.

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ERBIUM : Alternative poison? Stabilisation additive . What future ?

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Abstract

Erbium was proposed as alternative poison to gadolinium at a very early stage. The potential interest of this poison compared to gadolinium is that it presents a relatively low (^{167}Er) absorption cross section in the thermal range and a non-negligible resonance integral that lead to a relatively slow consumption kinetic rather adapted to long or even very long cycles.
The poisoning mode adapted to this poison, homogeneous in low concentration (< 3 %), does not downgrade the power distribution, on the one hand, as the absorption is low and spatially homogeneous, and the thermal conductivity, on the other hand, as the addition in the fuel oxide is in low quantity. A review of knowledge acquired as regards Er, from the 1960s to now, is presented.

1. Introduction

When the problem concerning the extension of the cycle length in water reactors arose, the control corollary of the potential reactivity in excess immediately emerged. Then, the use of the MOX fuel also transferred the considerations to the way of controlling the reactivity, to the extent that the significant reduction in the efficiency of traditional control means such as soluble or solid boron and gadolinium is explained by the fact that the neutron spectrum is harder than with a core using a UO_2 fuel. Finally, recent developments of fuel in inert matrix designed to achieve a better use of the plutonium in water reactors, either in a form designed for multirecycling, or in the form of a «Rock like» type fuel (or ROX) intended to achieve « Once Through» cycles, require the use of additional control means, on the one hand, to the extent that the absence of a conversion on ^{238}U (inert matrix) imposes a faster depletion slope for the fuel, therefore a higher original potential reactivity than a standard fuel with ^{238}U cores at equal cycle length. On the other hand, they require the use of stabilisation additives to obtain a perfectly stabilised Zirconia. Erbium, in the oxide form, often appears in the literature and research teams are focusing on it to achieve these various control and stabilisation functions.

2. Erbium history

As of 1970, Radkowski published articles highlighting the role of erbium as poison in LWR.

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However, it is only late in the 1980s (1988-1989) that the Combustion Engineering (CE) firm conducted numerous studies intended to promote erbium as an alternative burnable absorber to gadolinium and became the market promoter of this solution to reactivity control.Swiftly, the light was shed on the major neutron properties of erbium, and some integral experiments in the TRIGA reactor were conducted to validate the initial validity and the values of the moderator coefficient.
As of 1990, code cross-comparisons took place between CE and the Stüdsvik center (ABB) and significant differences appeared as regards the integral values, and of 2% on cycle lenght.
In 1991, measurement campaigns were undertaken in the "Reactor Critical Facility" (RCF) at the Rensselear Polytechnic Institute on a lattice of 236 UO_2 rods enriched at 3.6 % in ^{235}U, containing 56 rods poisoned at 1.5 % erbium. These experiments as a whole and the associated interpretations gave rise to calculation/experiment differences that gave CE the guarantee it had both the codes and associated libraries to obtain industrial level accuracy.

3. Erbium study at the CEA - First stage

In 1991, in order to reduce problems linked to the presence of hot points and to the bad thermal conductivity of the UO_2 gadolinium high content mixture to envisage the extension of cycles to 18 months, a first evaluation of erbium's potential took place. Early in 1992, a thesis was launched jointly with FRAGEMA [1].
Rapidly, the need to obtain a library of self shielded cross sections arose and a first library was created grounded on an OECD database from 1973. At the time, neither a validation nor a qualification was associated to this elementary database.
Work was conducted and the results compared to those available in the international literature. Generally, comparisons showed a good convergence. However, no comparison is possible to accurately validate the value of erbium's initial efficiency; as isotopes are missing in the evolution chains that were built in a hurry and when CE asserts residual penalties of the same order of magnitude as for gadolinium, CEA calculations led to a factor 2 in disfavour of erbium. Concurrently, studies [2] were undertaken to develop manufacturing processes ensuring a homogeneous distribution, as well as techniques for the sintering of erbium pellets in accordance with the PWR manufacturing specifications [3, 4].
Finally, in 1996, work to develop a coherent library (as of JEF2) was finalised. This completed the first stage of work on this body.

4. Erbium from the neutron standpoint

The following table (Table 1) lists the main neutron properties of erbium that has the particularity of having a single absorbing isotope in the thermal range and with a rather high resonance integral. In fact, the evolution chain is rather short and is simplified due to low absorption rates and to low proportions of the various isotopes (figure 1).
The fundamental difference with gadolinium is that the poison's efficiency according to the content and number of poisoned rods is nearly linear. and that when cycle length effect(s) and initial efficiency were clearly separated by gadolinium, with erbium, the poison may be selected according to two parameters and the lowest possible content can be selected, or in any case, the contents that will not downgrade the thermal conductivity.
. Erbium does not downgrade the power tray (low thermal cross section). The moderator coefficient is more negative, therefore, a higher initial soluble boron concentration can be used.

Table 1
Erbium main neutron properties

Z	A	σa (barn)	I_R (barn)	% isot.
	162	19 ± 2	480 ± 50	0.14
	164	13 ± 2	105 ± 10	1.6
	166	40 3.± 4	100 ± 20	33.4
^{68}Er	167	**670 ± 30**	**2970 ± 70**	22.9
	168	1.95 ± 0.05	36 ± 7	27.0
	170	5.7 ± 0.2	20 ± 2	15.0
	171(*)	280 ± 30		

* ^{171}Er → ^{171}Tm (σa = 4.5 ± 0.2 barn, IR = 118 ± 6 barn)

The moderator coefficient is more negative, therefore, a higher initial soluble boron concentration can be used.

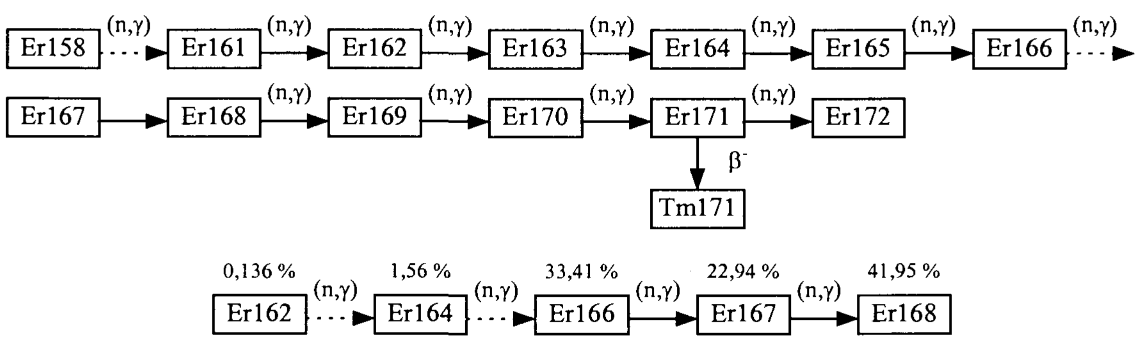

Fig 1. Erbium depletion chain and simplified chain

5. Studies at the CEA - Second stage

In 1996, a reflection followed by a series of experiments concerning the CERMETs were conducted, with the TANOX CCE irradiation in the TANOX device (figure 2) with composite fuels Mo 80 % UO_2 20 % (U5 40 % + 2 % Er_2O_3) that reached 130 GWd/t.
The main results showed [5] that, amongst others, the introduction of erbium did not modify the fuel behaviour, that the thermal conductivity is effectively that planned by a Maxwell Euken law.
Initial efficiency measurements of the natural erbium were conducted in the EOLE reactor both in a UOX and in a MOX lattice. The detailed results [6] presented in an compagnon paper show a good coherency between the calculation and the experiment, that makes it possible to qualify the efficiency of erbium at least at time 0 for an industrial certification. To prevent manufacturing problems linked to the pollution of the UOX and MOX manufacturing lines, studies were launched to integrate quantities of erbium neighbouring 2 % in weight in the rod claddings.
Studies and developments led to the registration of the manufacturing patent [7] that defines a poisoned cladding with the characteristics required for a use in PWR and that represents a major control improvement, that notably makes it possible to reduce the quantity of soluble boron.

From another standpoint, this technique may be very useful in the scope of CERMET composites where the poison may be introduced in the metallic matrix.

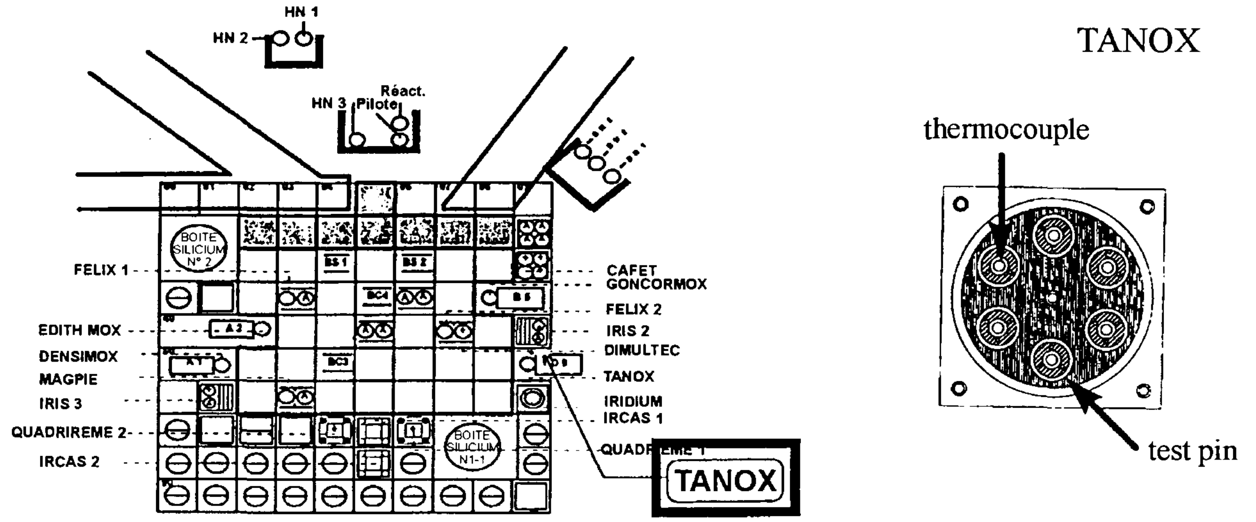

Fig. 2. TANOX device and in SILOE reactor location

Finally, erbium also seems very interesting as compound stabiliser (Fully Stabilized Zirconia) when quantities to ensure the thermal dynamic stability of $(Zr,PuO)_2$ [8] are similar to the quantities required for the neutron control. Therefore, this gives us the opportunity to kill two birds with one stone, by ensuring both the compound's stability, and the compound's neutron control.

6. Conclusion

The sensitivity studies presented in [9] show that there are still problems at erbium library level and that the generated uncertainties are penalising for the industrial future of these poisoning methods. Although the initial anti-reactivity values are correctly calculated at time 0 in UOX lattice and in MOX lattice, a major doubt remains concerning the consumption kinetics and in particular the resonating sections of ^{166}Er. These doubts and inconsistencies must absolutely be dealt with and analysed to achieve the industrial qualification of this body so that it can fulfil the major role expected from it in the field of compound fuels.

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