SOME SPECIFIC ASPECTS OF HOMOGENEOUS Am AND Np BASED FUELS TRANSMUTATION THROUGH THE OUTCOMES OF THE SUPERFACT EXPERIMENT IN PHENIX FAST REACTOR

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ABSTRACT

In a private communication[1], wc reported the results of non-destructive and destructive examination of the SUPERFACT 1 experiment carried out by both the TransUranium Institut (TUI) and the french Commissariats à l'Energie Atomique (CEA). This experiment aimed to study, the behaviour of fuels made up with Neptunium or Americium (from 2 Weight'% up to 45 wt% part of heavy atoms) under irradiation in the Phenix french fast reactor (FR), Post test examinations, jointly performed by the CEA and TUI, allowed to position this behaviour with the standard oxide fuel reference[2].

This paper first reviews this experiment examination main results. Then, it puts the real interest of the FR forward for high rate transmutation of actinides and also some of their limitations.

I. FUEL ELEMENT CHARACTERISTICS

The test section of tbe Superfact experiment was a standard experimental cluster used in PHENIX that holded 19 pins among with 8 were named "actinide pins" due to their particular fuels enriched in americium (Am) or neptunium (Np). The external geometry of the pins is a Phenix standard type. In any cases, the cladding is 15-15 Ti CW stainless steel with a standard PHENIX design. Fuel pellets and pins and cluster were manufactured together by CEA and TU1[2].

The fuel of the so-called homogeneous mode actinide pins is a mixture of the different actinide oxides. Two types of actinide pins were completed, namely the high or low actinide concentration pins according to their americium or neptunium concentration respectivel y in an u ranium or uranium-plutonium oxide fuel. Table 1 shows the initial weight composition of the different actinide pins (standard Phenix oxide fuel is also given). The low concentration actinide pins are characterized by a 2 wt% americium or neptunium content whereas the high ones present a 40 wt% Np or an intermixture of 20 wt% Am and 20 wt% Np content.

The 2 wt% actinide concentration was considered as too low to significantly alter the thermal and thermodynamic properties of the low actinide pins compared to standard oxide ones[3]. Concerning fuels with high actinide concentration, the thermal conductivity of the neptunium fuel is not very far from a standard $(UPu)O_2$ onc[4]. Chiefly duc to the americium, the 20 wt% Np - 20 wt% Arn fuel exhibits a smaller thermal conductivity that is around what could be awaited for an U,PuO_2 fuel *with* a 1.93 low O/M ratio[4]. Thermodynamic properties are mainly ruled by the oxygen potential. For a same O/M ratio, mostly the americium presence involves higher oxygen potential than a standard $(UPu)O_2$ fuel. The *oxygen* potential of the above dcscribcd 20% Np - 20'% Arn fuel (0/M - 1.93), assessed thanks to the reference[5] is about -575 kJ/mole at 1200 K, i.e. the value for a standard oxide fuel $(UPu)O_2$ with a 1.98 O/M ratio. Such a low O/M ratio was pursued to come to an oxygen potential very close to an ordinary oxide one.

Table 1- Composition of homogeneous and frcsb Np and Arn based fuels (pellet diameter = 5,42 mm, height= 7 mm).

pin No *and type*	composition (as % of weight for actinides) required	measured after fabrication
8 and 16 *standard*	$(U_{0.75}Pu_{0.25})O_{2-x}$	$(^{\circ}0.718\ Pu_{0.282})O_{1.983}$
7 and 13 *2% Np*	$(^{\circ}0.74^{\circ}0.24^{\circ}p0.02)\ ^{\circ}2-x$	$(U_{0.741}Pu_{0.244}Np_{0.0150})01.973$
4 and 16 *2% Am*	$(U_{0.74}Pu_{0.24}Am_{0.02})O_{2-x}$	$(U_{0.745}Pu_{0.237}Am_{0.0184})O_{1.957}$
5 and 15 *45% Np*	$(U_{0.55}Np_{0.45})O_{2-x}$	$(^{\circ}0.552\ Np_{0.4482})O_{1.996}$
6 and 14 *20%Np +20%Am*	$(U_{0.60}Np_{0.20}Am_{0.20})\ O_{2-x}$	$(U_{0.596}Np_{0.2118}\ Am_{0.1918})O_{1.926}$

11. IRRADIATION CONDITIONS

SUPERFACT 1 was irradiated in PHENIX from the 38111 up to the 42th run (October 86 to January 88) in the same position in the most outer position (4th crown) of the internal core of the reactor. Maximum bum ups of the actinide pins were assessed by calculation to be about 6.8 %at for the low actinide concentration pins and around 4,5 %at for the high ones[6]. Rcccnt results[7] issued from chemical analysis (Nd 148) show a good agreement with the reckoned values (table 2),

In order to give an idea of the irradiation history of SUPERFACT, table 3 shows the i rradiation parameter evolution for the standard oxide pins. With the exception of a 2/3 level of

power, 77 EFPD long, at mid life during the 39th run, SUPERFACT ran across a full power irradiation. The standard oxide pins reached a bum up of 8.5 %at within 383 EFPD. The linear power rate continuously decreased from 430 W/cm at beginning of life to 380 W/cm at the end.

Table 2- Irradiation conditions in Phenix.

Pin No	Type	Burn-up (at%) CEA calculation	Burn-up (at%) ITU measurement	Linear power (W/cm) CEA calculation beginning & end of tife	
8 and 16	standard	8.5	/	430	370
7 and 13	2% Np	6.8	6.4	380	325
4 and 16	2% Am	6.8	6.4	380	325
5 and 15	45% Np	4.6	4.5	206	283
6 and 14	20% Np / +20% Am	4.3	6.5	174	273

Let us recall that irradiation conditions for actinide pins were obviously different and conceived in such a way that power evolutions had to observe the reactor safety rules. At the stage of the experiment design, the power progress were forecast[8] as reported in table 2. Contrary to a standard oxide pin, the linear power rate of the pins with high actinide concentration rose continuously during irradiation. That particular behaviour was expected since the elimination of actinidcs produces significant amounts of ncw fissile elements, principally plutonium.

Table 3- Irradiation conditions in Phenix (internal core 4th crown*) for standard oxide pins (n" 8 and 16)

date	cycle	total EFPD	total dpa NRT	total at %	Phenix 'we'	linear power (W/cm)	max temp. (°C)
october 1986	38	76.5	11.7	1.82	Pn	429	631
	39	87.3	13.4	2.07	Pn	427	630
	39	164.4	25.2	3.83	2/3 Pn	/	/
	40	219.5	33.6	5.06	Pn	420	620
	41	290.1	44.4	6.56	Pn	388	603
january 1988	42	383.4	58.9	8.51	Pn	383	600

* calculated flux: 5.8 x 10^{15} n/cm².s

HI. POST IRRADIATION EXAMINATIONS

Pins were distributed equally among the two partners of this experiment and 4 actinide pins were shipped from Phenix FR to the hot cells of TUI and CEA for post irradiation examination.

III.1. Non Destructive Examination

The non destructive examination does not give any cluc of an abnormal behaviour of the actinide pins compared to the standard oxide sibling pins irradiated in the same test cluster[9].

Profilometry measurements display insignificant clad deformation with the exception of the pins containing americium that present some hints of a possible beginning of a fuel clad mechanical interaction. However, this tendancy is supported only by a tiny deformation and will have to be checked *on* **higher burn up pins.**

Gammascannings evidence that fission products such as cesium 137 and 134 are axially displaced in the fuel column. Yet, both the intcusity of this movements and the accumulation axial locations do not digress from one could expected in a standard oxide pin irradiated under the same conditions.

111.2. Destructive Examination

Fission gases xenon and krypton released by fuels during irradiation and gauged thanks pin piercing and mass spectrometric analysis present, whatever the nature of the actinide pin considered, roughly the same average value of about 60 % of the total xenon and krypton fission yield. This release rate is in rather good agreement with what could be expected from a standard oxide pin in the same conditions of irradiation. Unlike a standard oxide pin, the actinide ones with fuel containing americium reveal a large amount of helium within the release fission gases. The amount of helium measured in the 2% Am pin is already roughly four times greater than what can be awaited using the standard oxide fuel production rate, i.e. assuming a total helium relcasc. This ratio increases up to 60 times for the 20'%. Am -20 Np% pin whereas it remains only twice the theoretical production in the 45'% Np pin. This unanticipated helium presence is noticed as well within the in-fuel retained gases. Gas retention measurements through a fuel vaporization at high temperature under vacuum indicate that up to 660 mm^3 He NTP/g_{fuel} arc retained in the fuel of the 20% Am -20 % Np pin. This value has to be compared to the typical few tenth mm^3 He NTP/g_{fuel} recorded in a standard oxide pin fuel. The average value for the 45% Np pin is "only" 130 mm^3 He NTP/g_{fuel}. The likely origin of this helium is discussed hereafter.

Metallographics performed at peak power node bring to light in a certain extent the thermal level experienced by fuel during irradiation through features such as central hole and the fuel restructured aspect. The macrographs of the 45% Np pin and the 20°/0 Am - 200/. Np pin (figure la) display ahnost the same. lack of restructuration with no central hole. This is to be related to the same low linear power, those pins encountered during irradiation, The by far more porous aspect of the 20'% Am - 20% Np pin is presumably connected to the huge helium production seen above, The higher linear power met by the 2'%. Np (figure 2a) or Am pins is revealed by the presence of a central hole and some columnar grains around. Insofar as the central hole and columnar grains diameters, one can measure on a standard oxide pin withstanding the same condition of irradiation, would be very close. it is thus confirmed that a 2% Am or Np addition does not lead to a notable different behaviour.

Fuel Clad Chemical Interactions (FCCI) between actinide based fuels and the cladding (15- 15 Ti CW) were observed for the 2% Np or 2% Am up to 50 microns deep (figure 2b) and the 20'?4. Am -20 '% Np pin (figure lb) up to 60 microns deep[10]. No trace was detected on the highest actinidc content pins (45% Np). No correlation can therefore be drawn out neither

with the actinide nature nor with its concentration inside the fuel. This corrosion is characterized by an irregular radial occurence. The corrosion aspect is not looking like the current features for an oxide pin FFCI. In particular, the 60 microns deep corrosion observed on the 20°/0 Am -20 ·/· Np pin is purely intergranular and the 40 μm deep corrosion for the 2°/0 Np pin presents a dense volumic aspect.

Microprobe analysis[11] (MPA) show a radial flat distribution of the actinidcs in the pellets, whatever the actinidc nature or concentration. Figures 3 evidence such an even radial distribution in several cases. The radial profiles of actinidcs in the 20'%. Am - 20'%. Np fuel appear less smooth (figures 3C to 3e) and in addition to that, some variations are seen inside the same sample on the two radii reported (figures 3d and 3e). None of these radii present an appropriate shape in terms of nuclear cross sect ion explanation (a drop towards the pellet center). Now, considering on one hand that the figure 3f, we set up by averaging each radius, shows a fairly flat profile and on the other hand that all the other Np profiles are still flat particularly the initial 45 wt% one (fig. 3h), we thus tend to conclude to a possible inhomogeneous intermixture during fabrication of the fuel pellets. The particularly porous aspect of the 20% Am - 20% Np fuel sample adjoined to the well known sensitivity of the MPA measurements to the porosity could bring an explanation. Now, the fact that the tendancy are recorded on several points with a rather low dispersion for each point makes that hypothesis doubfull. The mean Np content radially weighted matches pretty well the chemical analysis (transmutation yield 27%)

Since no reliable Am standard was available, the concentration values must be regarded with cares (figs. 3g and 3b).

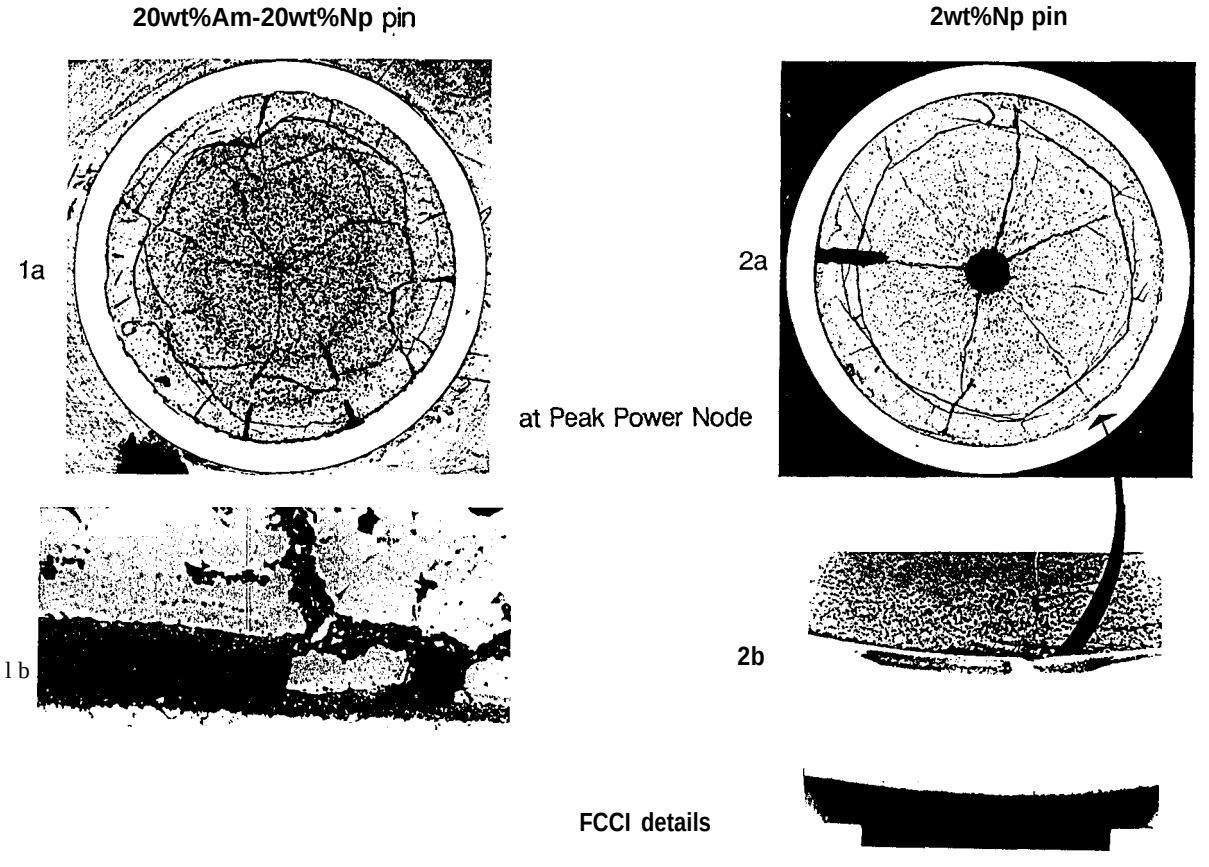
IV. TRANSMUTATION YIELDS

After irradiation in Phenix FR, a preliminary analysis confirms the good agreement of experimental values with Np and Am^{241} nuclear data calculations[12] for Supcrfact 1 transmutation rates (Table 4),

Table 4- Transmutation rates (τ.). Consumption calculation and experimental values after irradiation

Minor actinide	Np %			Am%	
Type of fresh fuel	2	20	45	2	20
Measurement	*	*	27	27 to 30	27 to 32
Calculation	24-26*	27.4	27.5	27.3	31.6

* in progress



Figures 1 and 2- Minor act inidc based fuels and cladding : structural and interaction aspects.

v. DISCUSSION

V.I. General behaviour

The usual examinations carried out have shown that the behaviour of the actinide pins whatever the actinide nature or their concentration, is not so far from a standard oxide pin one when taking into account the rather low linear power and burn up. Regarding the fuel clad chemical interaction, the aspect and phenomena involved seem to be not quite the same as those usually met on the standard oxide pins. Therefore, these observations do not allow direct extrapolation for longer irradiation using the corrosion model based on the standard oxide fuel. Nevertheless, these observations have to be softened by on one hand the moderate depths that lay inside the current viewed depth on the standard oxide pins at this burn up and on the other hand the irregular radial occurence with is commonly known on the standard oxide pins as a hint of the initiation of this phenomenon. Thus, nothing particularly worrying has to be assumed from this chemical interaction in longer irradiation.

V.2. Helium production and consequences

The unusual large quantities of helium retained in fuel originate only with the post irradiation period since helium diffusion rate at the fuel temperature during irradiation is high enough for helium to leave the fuel almost entirely. Consequently, the origin has to be searched within a nuclear process implying an α decay. Among the numerous transmutation products, reference[1] explains that chiefly Cu242 and in a less extent Pu 238 arc relevant sources because of their relative short a desintegration period, Fuel containing Am24 I and then producing appreciable amount of Cu242 during irradiation by a single neutron absorption will thus be able to yield large amount of helium bccausc of the Cu242 half time decay period of 162.5 days. At the opposite, Np237 fuel produces under irradiation mainly Pu238 that owns a 86.4 years period. When compared to an average irradiation duration (i.e. 2 or 3 the half periods), one readily secs that fuel containing Am241 will provide substantial helium quantities during irradiation and a few months after.

Helium created by this path during irradiation leaves the fuel and is then collected with the other fission gases released, namely Kr and Xe, in the plena of the pin. The helium thus acts as an extra source of the pin pressurization that increases the clad strain. In order to represent the importance of this helium pressurization on an irradiated 15-15 T1 CW cladding[13] we have assessed the evolution of the clad stress of the Superfact

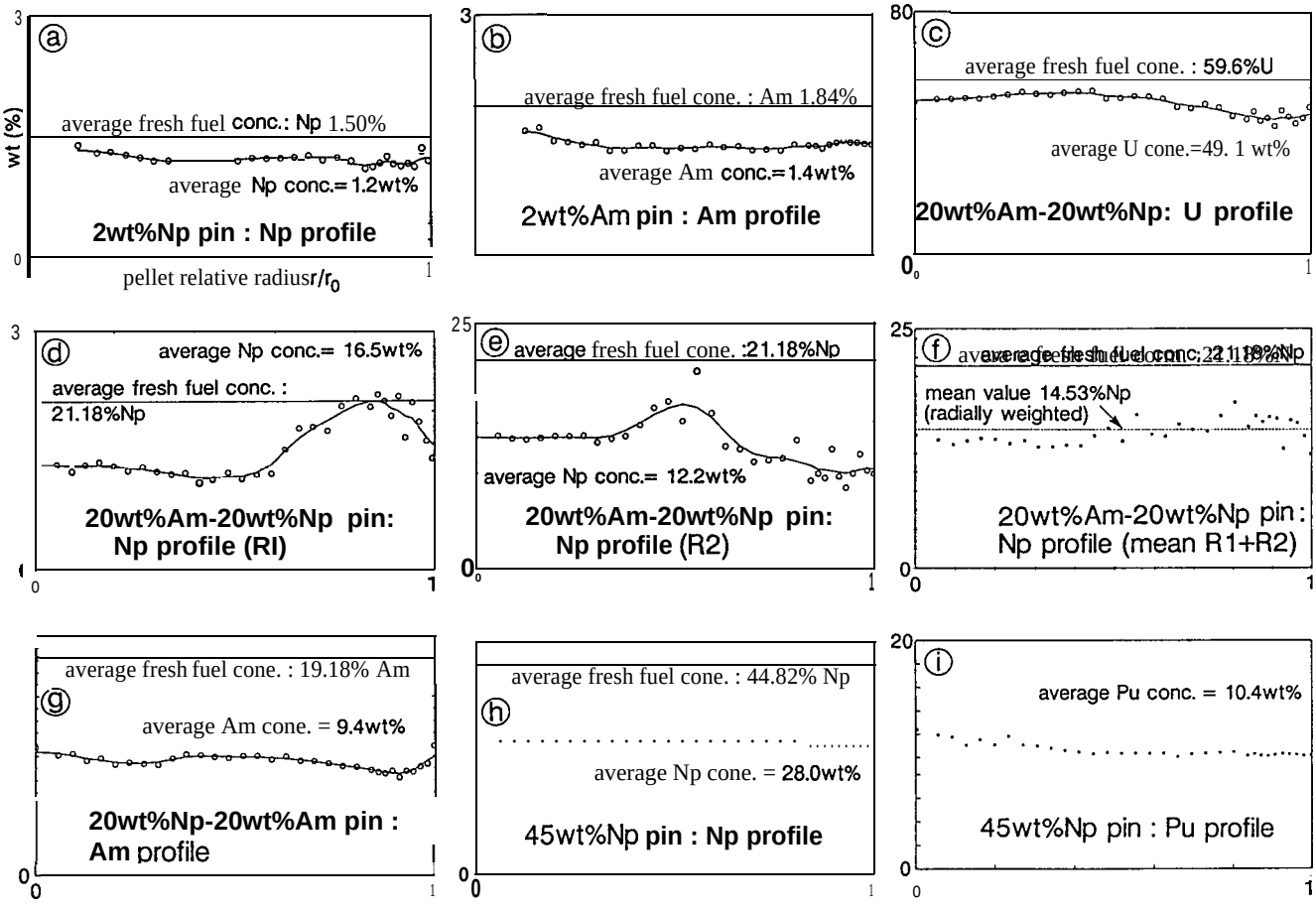


Figure 3- Microprobe analysis radial distribution of actinides in pellets

20 Am-20 Np pin assuming a linear helium production with the bum up. Then we transposed this estimation to a Phenix and Superphenix designed pin with the same kind of actinide based fuel. Reported on figure 4, this evolution assessment points out a mechanical limitation that could arise from this helium production on a irradiated 15-15 Ti CW cladding. Yet, this limit remains sufficiently far enough to be troublesome, and at least could be solved by a specific pin design (plena).

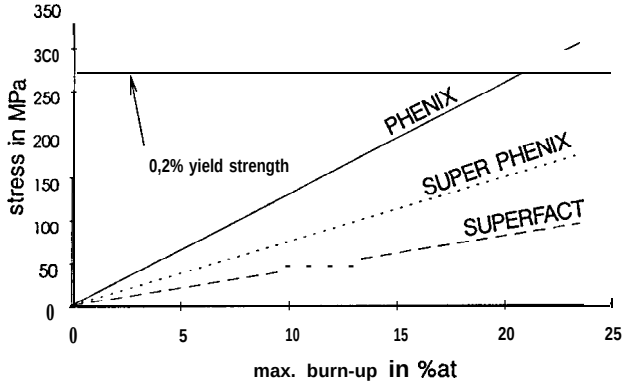


Figure 4 - Irradiated 15-15 Ti CW cladding stress through fission gas pressurization versus the bum-up (production base : Superfact, pin 6,20 % Am- 20 % Np).

V.3 Fast Reactor for transmutation

The radial distribution of transmutation products is flat as foreseen in a FR flux, because of the cross section that leads the mean path length of the neutrons to be by far larger than a pellet diameter. This caracteristic, specific of the FR, is rather an advantage compared to a reactor using a thermal neutron flux such as PWR. As a matter of fact, thermal flux transmutation experiments [14] has shown unflat radial distribution ($max/_{min} \approx 8$) with a peak located near the edge of the pellet. This overreactivity near the edge of the pellet is well known in the standard PWRas the "rim effect" and is still intcrprctcd in terms of self shielding.

Respecting the actinidc fuels, it remains that such an inhomogencous radial profile will not favour parameters such as corrosion rate and thermal conduction because of the accumulation of fission products (gazeous or elsc) at the edge of the fuel pellets. Moreover, the mean radial transmutation yield might stay low becausc of the shielding effect of the outer rim. That is why, thanks to the radially uniform transmutation yield these fuel elcmcnts present, the FR appears to bc an adapted type for transmutation of actinidc at high burn up.

Furthermore, an interesting point lies in the use for the actinide based fuels of an inert matrix instead of uranium in order to lessen the correlative plutonium production rate by neutron absorption of uranium and to accordingly improve the final actinide transmutation rate. A rough calculation, hypothesizing an irradiation of two pins holding 40 wt% Np, one with an uranium oxide matrix (near the Superfact fuel pins at 45 wt% Np) and the other with an inert matrix (Al_2O_3) at 1500 Effective Full Power Days conditions, leads to the following results[15]. After irradiation, around 21 wt% of neptunium are predicted to remain in the uranium matrix based fuel and 18 wt% in the alumine based fuels, i.e a weak advantage of 3 pts for the inert matrix based fuel. But, if we look at the plutonium creation, around 24 wt% are awaited within the uranium matrix based fuel under irradiation when only 18 wt% are produced by the alumine matrix based fuel. From theactinide complete balance point of view, such an inert matrix based fuel irradiated under the conditions seen above, turns out to be 10w% less producing. The rather good behaviour previously seen regarding the high actinidc concentration in an uranium matrix, promotes a promising opening to the use of inert matrix actinide based fuels.

VI. CONCLUSION

The first outcomes of the Superfact experiment is that the behaviour of the actinide based fuels is satisfactory and does not badly differs from the standard oxide one, at least until the rather low burn up reached (4.5 06.5 %at). Moreover, the theoretical background we had about those particular fuels seems adequate enough in the sense it allows the forecast in terms oftransmution yield, burn up and thermal level not to bc out of range of the post irradiation examination results, Superfact 1 experiment fullfilled the aims in presenting a satisfactory trail prior to a large scale (subassembly) semi-industrial demonstration[15].

The radial flat transmution rate, the actnide based fuels withstand in a fast neutron flux, promotcthc FRas an adapted reactor type foractinide high transmutation rate aims. The huge helium quantities, the americium based fuels provide during irradiation, have to be taken into account forthc pin design, but do not represent a barring problem. Though fuel clad chemical interaction in the actinidc pins is observed nothing bothersome has to bc feared because on one hand no correlation with neither the actinidc nature nor its concentration was noticed and on the other hand because the dephts measured lay inside the conventional range for the standard oxide pins at those bum ups. Nevertheless, this point remains to bc verified in higher burn up and larger scale experiment, Considering the total balance of actinide transmutation, some advantages seem to bc drawn with the use of an inert matrix for the actinide based fuel instead of uranium[16,17].

Eventually, the Superfact experiment is as well an outstanding and enjoying collaboration between the two CEC and CEA partners.

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