



TECHNICAL ABILITY OF NEW MTR HIGH DENSITY FUEL ALLOYS REGARDING THE WHOLE FUEL CYCLE

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ABSTRACT

The development of new fuel alloys could provide a good opportunity to improve drastically the fuel cycle on the neutronic performances and the reprocessing point of view . Nevertheless ,those parameters can only be considered if the fuel manufacture feasibility has been previously demonstrated .

As a matter of fact , a MTR work group involving French partners (CEA , CERCA , COGEMA) has been set up in order to evaluate the **technical ability** of new fuels considering the whole fuel cycle . In this paper , CERCA is presenting the preliminary results of UMo and UNbZr fuel plate manufacture, CEA is comparing to U_3Si_2 the Neutronic performances of fuels such as UMo , UN , UNbZr; while COGEMA is dealing with the reprocessing feasibility .

INTRODUCTION :

Within the framework of the Reduce Enrichment of Research and Test Reactors programme (RERTR) involving international operators, manufacturers and laboratories, the LEU silicide fuel U_3Si_2 has been qualified in 1988 by the American NRC up to a density of 4.8 g Ut / cm³. After 10 years background it appears now obvious that this fuel cannot fully comply with the needs of existing and future reactors. The density reached is not high enough to convert some HEU reactors satisfactorily and the back end solutions are not fully assured.

Based on these facts, the Argonne National Laboratory on behalf of the RERTR Programme presented, by the end of 1996, an irradiation plan for new fuels allowing theoretically to reach densities up to 8 or 9 g Ut / cm³ [1]. The Uranium alloys used for these fuels were supposed to be a γ stabilised phase. For really higher densities compared to U_3Si_2 , Uranium Molybdenum and Uranium Niobium Zirconium alloys were mainly considered.

Considering the whole MTR fuel cycle (Manufacturing, reactor operation and reprocessing) the major French actors have pointed out the need to analyse these new fuels and others that have been proposed before.

For this purpose a working group has been set up : CERCA has tested the manufacture feasibility, CEA has developed neutronic calculations and core performances more particularly considering the new LEU JHR reactor, whereas COGEMA has studied the reprocessing feasibility .

The main preliminary results are presented hereafter .

1 : THE FUEL MANUFACTURE

1-1 : γ alloy optimisation

1-1-1 : Aims of the optimisation and problems to be overcome

Unlike U_3Si_2 and UN , UMo and UNbZr alloys are not definite compounds with their own crystal structure but a solid solution where Molybdenum or Niobium and Zirconium are included within the Uranium metal crystal structure . The γ phase of the Uranium is required in this particular case because of the body centred crystal structure which is expected to have a good behaviour under irradiation .

At room temperature and in equilibrium conditions , the α stable crystal structure is generated. Therefore, γ phase can only be obtained in a metastable state at room temperature when quenching the alloy.

The optimisation work to be done related to the manufacture of the γ Uranium alloys can be summarised as followed :

- Obtain pure metastable γ phase at room temperature
- Keep γ phase during hot rolling (stability increase with the alloy content)
- Get an homogeneous alloy content with no segregations
- Be sure the refractory alloy is fully dissolved
- Obtain a dense compound (with a minimum alloy content)

CERCA has focused its development effort on two types of γ phase alloys which have their metallurgical specificity's :

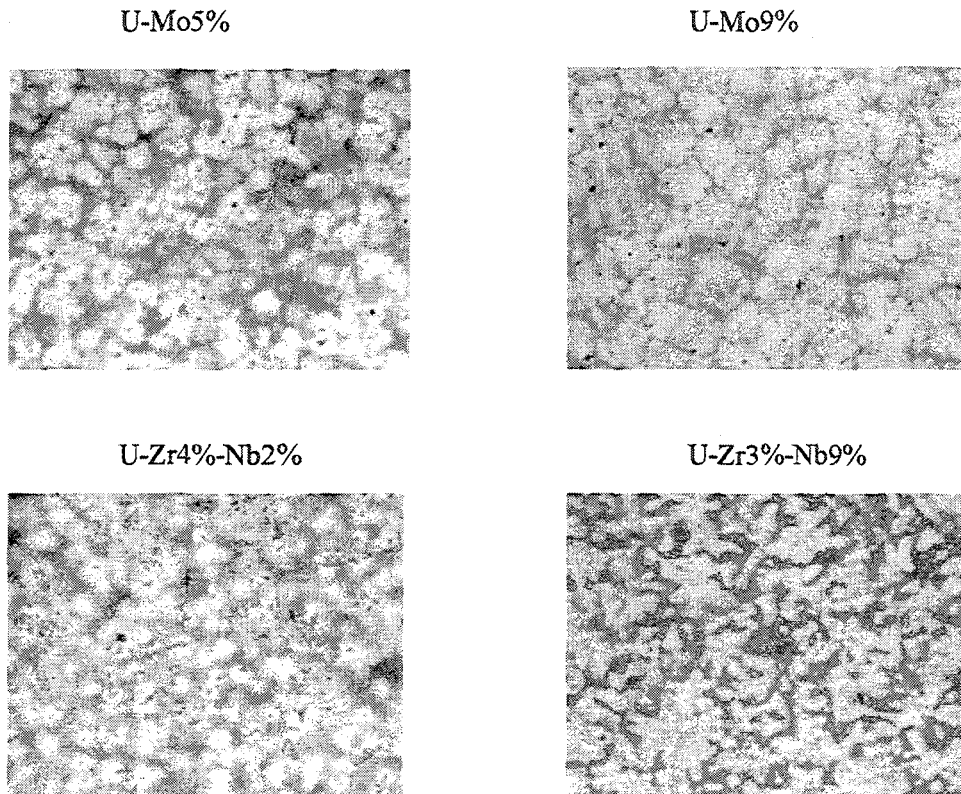
- Low alloy content (U-5%Mo and U-4%Zr-2%Nb)
High densities but may be poor thermal stability
- High alloy content (U-9%Mo and U-3%Zr-9%Nb)
Lower densities but higher thermal stability

1-1-2 : results of the casting developments :

Taking into account all the difficulties previously explained , a special procedure of casting and heat treatment has been developed in order to reach our goal : an homogeneous γ phase of Uranium alloys .

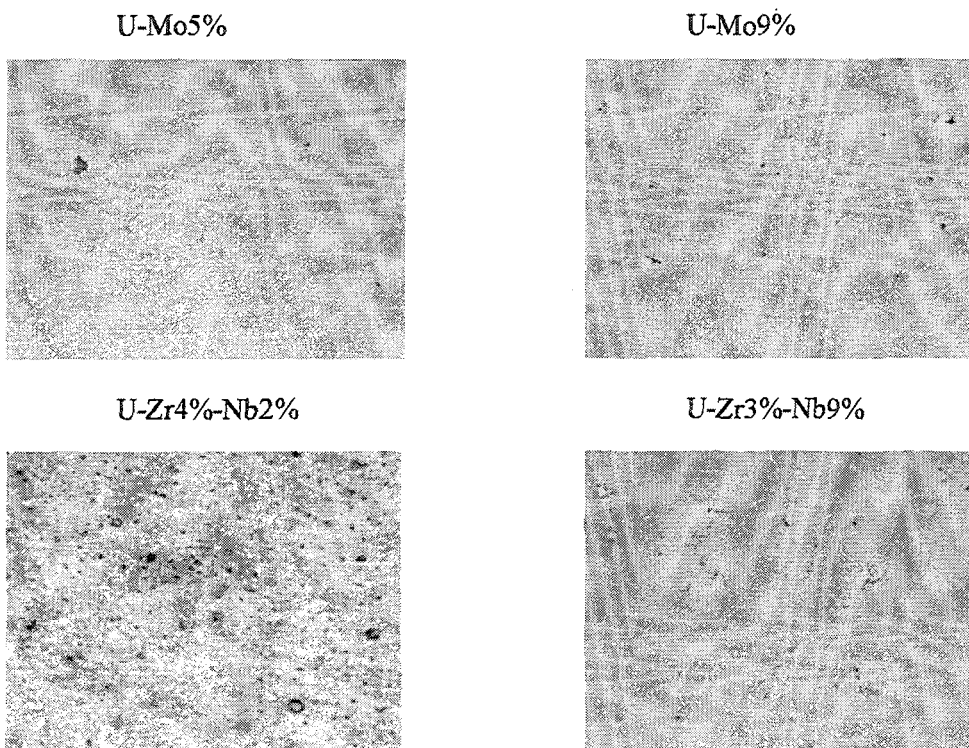
Figure 1 shows micrographs of "as cast" alloys ingots . Whatever alloy is considered, lighter areas can be seen in the centre of metallurgical grains which means a low content of Uranium whereas, the darker areas represent the grain boundaries with a high level of Uranium.

Figure 1 : As cast alloys ingots



On the UMo heat treated ingots (figure 2) no more segregation can be detected. Concerning the U-Zr3%-Nb9% alloy it still remains traces of precipitates which are made of roughly pure Zirconium whereas on the U-Zr4%-Nb2% sample it can be noticed that the grain boundaries segregation's have disappeared and an other type of inhomogeneity can now be seen : the mixing of α and γ phases .

Figure 2 Heat treated ingots



X ray diffraction patterns carried out on “as cast” alloys show pure γ phases for all the samples except in the case of U-4%Zr-2%Nb alloy which is composed of α and γ phases. The same characteristics can be analysed on the heat treated ingots .

After the development of the casting and heat treatments, the ingots were crushed and ground in order to get powder . The mechanical behaviour of γ alloys is completely different from U_3Si_2 , the comminuting parameters have been consequently adapted to those new fuels .

1-2 : The fuel plate manufacture with UMo fuels

As CERCA masters the manufacture of high loaded plates thanks to a proprietary advanced process allowing to reach 6g Ut / cm³ with U_3Si_2 [2], the same process has been tested with new fuels.

For a given volume fraction of fuel particles of 53 %, when replacing U_3Si_2 particles by γ phase alloys the following densities (g Ut / cm³) can be reached :

Vol fraction of fuel	U-5%Mo	U-4%Zr-2%Nb	U-9%Mo	U-3%Zr-9%Nb
53 %	9	8.6	8.3	7.6

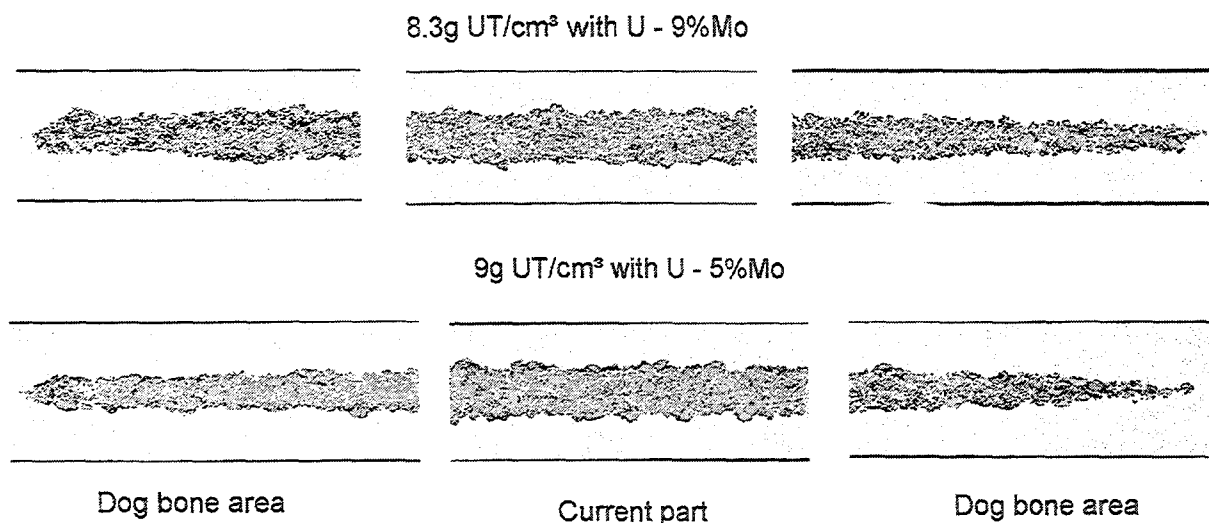
As it seems to be impossible to get U-4%Zr-2%Nb alloy with a pure γ phase, this fuel has not been selected for the manufacture test. In a first step, only UMo plates have been manufactured .

The first manufacture trials showed the advanced process could not be applied as existing due probably to the difference in mechanical behaviour between U_3Si_2 and γ phase alloys . The process has been consequently adapted and the main results are presented hereafter

1-2-1 : Plates micrographs .

Figure 3 shows micrographs in current part and the dog bone areas of 8.3 and 9 g Ut / cm³ plates manufactured with respectively U-9%Mo and U-5%Mo alloys. First of all, it must be noticed that the meat geometry of both plates are quite similar. This is as expected because of the same volume fraction of fuel particles within the meat (53 %). On both plate, no dog bone effect can be detected leading up to an acceptable minimum cladding thickness. Furthermore, the meat thickness is rather regular on the current area .

Figure 3 :



1-2-2 : Homogeneity of Uranium distribution :

Due to the difference in density between aluminium and UMo powders, the first plates manufactured had a very poor homogeneity quality. After a careful development work, a special procedure has been set up for the manufacture of the fuel meat in order to perform good plates.

For the 8.3 and 9 g Ut / cm³ UMo plates, the X ray inspection homogeneity diagrams stood easily between the allowed limits of the commonly used Silicide fuels specification when scanning the whole meat area

Although U-3%Zr-9%Nb has not been tested yet , the manufacture results are supposed to be equivalent since this alloy has roughly the same mechanical behaviour as UMo alloys because of the same crystal structure .

1-3 : Thermal stability:

During the manufacture, the fuel plate is maintained for a few hours at several hundred Celsius degrees, while, under irradiation, it has to endure one or two hundred Celsius degrees for many days.

The thermal stability of γ phase alloys is therefore very important to test. For that purpose, samples made of aluminium and γ phase alloys were tested at 400 ° C for several days. This test is very severe regarding the irradiation conditions . It must be pointed out that the U-4%Zr-2%Nb alloy has not been tested as it has not been possible to get pure γ phase.

After three days at 400 °C the U-9%Mo sample kept its perfect geometry and the γ structure remained unchanged . With the U-5%Mo ; U-4%Zr-2%Nb and U-3%Zr-9%Nb samples, X ray diffraction pattern showed α and γ phases with traces of UAlx. It can be suggested that γ phase of those alloys has returned to the equilibrium state, the α phase . Afterward the α phase has diffused with aluminium leading to UAlx .

1-4 : Conclusion about the fuel manufacture :

It has been demonstrated that the fuel manufacture of high loaded plates with γ alloys is feasible . Densities up to 9 g Ut/ cm³ can be obtained with U-5%Mo and 8.3 gUt/cm³ with U-9%Mo.

Nevertheless, not only for the fuel manufacturer but also for the reactors, the thermal stability is a very important selection criteria for the γ phase alloys. From that point of view U-4%Zr-2%Nb alloy is the worst candidate because it seems to be impossible to get a pure γ phase after casting. The U-5%Mo and U-3%Zr-9%Nb alloys are composed of pure γ phase after casting but are not very stable at 400 °C. The best candidate seems to be U-9%Mo as it fulfil the criteria of thermal stability.

In order to combine high density and thermal stability it could be considered an intermediate percentage of Molybdenum. An other solution lies in adding metal traces such as Ruthenium, Platinum or other to thermally stabilise U-5%Mo. In this particular case ANL has scheduled an irradiation test programme.

Neutronic performances evaluations of γ phase fuels could make a selection among the feasible and stable fuels .

2 : THE FUEL NEUTRONIC PERFORMANCES

This study aims at analyzing the influence of LEU fuel densification on the main neutronic physical characteristics of the Jules Horowitz Reactor core, by considering different types of dispersing agents presently under study.

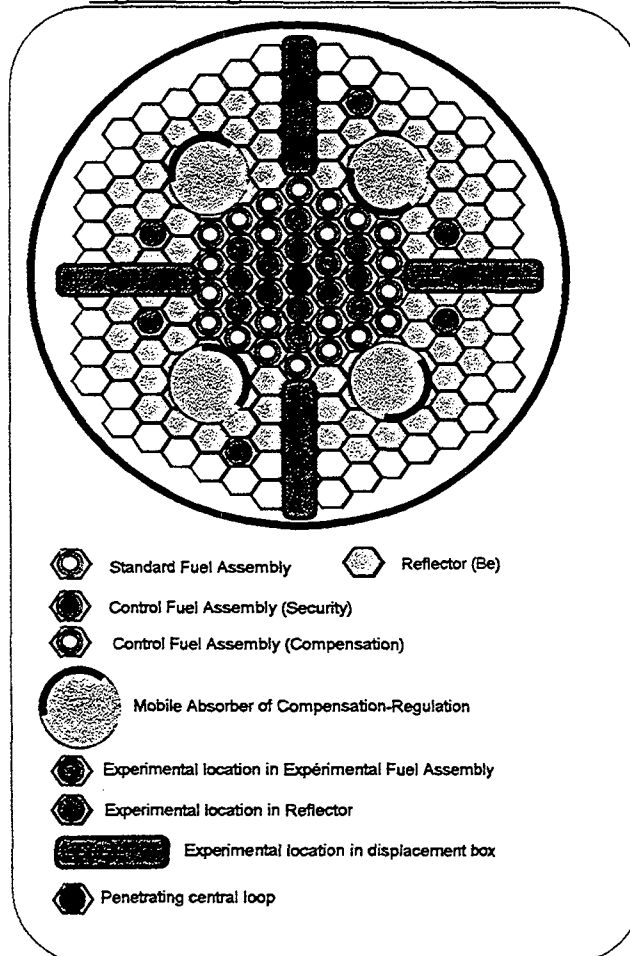
2-1: The Jules Horowitz Reactor

The Jules Horowitz Reactor (JHR) is a new research reactor project totally dedicated to nuclear material and fuel tests. It will eventually be one of the main European technological irradiation tools, and will provide its support to French nuclear power plants for the qualification under irradiation of new nuclear concepts and fuels, of materials and components. Its installation is foreseen on the CEA-CADARACHE site and its startup programmed for 2005.

In view of the present scope of irradiation requirements it will have to meet, and of the uncertainty as to their evolution at the beginning of the 21st century, the objectives attributed to the JHR core are ambitious. Besides high performances and great operating flexibility, it is expected to be of large experimental volume and great adaptability in order to meet future demands. Finally, it will have to be both highly available and generate the lowest possible cycle costs.

The choice of a non-proliferating LEU fuel (enrichment limited to 19.75 % in mass) was immediately made based on the co-laminated plates containing a dispersed silicide of density 4.8 g U.cm^{-3} in fuel meat. This choice is constraining for cycle costs, with a high yearly consumption of fuel elements.

Fig. 1 - Diagram of JHR reference core



The JHR has an open-pool type reference core, cooled by light water. With a maximum thermal power of 100 MW, it has a fuel height of 80 cm and an equivalent diameter of 51 cm. In order to reduce cycle costs and improve irradiation performances, it is surrounded by a thick Beryllium reflector in which experimental devices can be installed.

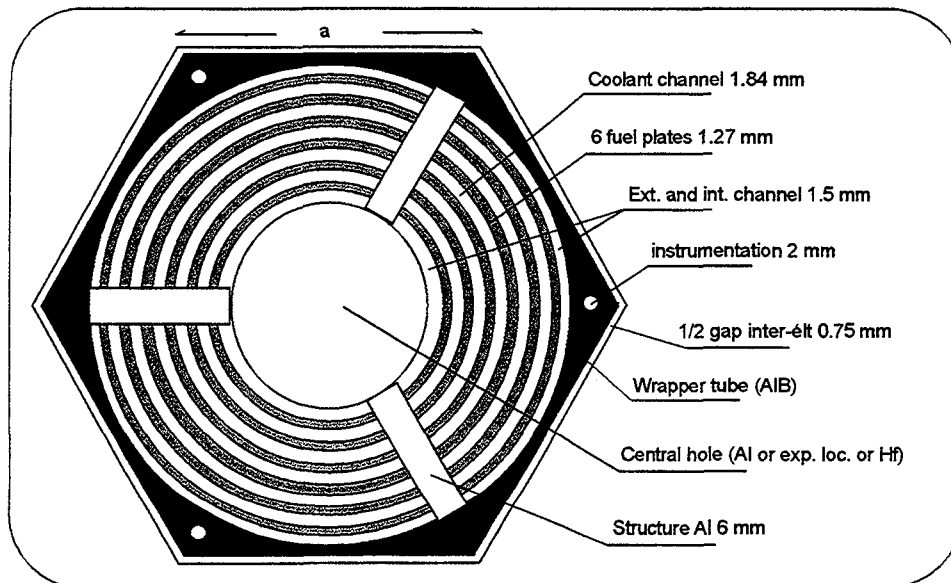
The core is formed by 37 fuel elements and is designed to receive a penetrating central loop dedicated to high flux irradiations, for purposes of FR representativity or achievement of power transients.

2-1-1 :The reference fuel element :

Because of the high specific power of the core (600kW/l), the primary water flow rate reaches 16 m/s crossing the core, thus implying large vibration risks. Also, the present reference fuel element is constituted by cylindrical plate sections, integrated in a hexagonal wrapper tube.

In order to obtain high in core fast flux performances, the plate lattice is greatly under-moderated with a reference thickness of the coolant channel of 1.84 mm (element called H10B') sometimes of 1.53 mm (element called H10B). The thermal component of the core is then mitigated. The reference fuel is made of co-laminated fuel plates containing a dispersed silicide of density 4.8 g U.cm⁻³ of fuel meat.

Fig. 2 - Diagram of H10B' reference fuel element



The characteristics of the reference elements considered are listed below :

	H10B'	H10B
Number of plates	6	7
Edge of wrapper tube (cm)	4.65	4.67
Coolant channel (mm)	1,84	1,53
Fuel volume (cm ³)	384	438
U Mass (g)	1842	2101
²³⁵ U Mass (g)	364	415
Specific power (W/gU)	1468	1287

2-1-2 : Operating characteristics and irradiation performances :

The yearly operation of the JHR is based on 9x(28 EFPD) cycles, in other terms a reactor availability factor of 0.69. Two operation modes are defined, one called the « S » reference mode with 37 fuel elements and the other called the « B » mode with a central penetrating loop in the core, with a diameter of between 80 and 150 mm.

The lifetime of the core operated in « S - one-batch » mode with 37 new H10B' elements is of 56 EFPD, authorizing a three-batch fractionated renewal on the basis of 28 EFPD cycles. The number of fuel elements annually consumed is close to 110, with a rate of fissioned ²³⁵U at discharge close to 70 %.

The maximum perturbed thermal (< 0.625 eV) and fast (> 1 MeV) fluxes respectively reach 9.E14 cm⁻².s⁻¹ and 8.E14 cm⁻².s⁻¹, depending on the operation mode.

2-2 : Neutronic study of the JHR with high density LEU fuels

2-2-1 : Objectives

This study aims at analyzing the influence of densification in U of the meat of the plates on fuel management, on the flux levels and the obtained neutron spectra, and on the negative reactivity of the control rods. The influence of the dispersing agent used is analyzed in terms of loss of lifetime (DL). A simplified calculation model of the one-batch lifetime is established for fuels with densities included between 4.8 and 8.5 g U.cm⁻³ in the meat.

2-2-2 : Modeling

A simplified 2D modeling of the core in the "S" mode was used for this study. It is based on the use of a calculation "project" scheme which links together the APOLLO1 and CRONOS2 CEA codes.

The aimed at eigenvalue at the end of cycle (VPFC) includes the calculation and technological uncertainties, a reactivity reserve at end of cycle of 1000 pcm and the negative reactivity of the non-modeled experimental load. It thus reaches VPFC = 1.11, and allows to deduce the t_b lifetimes of the one-batch operated cores and the optimum fractionated renewal modes on the basis of 28 EFPD/cycle.

The calculated reactivity worth are defined by : $\Delta\rho = \frac{K_1 - K_2}{K_1 K_2}$

2-2-3 : High density innovative LEU fuels

The innovative fuel characteristics considered in this study are summarized in the table below :

Fuel	Density (gU.cm ⁻³ meat)	Vol. Frac. of disp. agent (%)	M(²³⁵ U) H10B' (g)	Spec. P-H10B' (W/gU)	M(²³⁵ U) H10B (g)	Spec. P-H10B (W/gU)
U ₃ Si ₂	4,8	42,4	364	1468	415	1287
U ₃ Si ₂	5,66	50	428	1245	489	1091
UN	6,75	50	512	1044	584	915
UMo ₉	7,74	50	587	910	669	798
UZr ₄ Nb ₂	8,13	50	617	867	-	-
UMo ₅	8,5	50	645	829	-	-

These fuels are defined compounds (U₃Si₂, UN) or alloys (UMo at 5% and 9% in Molybdenum mass - respectively noted UMo₅ and UMo₉ -, UZr₄Nb₂ at 4% in Zirconium mass and 2% of Niobium).

2-2-4 : Results

The average fluxes obtained in unbounded lattice for the different fuels analyzed are the following :

H10B' fuel	Density (gU.cm ⁻³ meat)	χ^1 %	χ^2 %	χ^3 %	χ^4 %	Φ average 10 ¹⁵ cm ⁻² s ⁻¹
U ₃ Si ₂	4,8	24	33,6	27,3	15,2	1,62
U ₃ Si ₂	5,66	24,4	34,3	27,6	13,6	1,56
UN	6,75	25	35,3	27,8	11,9	1,52
UMo ₉	7,74	25,5	36,1	27,8	10,7	1,46
UZr ₄ Nb ₂	8,13	25,6	36,2	27,8	10,5	1,47
UMo ₅	8,5	25,6	36,6	27,9	10	1,46

H10B fuel	Density (gU.cm ⁻³ meat)	χ^1 %	χ^2 %	χ^3 %	χ^4 %	Φ average 10 ¹⁵ cm ⁻² s ⁻¹
U ₃ Si ₂	4,8	24,3	34,6	27,8	13,2	1,65
U ₃ Si ₂	5,66	24,9	35,5	27,9	11,7	1,59
UN	6,75	26	36,8	27,4	9,8	1,61
UMo ₉	7,74	26,2	37,7	27,5	8,8	1,59

Densification implies a "hardening" of the neutron spectrum. During a doubling of U density in core, the average thermal flux level in the core decreases by 41 %, a variation linked to the drop in specific power. The fast flux level slightly decreases by 4 %. These variations have little effect on the irradiation performances of materials in core, and of the fuel, the latter being placed in a Beryllium reflector.

However, the doubling of the U density results in an efficiency loss of roughly 26 % of the mobile core control absorbers made of Hafnium. It is therefore necessary to either increase their number in the core or to find an inherently more efficient material. This efficiency is 8 % lower in an under-moderated lattice (H10B).

The effect of the dispersing agent varies in evolution on the homogeneous core : in view of the relatively "hard" spectrum due to the under-moderation of the lattice and to the initially high U mass in the core, the fuel densification effect is not significant on the initial reactivity worth.

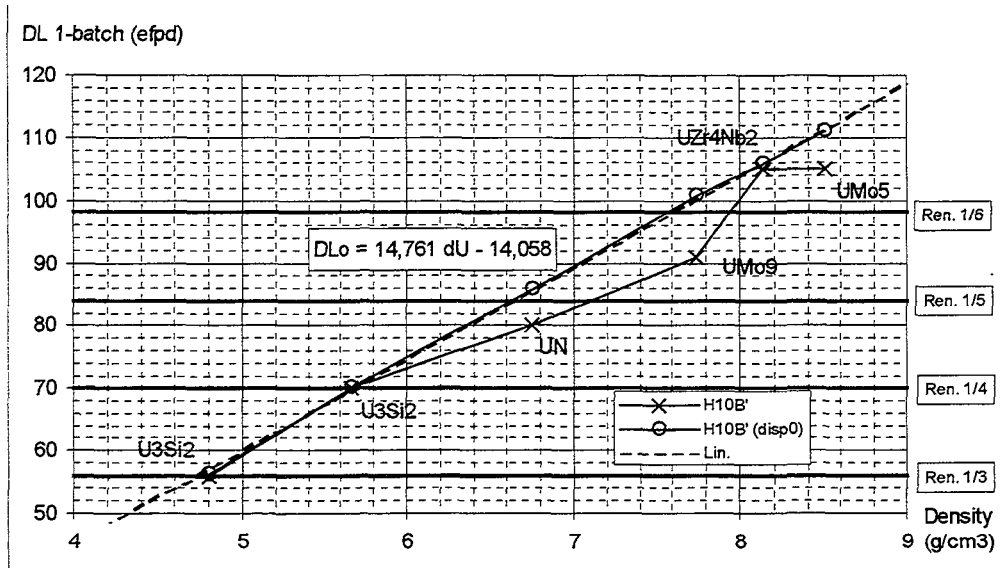
The higher the density, the lower the reactivity slope in evolution (lower specific power) and thus authorizes gains in one-batch lifetime which allows greater renewal fraction modes.

The negative reactivity of the dispersing agent and losses in lifetime t_b associated to a one-batch core are presented below :

H10B' fuel	Density (gU.cm ⁻³ meat)	DL 1-batch t_b (efpd)	EOL slope pcm/efpd	$-\Delta\rho$ BOL pcm	$-\Delta\rho$ EOL pcm	Δ (DL) efpd
U ₃ Si ₂	4,8	56	-399	22	44	-0,1
U ₃ Si ₂	5,66	70	-348	19	47	-0,1
UN	6,75	80	-277	1017	1639	-5,9
UMo ₉	7,74	91	-230	1673	2201	-9,6
UZr ₄ Nb ₂	8,13	105	-232	135	186	-0,8
UMo ₅	8,5	105	-215	952	1275	-5,9

H10B fuel	Density (gU.cm ⁻³ meat)	DL 1-batch t _b (efpd)	EOL slope pcm/efpd	-Δρ BOL pcm	-Δρ EOL pcm	Δ(DL) efpd
U ₃ Si ₂	4,8	58	-328	12	32	-0,1
U ₃ Si ₂	5,66	72	-265	8	27	-0,1
UN	6,75	81	-219	1058	1569	-7,2
UMo ₉	7,74	92	-186	1362	1734	-9,3

Fig. 3 - One-batch lifetime H10B' core with and without dispersing agent



As a rule, the one-batch core lifetime established without dispersing agent and for the density range analyzed can be approximately expressed by :

$$DL_0 = 14,761 dU - 14,058 \text{ (H10B')} \quad (1)$$

Due to the thermalization of the flux in evolution and to the difference in structure of the Nitrogen and Molybdenum cross sections, Nitrogen is 60% more penalizing at the end of cycle than at the beginning, whereas Molybdenum only increases by 30%.

In view of the low capture cross sections of Zirconium and Niobium, the UZr₄Nb₂ alloy appears to be very interesting from the neutronic point of view.

Finally, the gains in lifetime obtained lead to the forecasts on yearly consumption of fuel elements and to the discharge burnups (BU) presented below :

Fuel	Density (gU.cm ⁻³ meat)	Renewal fraction	M(²³⁵ U) cons./year (kg)	Nb. El./year	Disch. BU. (GWd/t _U)	Disch. BU. (%)	g ²³⁵ U/MWd
H10B'							
U ₃ Si ₂	4,8	1/3	40	108	123,3	70	1,13
U ₃ Si ₂	5,66	1/4	36	81	139,3	77	1,10
UN	6,75	1/4	43	81	116,8	65	1,10
UMo ₉	7,74	1/5	39	63	127,4	69	1,07
UZr ₄ Nb ₂	8,13	1/6	34	54	145,5	78	1,06
UMo ₅	8,5	1/6	36	54	139,2	75	1,07

Fuel	Density (gU.cm ⁻³ meat)	Renewal fraction	M(²³⁵ U) cons./year (kg)	Nb. El./year	Disch. BU. (GWd/t _U)	Disch. BU. (%)	g ²³⁵ U/ MWd
H10B'							
U ₃ Si ₂	4,8	1/3	46	108	108,1	62	1,13
U ₃ Si ₂	5,66	1/4	41	81	122,2	69	1,11
UN	6,75	1/4	49	81	102,5	58	1,11
UMo ₉	7,74	1/5	45	63	111,7	62	1,10

From the neutronic point of view, the most efficiently used fuels are U₃Si₂ (d=5.66) and UZr₄Nb₂. The latter allows an optimum use of fuel for a minimum yearly consumption of elements.

Besides their manufacturing abilities, their mechanical behavior and their reprocessing possibilities, these fuels must initially be qualified under irradiation to the discharge burnup values (probably over-estimated) obtained.

In the specific case of Nitride fuel, the capture reaction (n,p) on ¹⁴N produces ¹⁴C which is detrimental to fuel reprocessing. Based on conservative assumptions, we establish that the mass of ¹⁴C formed per element is 54 mg.

2-3 : Simplified calculation model of JHR core lifetimes

The loss in lifetime due to the dispersing agent evolves in a quasi-linear way versus its average macroscopic cross section adjusted to 1 energy group.

Hence, this cross section can be simply approximated directly using the data in the literature, by distinguishing for each dispersing agent, the thermal absorption (Group 4) of the resonant absorption (approximated to 3 other macro-groups). It is expressed :

$$\bar{\Sigma}_a = N_a \left(\sqrt{\frac{\pi}{4} \cdot \frac{T_0}{T}} \sigma_0 \chi^4 + \frac{I_R}{\ln \frac{E_2}{E_1}} (1 - \chi^4) \right)$$

with :

$$T_0 = 293.45 \text{ K}$$

T = 323.45 K (average temperature of the moderator setting the average "temperature" of the neutrons)

σ_0 = absorber microscopic cross section of the dispersing agent at 0.025 eV

IR = resonance integral of the dispersing agent (established in the E1, E2 energy range)

E1 = 0.5 eV (« limit » of the Cadmium)

E2 = 10 MeV

χ^4 = thermal contribution of the neutron spectrum (Group 4 = 0 to 0.625 eV)

Finally, we obtain :

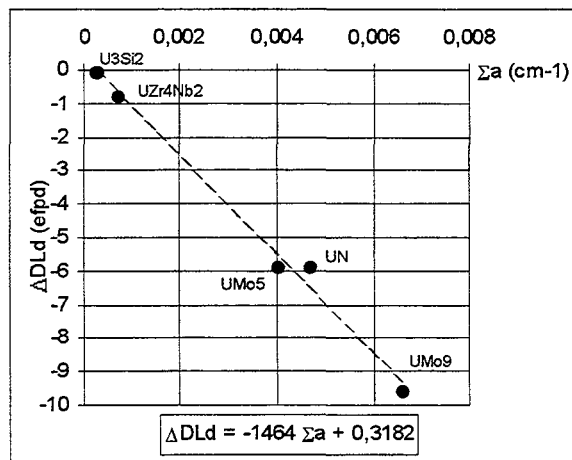
$$\bar{\Sigma}_a \approx N_a \left(0,844 \sigma_0 \chi^4 + 0,059 I_R (1 - \chi^4) \right) \quad (2)$$

The evolution law of the thermal component of the χ^4 neutron spectrum is deduced from transport calculations and is governed by the law :

$$\chi^4 = -0,0176 dU + 0,2691 \quad (\text{H10B}') \quad (3)$$

The loss in lifetime previously determined and plotted versus the average cross section calculated using the relations above, is represented in the figure below :

Fig.4 - H10B' model



The linear law obtained is :

$$\Delta DL_d = -1464 \Sigma_a + 0,3182 \quad (4)$$

Using Laws (1) to (4) previously established, the lifetime of the JHR core evolving in one-batch mode is expressed, in the area studied, by :

$$DL = DL_o + \Delta DL_d$$

thus allowing to deduce the optimum fractionated renewal mode. This lifetime is slightly dependent on the value of the two thicknesses of the coolant channel.

2-4 : Conclusion about JHR neutronic calculations

Within the framework of the study on the use of LEU - MTR fuels, the densification in Uranium of the fuel plates is of interest to the Jules Horowitz Reactor core. Despite the significant penalizing reactivity effect on the lifetime due to the dispersing agent used (Nitrogen and Molybdenum), the lifetime can be approximately doubled when the density is doubled, which does not significantly affect the irradiation performances. However, the significant loss in efficiency of the mobile reactivity control absorbers and probably that of the burnable poisons will have to be countered.

Based on a simplified modeling of the present JHR core concept, we established the correlations which allow the simple calculation of the gains in lifetime expected from any dispersing agent, of a density between 4.8 and 8.5 g U.cm⁻³, taking into account the basic nuclear parameters of the dispersing agent or of the several dispersing agents used.

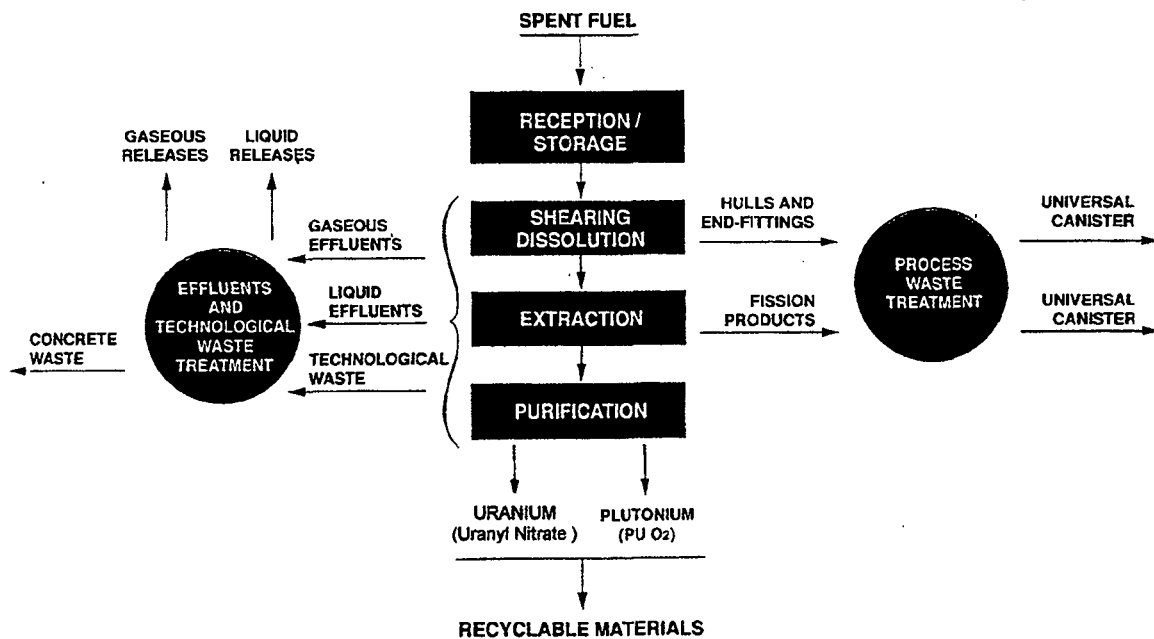
A more rigorous study taking into account a more realistic modeling, such as the presence of experimental devices and the integration of burnable poisons, needs to be carried out.

In view of the usual delays involved in the development and qualification of new fuels, the latter can nevertheless only be envisaged in future JHR irradiation programs, the reference fuel presently foreseen for its startup being U₃Si₂ with a density of between 4,8 and 6 g U.cm⁻³.

3 : THE FUEL REPROCESSING

Today the only proven back end solution for MTR spent fuels is the reprocessing. COGEMA is already proposing reprocessing services for Aluminide spent fuels, based on La Hague capability, and the reprocessing ability of the new LEU MTR fuels is a key issue for the spent fuel management. For this purpose, COGEMA is involved in the R&D activities to support this new fuel development .

The standard reprocessing operation for LWR spent fuel is summarized in the following diagram:



In the case of the MTR spent fuels, the diagram for reprocessing is the following :

- Reception of MTR spent fuels.
- Unloading under wet conditions.
- Storage in pool.
- Dismantling and dissolution.
- Dilution of dissolution mixture with LWR dissolution mixtures.
- Extraction and purification of Uranium and Plutonium.
- Vitrification of Fission Products.
- Treatment and conditioning of the wastes into residues.

Also, for reprocessing ability evaluation, the main step to be studied is the dissolution because after this step, the MTR dissolution mixture is diluted with a large amount of LWR mixture reducing significantly the impact on the following steps.

The standard dissolution conditions are the following :

- Boiling nitric acid.
- Final acidity of the mixture less than 3.5 N
- For MTR spent fuels, the final concentration of uranium in the mixture is limited by the aluminum solubility

In order to evaluate the behavior of the new fuel dispersant during the dissolution, COGEMA realized a R&D Program. The UN has not been included in this Program, because the feasibility of its dissolution has already been proven.

3-1 : The R&D Program

This Program has been implemented at the COGEMA/SEPA research laboratory which is mainly involved in ore treatment and natural uranium material tests and analysis.

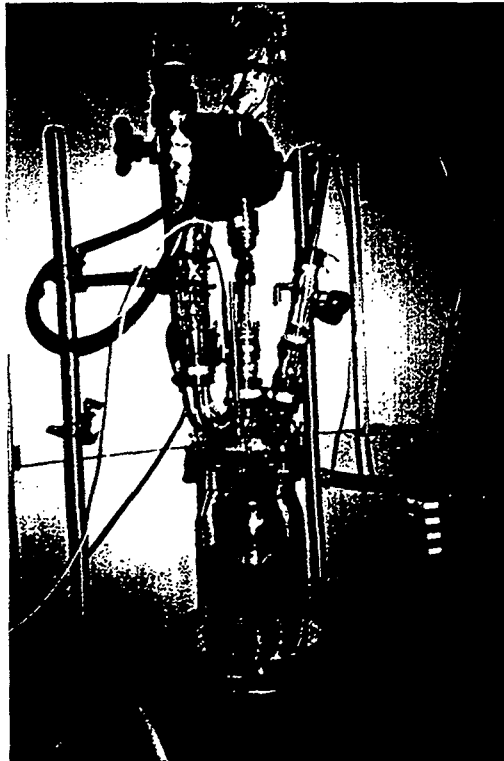
The main objectives of this Program were :

- Dissolution kinetic evaluation.
- Quantities and nature determination of the residual solid if any.
- Evaluation of hydrogen in the gas produced for security purpose.
- Determination of the NH_4^+ concentration in the mixture also for security purpose.

A simple testing device has been designed for that purpose at the SEPA. It consist mainly of a laboratory vessel of 1 liter equipped with a total reflux condenser and a gas monitor
The sample weight around 20 g to have a final uranium concentration in the solution representative of the one obtained in the industrial process.

The sample was introduced in the vessel already filled with cold nitric acid. Then the temperature was increased slowly until boiling. Every hours, a solution sample was collected to be analyzed. At the end of the dissolution, the solution and the remaining solid was analyzed.

Laboratory testing device for dissolution ability evaluation :



To realize this Program, CERCA provided us with depleted uranium fuel dispersants of UMo5, UMo9, UZr4Nb2, UZr3Nb9 in bulk solid pieces and in powder (particles size range of 40-90 µm or 90-125 µm)

Matrix	Al compacted powder	U-Mo5%	U-Mo9%	U-Zr4%-Nb2%	U-Zr3%-Nb9%
Preliminary test		N°1 HNO ₃ 3,5N		N°2 HNO ₃ 3,5N	
Solid pieces			N°3 HNO ₃ 3,5N		N°4 HNO ₃ 3,5N
Complementary Test on Solid pieces	N°9 HNO ₃ 3,5N final		N°6 HNO ₃ 3,5N + Al	N°7 HNO ₃ 3,5N + Chelating agent	
Powder			N°10 HNO ₃ 3,5N (1)	N°11 HNO ₃ 3,5N + Chelating agent	

(1) Compacted powder of UMo + Al

3-2 : Main results on dissolution

3-2-1 : Al dissolution

A test has been conducted with Al compacted powder for kinetic comparison purpose. The reaction started at 75°C, with an important hydrogen production increasing with temperature. It was completed after 7 hours. The final mixture contained a significative amount of NH₄⁺. The acid nitric consumption was around 3.8 moles per mole of aluminum (to be compared with theoretical value : 3.75).

This confirm that the dissolution is possible without any catalysis agent. Regarding the hydrogen production, the production rate could be monitored by adjusting the initial acid concentration and the temperature.

3-2-2 : UMo dissolution

These tests show a very good behavior of UMo regarding dissolution.

No solid residue and no precipitate have been found at the end of the dissolution which is quite fast (less than 5 hours for bulk solid pieces).

The test with the UMo + 9.3% Al compacted powder shows a high reactivity : the dissolution started at 65°C and has been completed in around 30 mn. If necessary, in the industrial process, this reactivity can be monitored with the temperature parameter.

The tests show also that no significant quantities of hydrogen or ammonium ion has been formed.

The dissolution kinetic is close to the one observed with UO₂ fuels.

According to this preliminary results, the UMo appears to be a really good candidate for its dissolution behavior.

3-2-3 : UNbZr dissolution

During the first test with nitric acid alone, the dissolution has been much longer than in the case of UMo (from 24 to 48 hours for bulk solid pieces). As soon as the dissolution started, the dissolved Nb and Zr elements precipitated. At the end, the precipitate represented a few percent of the initial mass, while the residual solid represented another 5%.

Some complementary tests allowed us to define an appropriate chelating agent to avoid this precipitate for Nb and Zr. With this chelating agent, the complete dissolution for powder should be reached after 10 to 20 hours. Due to an experimental device failure, the corresponding test has to be done again. But the analysis of the partial results allows us to suppose that the dissolution will be complete, with no residual solid and no precipitates.

The dissolution kinetic seems to be of the same order of magnitude as for Metallic Uranium fuels.

3-3 : Extraction and vitrification aspects

3-3-1 : Extraction

As stated before, the dilution rate of the MTR dissolution mixture with the LWR mixture is high enough to say that no impact is forecasted on the extraction units. But, in the case of UZrNb, the addition of a chelatic agent could disturb the solvent extraction process. This point has to be checked by a specific R&D Program.

3-3-2 : Vitrification

The elements Mo, Zr, and Nb are already present in LWR spent fuels as fission products. The reprocessing of MTR spent fuel is limited by the aluminum content of the MTR fuels. Tacking into account this aluminum limitation, the impact of new fuel dispersants has been evaluated regarding the vitrified residue composition in comparison with LWR fuels alone :

- For UN, the N element has no impact on vitrification
- For UMo, the Mo content increase of a maximum of 2% (for UMo9)
- For UZrNb, the Zr and the Nb content increases are negligible

Also, for these new fuel dispersants, the slight increase of Mo, or Zr and Nb, will have no significant impact on the vitrification process. Further, the already internationally approved specification of the vitrified fission products residue will be met.

3-4 : Conclusions about fuel reprocessing

This preliminary review of the new fuel dispersants, candidates for a high density fuel development, allows us to classify them, regarding the reprocessing ability, as follows :

- At the first rank, we will place UMo and UN. The reprocessing of such fuels will be very similar to aluminide fuels.
- Then will come UZrNb fuels. The management of Zr and Nb precipitates will mean the addition of a chelating agent which might have some disturbing effects in the case of a real dissolution mixture containing a wide range of chemical species.
A specific R&D Program will be necessary to evaluate these effects.
In addition, the dissolution kinetics is less favorable than for UMo

The choice of a new fuel dispersant will obviously rely also on fabrication and irradiation behavior matters. But, to allow the operators to run smoothly their Research Reactors in the future, it is essential that a new fuel development includes the insurance of a steady back-end solution.

CONCLUSION :

There is not one fuel candidate which is preferred simultaneously by all the three steps of the fuel cycle: the manufacture , the irradiation and the reprocessing .

A compromise must be found between the neutronic performances (low alloy content needed) and the thermal stability which require high alloy content.

In any case, the reprocessing feasibility has been demonstrated . After UN , the UMo alloys seem to be significantly easier to reprocess as compared to UNbZr

As a preliminary conclusion, a UMo alloy with a quite low Mo content and an additive to enhance its thermal stability might be a good compromise.

This point of view has to be considered as a first evaluation approach linked to the French needs and production processes. It could be a basis for further discussion with each international actor involved in the development of a new LEU MTR fuel.

References :

[1] : DEVELOPMENT OF VERY HIGH DENSITY FUELS BY THE RERTR PROGRAM

presented at the RERTR meeting in SEOUL (KOREA) , October 1996

by J.L. Snelgrove, G.L. Hofman , C.L. Trybus , T.C. Wienczek

Argonne National Laboratory (ANL) ; Argonne , Illinois , USA

[2] : DEVELOPMENT OF HIGHER DENSITY FUEL AT CERCA

Presented at the RERTR meeting in Roskilde (DENMARK) , October 1992

by JP Durand , Y. Fanjas , A. Tissier (CERCA)

[3] - B. BARRE - F. MERCHIE - P. RAYMOND - S. FRACHET - B. MAUGARD - « REX 2000 : a new material testing reactor project », Proceedings of the « Research Facilities for the future of Nuclear Energy » ENS topical meeting, June 4-6, 1996, Brussels, Belgium.

[4] - B. MAUGARD - D. GALLO - S. FRACHET - P. RAYMOND - F. MERCHIE - « REX 2000 core : a new material testing reactor project », Proceedings of the « Breakthrough of Nuclear Energy by Reactor Physics » PHYSOR 96, September 16-20, 1996, Mito, Ibaraki, Japan.

[5] - S. FRACHET - P. MARTEL - B. MAUGARD - P. RAYMOND - F. MERCHIE - « The 'reactor Jules Horowitz' : a new experimental reactor project », Proceedings of the 5th International Conference on Nuclear Engineering ICONS 5, May 26-30, 1997, Nice, France.