THORIUM UTILIZATION IN PWRs: STATUS OF WORK IN THE COOPERATIVE BRAZILIAN/GERMAN PROGRAM

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Abstract

The major results of the program "Th-Utilization in PWR's" are presented and discussed: The investigations show that the standard KWU-PWR can accomodate $(Th,U)O_2$ and $(Th,Pu)O_2$ fuel without changes in the fuel element design, in 3 and 4-batch operation scheme, without penalties in the reactor performance. An advanced fuel fabrication scheme using direct pelletizing methods out of ex-gel microspheres has been developed on laboratory scale. The $(Th,U)O_2$ test fuel produced satisfies PWR-specification and is currently undergoing irradiation testing. Thermal and mechanical design of fuel pins with thoria-based fuel is validated by the current instrumented single rod irradiation test. Cold laboratory investigations indicate that the $(Th,U)O_2$ PWR-fuel can be reprocessed using presently known technology, including the chop-leach technique and modified THOREX extraction process.

I. Introduction

The utilization of thoria fuel in thermal reactors can result, due to a high η-value of U-233, in higher conversion ratios and consequently in better resource utilization compared with

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the conventional uranium fuel. This holds particularly for advanced reactor types specifically designed for thoria application, as shown by numerous R+D activities in many countries, among others Brazil and Germany. But also in Light Water Reactors substantial advantages are anticipated in a case of a closed fuel cycle /1/. However the thorium fuel cycle technology was not as mature to permit well based feasibility statements in this area.
Therefore the cooperative R+D program on "Thorium Utilization in Pressurized Water Reactors" between NUCLEBRAS/CDTN on the Brazilian side and KFA, KWU and NUKEM on the German side aims to an improvement of our knowledge on this subject and contributes to fullfill in practice the "Governmental Agreement on Cooperation in the Field of Science and Technology" of 1969 and the "Memorandum of Understanding between KFA and NUCLEBRAS" of 1978 / 2 - 5 /.

The objectives of the program running since mid 1979 are:

- to analyse and prove the thorium utilization in pressurized water reactors,

- to manufacture, test and qualify Th/U and Th/Pu fuel rods and elements under operating conditions,

- to study the back end of thorium fuel cycle by reprocessing spent Th-containing PWR fuel elements where it seems to be reasonable, or by applying available storage techniques.

The program utilizes as much as possible the existing technologies and available know-how for fuel element and nuclear core design, fuel element fabrication and reprocessing. Therefore the existing techniques and equipment for the fuel cycle in High Temperature Reactors, i.e. manufacturing and reprocessing of fuel, as well as for the fuel cycle in Light Water Reactors, i.e. fabrication and head end treatment, have been applied.

2. Nuclear Core Design

The KWU standard 1300 MWe PWR is the reference reactor for the present study. The results of nuclear core design are based on Th-fuel cycles using highly enriched uranium (HEU) or LWR recycle Pu as initial fissile material / 1, 5 /. It was found that the present design KWU-type PWR can be operated without changes and restrictions with 3 and 4-batch loadings in open and closed fuel cycle modes with all types of fissile material investigated / 1, 6 /.

It is to be anticipated that the realistic way of introducing Th-fuel to a PWR would be via a partial and later complete Th-fuel assembly reload / 1, 7 /. Results gained so far in the cooperative program show that no changes of the Th/HEU fuel assembly design - in comparison with the standard U-fuel assembly - to be loaded into an U-core is necessary. The use of Pu instead of HEU reveals also some interesting feature. Fig. 1 shows the relative energy production of Th/Pu-fuel inserted in a standard PWR core. However the high fission cross sections of Pu combined with a high thermal flux of neighbouring U fuel assemblies would cause unacceptable power peaking / 1, 7 /. Thus, Th/Pu-MOX fuel assemblies - as used for Pu recycling via U/Pu-MOX assemblies - need 2 - 3 types of fuel pins with a different fissile material content as shown in Fig. 2, to avoid local power peaking when loaded adjacently to U-fuel assemblies / 1, 7 /.

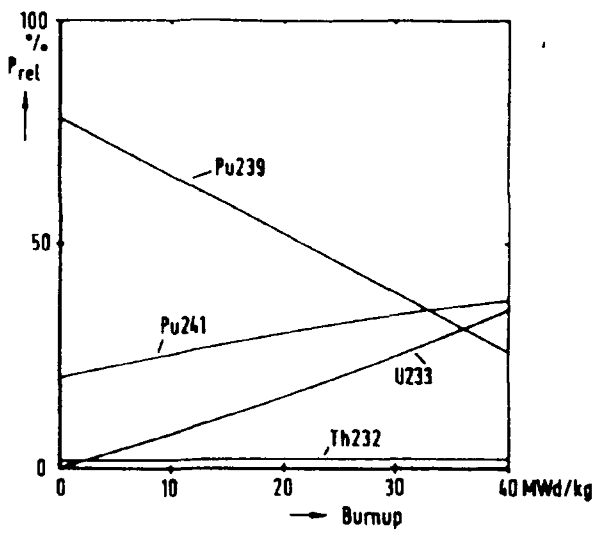


Fig. 1 Relative Energy Contribution of the Isotopes of Th/Pu Fuel

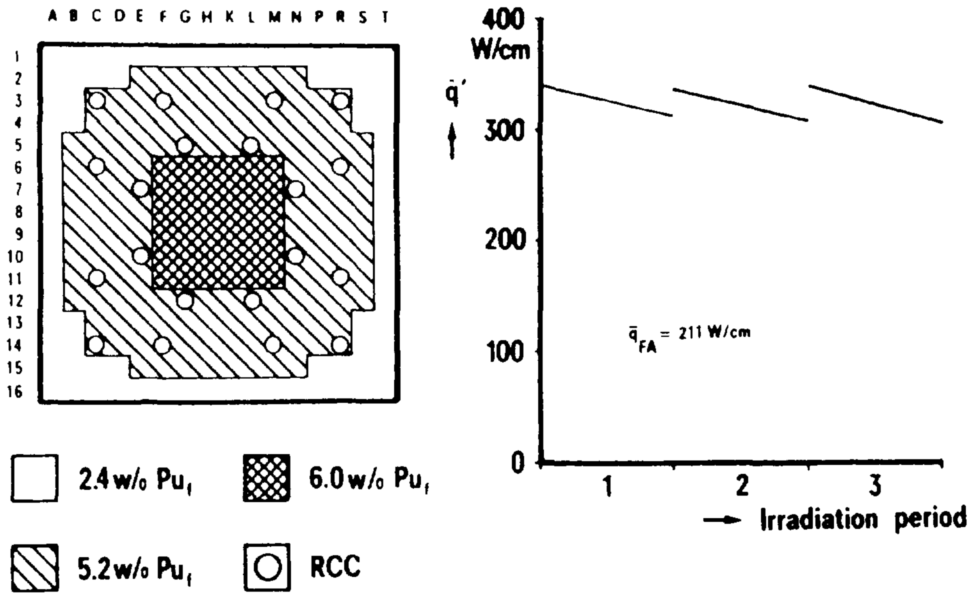


Fig. 2 Th/Pu Fuel Assembly with 3 Enrichment Zones

3. Strategy investigations

Strategy investigations are based on the need for natural uranium and separative work, calculated from the fuel cycle data of the nuclear core design / 1, 6 /. Recycling is found essential only for the Th/HEU to achieve substantial savings.

Recycling, however, may not be needed in the case of Th/Pu-cycles to realize savings in the need for natural uranium and separative work. Considerable saving come from Th/Pu-fuel via the replacement of U-fuel in existing reactors. The Th/Pu-fuel offers the potential of extending burn up to values where early closure of the fuel cycle by reprocessing Th-fuel could be avoided.

4. Fuel Technology

As reported above the present available fuel assembly design for KWU 1300 MWe PWR can be used to full extent to realize a thoria based fuel element. Thus, there is only a need for developing the oxide fuel manufacturing technology. This result from the nuclear core design is to be regarded as a major benefit for the Th use in çommercials PWRs.

Since there are large experiences available for chemical conversion processes for $(Th,U)O_2$ fuel production from HTR work / 8 / and for direct pelletizing of UO_2 / 9 / from PWR technology it was emphasized to use, as shown in figure 3:

- the chemical ex-gel conversion process in combination with

- standard pelletizing processes for UO_2 ex-AUC.

Selecting this combination offers the unique possibility to use the existing manufacturing equipment and quality assurance programs from commercial PWR and HTR / 8, 10, 11 /.

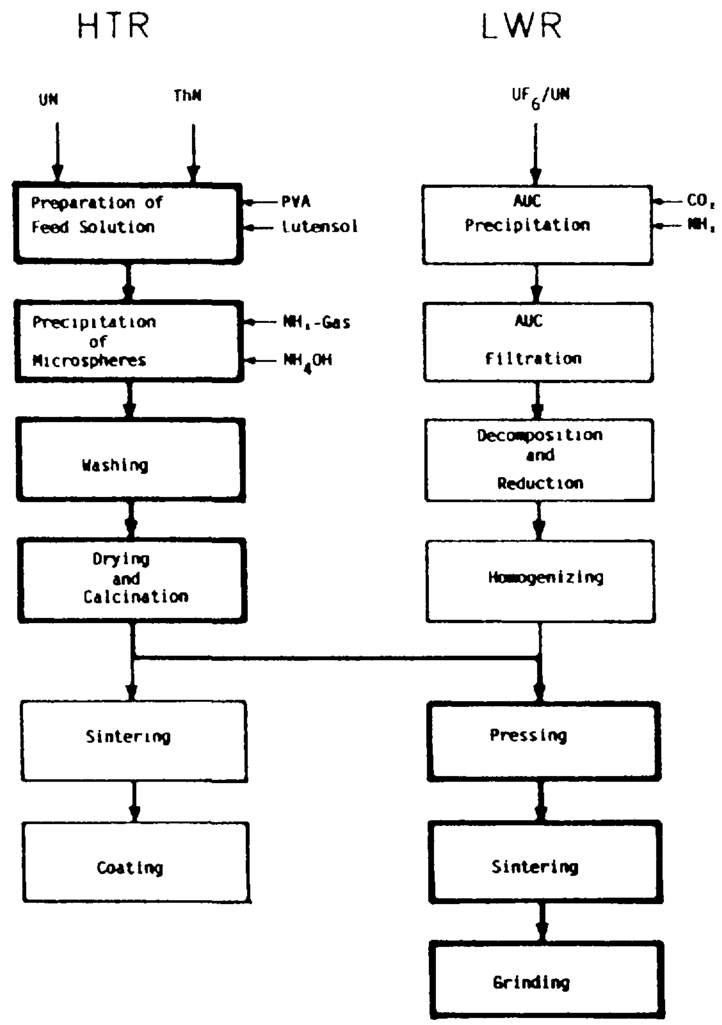


Fig. 3 Combination of Available Technologies for the Manufacturing of Mixed-oxide Fuel

Table 1 : Criteria for $(Th,U)O_2$-LWR-fuel for high burnup

Criteria		**High burnup targets for UO_2**	**High burnup targets for $(Th,U)O_2$**	**Status of $(Th,U)O_2$-fuel properties**	
Density	(g/cm^3)	(95 % td) 10.40	9.5	9.00 - 9.80	
Open porosity	(Vol.-%)	0.5 - 1	0.5 - 1	0.5 - 3	low op.por. can be controlled
Grain size	(μm)	10 - 20	10 - 2	10 - 15	
Average pore diameter	(μm)	2.5 at 10.40 g/cm^3	not yet evaluated[1)] (2.0 - 3.0 preliminary)	1.8 - 3	
log. scattering factor of pore size distribution	./.	0.25 - 0.30	0.25 - 0.30	0.25 - 0.3	
Shape factor[2)] of pores		> 0.7	> 0.7	> 0.7	

1) the results of FRJ-2 tests are necessary to confirm these values

2) $\frac{1}{F} = \frac{\text{Circumference of a pore in the microsection}}{\text{Circumference of a circle with the same area}}$

Table 1 summarized the target properties of the fuel with respect to the burn up defined in the nuclear core design and strategy studies / 1, 11 /. Two major areas of concern have been identified in the early stage of $(Th,U)O_2$-fuel development:

- the improvement of the fuel microstructure, especially to avoid the so-called black-berry structure.

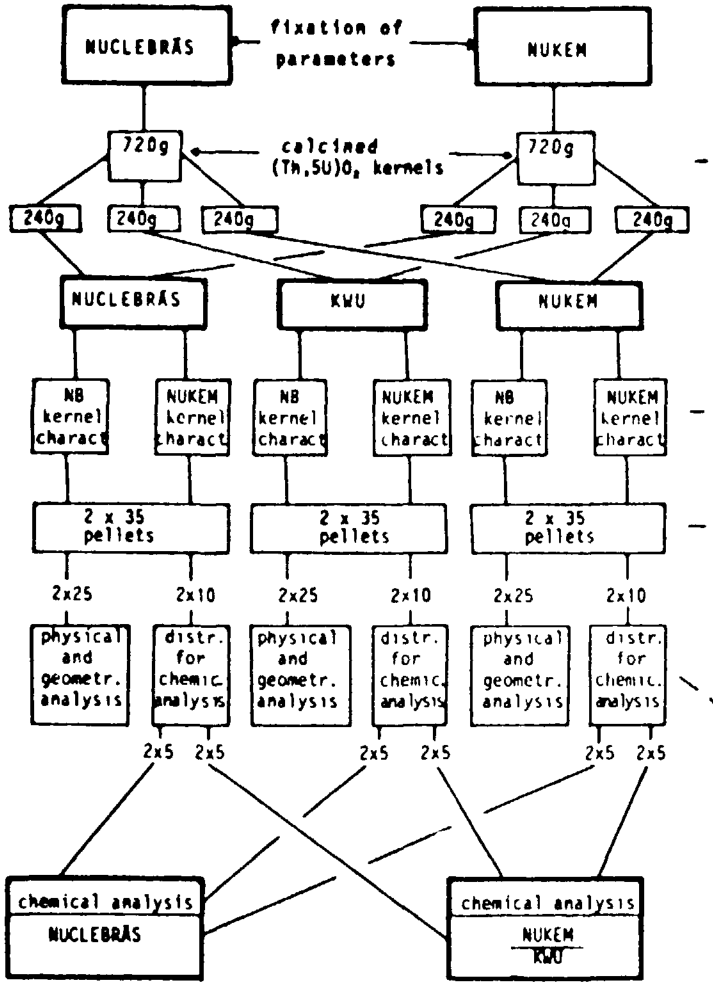
- the adjustment of the pore size distribution due to requirements given in table 1.

Details of the performed R+D-work have been reported earlier / 11, 10, 12 /. It turned out very soon that standard ex gel kernels are not suitable for the direct pelletizing process. However, the use of carbon black as additive to the feed solution permitted the adaptation of the ex gel kernels to the requirements for pressing and sintering, as well as to the control of density, microstructure and especially of the pore size distribution. The use of carbon black needs, of course a treatment for its removal after the kernel precipitation. The carbon black is completely oxidized during the calcination to CO_2 leaving the fuel kernel easily. Selecting its particle size distribution adequately, the burning carbon black leaves not only "press pores" behind improving the pelletizing, but also pores needed for the accomodation of swelling /shrinkage behaviour during burnup / 16 /. Table 2 compiles the results from test fuel production.

The reliability of the results from the R+D-program for $(Th/U)O_2$-fuel the fuel development is assured by a "Round Robin Test" involving a cross check of the manufacturing procedures performed at the different laboratories, as well as a cross check of the properties of the prematerial and the final pelletized fuel (Fig. 4).

Table 2 : Comparison of property-requirements and results from test fuel production

Properties	**Unit**	**Specified**	**Observed**
(Th+U)-Content	w/o	min. 87.7	87.83
U-Content	w/o HM	5 ± 0.1	5.03
Stoichiometry	MolO : MolU	1.99 - 2.05	2.014
H_2-Content	$Nmm^3/g\ (Th,U)O_2$	max. 10	9.4
Residual Gas Content	$Nmm^3/g\ (Th,U)O_2$	max. 40	17.6
Impurity Content:			
F	ppm (max.)	10	3
Cl	ppm (max.)	15	6
C	ppm (max.)	100	< 10
S	ppm (max.)	500	316
N	ppm (max.)	30	< 14
Ca	ppm (max.)	100	< 51
Si	ppm (max.)	100	51
Ni	ppm (max.)	50	10
Fe	ppm (max.)	100	23
Boron equivalent (Gd, B, Sm, Eu, Cd, Li)	ppm (max.)	1	0.911
Density	g/cm^3	9.5 ± 0.15 - 0.2	9.47 - 9.58 adjustable
Mean grain size	µm	4 - 15	8 - 15
Pore structure	in line with requirements		
Resintering Test	g/cm^3	max. 0.20	0.08
Surface Roughness	µm	2	0.89 - 1.14



Kernel fabrication (2x5 batches with 720g each)

Kernel characterization (each partner)
- . diameter n=150
- . crushing strength n=30
- . particle density 1X
- . BET 1X

Pellet fabrication (5x2 sinter batches with 35 pellets for each partner, totally 30 sinter batches)

. pressing of green pellets (each partner)
- . ρ_{green} accord. to indiv. working point
- . $\Delta\rho_{green}$ should be as low as possible

. sintering of pellets (each partner)
- . sintering of pellets
- . $\rho_{sint.}$ = 9.50 g/cm³ nominal value
- . sintering parameter 1700°C, 2 h, H_2 or H_2 mixture

Pellet characterization

. physical and geometrical characterization of pellets (no exchange of samples)
- . geometry, density/open porosity, and roughness ... total
- . resinter test ... 2x
- . microstructure
  - pore structure ... 2x
  - grain size ... 2x
  - pore size distr. ... 1x

. chemical characterization: separate Round Robin Test (not all chemical analitical methods available at all partners, exchange of samples in these cases)

Fig. 4 Detailed Procedure of the Round Robin Test

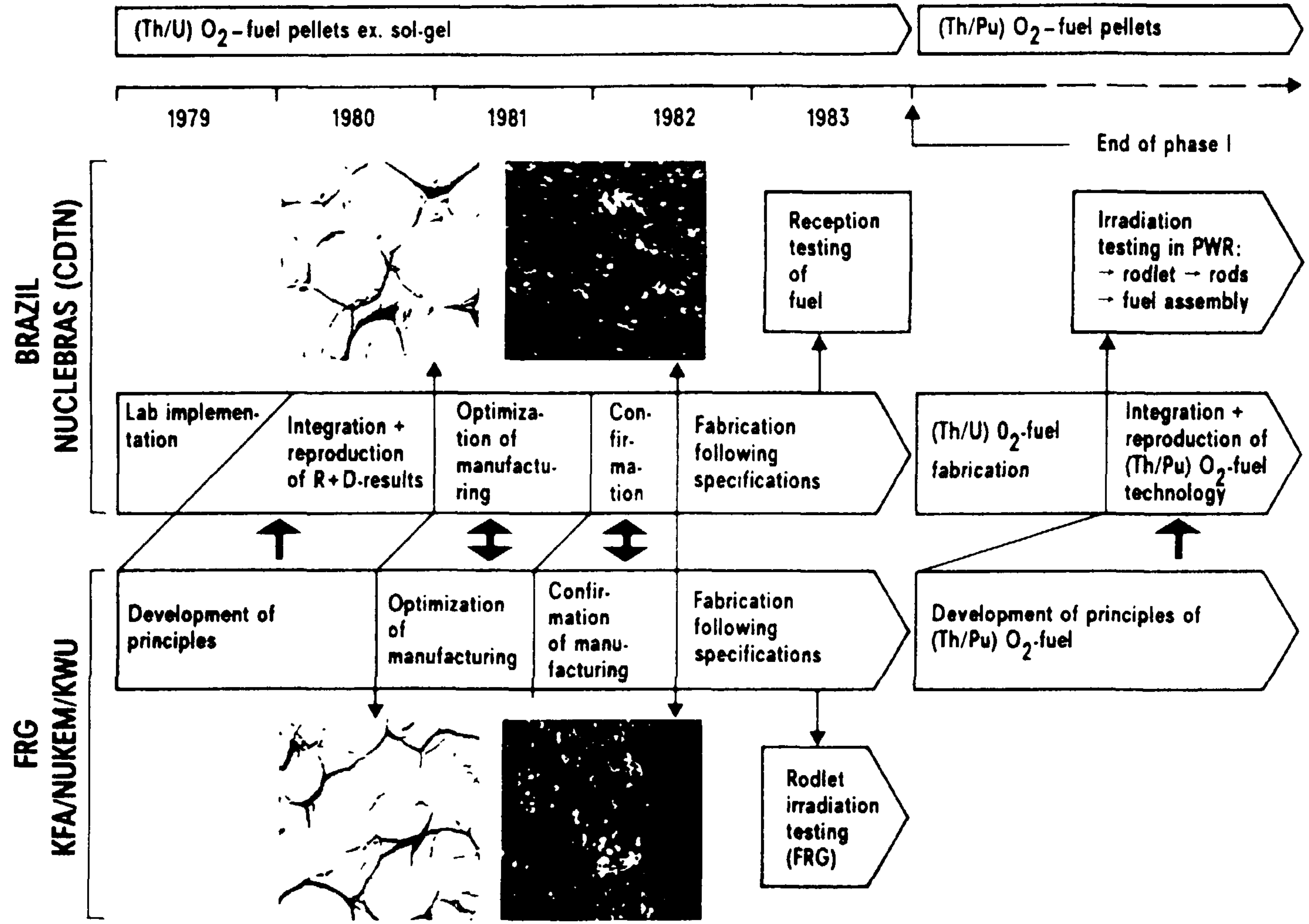


Fig. 5 Th-Utilization in PWR: Fuel Development and Fuel Fabrication

After completion of the $(Th,U)O_2$-fuel technology development the transfer of the gained knowledge to $(Th,Pu)O_2$-fuel production is initiated by the use of Ce as a Pu-simulator. Ce has proved to be an good Pu simulator under the aspect of its chemical, ceramurgical and physical properties / 13 /. The use of Ce as simulator allows for the development of the $(Th,Pu)O_2$ technology at a low cost basis. Only the final confirmatory investigation will use Pu. Fig. 5 comprises an overview on the strategy persued in fuel development and fuel fabrication.

5. Thermal Fuel Rod Design and Performance Prediction

Since the fuel assembly design and the linear heat rating of the Th-containing bundles are practically identical, thermal design work and performance prediction can be concentrated on the fuel rod analysis. The thermal conductivity for $(Th,U)O_2$ was determined also for the fuel with its typical microstructure. The results show that it is very near to the data known from UO_2 / 11 /.

To describe the fuel behaviour during burnup, the mechanistic densification-swelling model developed for UO_2 / 14 / has been adapted for thoria based fuel / 6 /. Also the CARO-code in its most recent version CARO D 5 / 15 / has been adapted also to describe in service fuel rod performance.

After completion of the code adaptation and having measured all necessary fuel properties, the thermal design and performance prediction were made for the test fuel rods currently under irradiation in the FRJ-2 at Jülich. The work resulted so far in a fairly good prediction for the beginning of life of the test fuel rods.

6. Irradiation Testing

The irradiation testing is performed in two complementary programs:

- an instrumented short length single pin irradiation in the experimental reactor FRJ-2 at KFA Jülich/FRG

- a segmented fuel rod and full length fuel rod irradiation in a host FA inserted into a commercial power reactor in Brazil.

The strategy behind these two branch program is to have a fast performing single rod irradiation, evaluating the burnup performance of the as-developed fuel and to generate the data base needed for licensing the pathfinder irradiation in the power reactor. The pathfinder irradiation provides the performance data under power reactor conditions. The single rods will be examined in a detailed PIE, whereas the pathfinder fuel rods will be inspected initially by available pool inspection techniques / 16 /. However, pool inspection is able to indicate the existence of any clad degradation, also from the inside, or the existence of defects if there are any. Thus, the results from both test series provide a reliable data base to assess the burnup performance of fuel pins with $(Th,U)O_2$ under power reactor conditions.

Table 3 gives an overlook on the single rod irradiation program and its actual status. The defect in the test LV 9.6-E 62 is due to a failure in the instrumentation.

Table 3 : Overview on FRJ-2 irradiation experiments
Values for E 61 in brackets were reached till 30 Sept. 84

Rig. No.	Test fuel rod (position)	Max. rod power from W/cm	till W/cm	Burnup GWd/tHM	Irrad. time d	PIE
LV9.6-E60	TT80 (top)	167	346	6,82	116,1	completed
	TDT81 (bottom)	391	430	8,22		
LV9.6-E61	TT82 (top)	111	234	8,2		in progress
	TDT83 (bottom)	278	308	10.0	176	
LV9.6-E62	TT84 (top)	112	235	1,5 *	34	in progress
	TDT85 (bottom)	276	300	1,26*		

* terminated after a defect in the instrumentation

Table 4 summarizes the pathfinder irradiation program. In order to investigate the operational behaviour and the dimensional stability of the fuel at different temperatures, the clearances between the fuel pellet and the cladding shall be varied within the full tolerance scale, i.e. in the range of 120 to 220 µm, in the segmented rods. Thereby first priority shall be given to the medium clearance of 170 µm. A full-length rod shall be irradiated to show the operational performance of a standard fuel rod with that type of thoria fuel.

To get information on the fuel performance in the whole burn-up range to 45 GWd/kg HM segmented fuel rods shall be irradiated for 1 to 4 operational cycles. This means, one segmented fuel rod shall always be un-loaded during the refueling shutdowns of the power plant. The full-length fuel rod shall be irradiated for altogether 4 operational cycles and finally be examined in the spent fuel pond of the power plant. Intermediate examinations are planned to be performed during each refueling shutdown if the time schedule for the refueling allows.

7. Investigation on the back end of the fuel cycle

For the most interesting case - the use of LWR recycling Pu as fissile material in Th fuel - worth mentioning savings are already achieved in the once through-put away cycle. Thus no reprocessing is needed for this fuel. But the long term storage of spent thoria fuel has to be still considered. Preferentially dry storage techniques will be used. Based on the excellent experience on the dry storage of spent UO_2 containing LWR-fuel / 17 /, no major problems should arise in this field. Only the different decay heats from $(Th,Pu)O_2$, in comparison to the UO_2 fuel, require an assessment at which periods after shut down dry storage may start in both versions: unconsolidated or consolidated manner. The problem areas of final disposal have not yet been addressed.

Using HEU as fissile material reprocessing is needed to achieve resource conservation (U- ore savings) and savings in the fuel

Table 4 : Overview on the pathfinder irradiation to be performed in ANGRA I
(Beginning with 3. reload 1988)

Test rod identification	**Test rod insertion period**			
	host fuel assembly No. 1			**host fuel assembly No. 2**
	1 cycle	**2 cycle**	**3 cycle**	**4 cycle**
Segmented fuel rod No. 1	X target burnup 11 GWd/tHM			
Segmented fuel rod No. 2		X target burnup 27 GWd/tHM		
Segmented fuel rod No. 3			X target burnup 33 GWd/tHM	
Segmented fuel rod No. 4				X target burnup 45 GWd/tHM
Full length fuel rod				X target burnup 45 Gwd/tHM

cycle cost. A scoping study on reprocessing showed the following results:

Cold dissolution experiments have shown that in the dissolution of $(Th,U)O_2$ fuels by Thorex solution, the Zircaloy clad is also dissolved to some extent and, besides this the dissolution time is 30 % increased if Zircaloy cladding is present. Nevertheless, final statements about dissolution behaviour of fuel with high burn up can only be made after hot experiments / 6 /. Those experiments are in progress.

For an optimization of the solvent extraction processes, distribution data covering the whole range of interest have been evaluated. Interpolations and extrapolations are possible by a computer program. Considerations about the readioactivity of reprocessed U-233 fuel from power reactors on the one side and the experience gained so far with the THOREX process variants on the other side lead to the proposal of a single-cycle THOREX process / 6 /.

8. Conclusions

The major results of the program work, confirmed in all related working areas, can be summarized so far:

- The standard PWR may use $(Th,U)O_2$-fuel and even $(Th,Pu)O_2$ fuel without any changes within the reactor system, within the core or in the fuel assembly design.
- Nearly the same performance of the reactor can be expected using $(Th,U)O_2$-fuel or $(Th/Pu)O_2$ as for of UO_2-fuel in a standard PWR nearly
- $(Th,U)O_2$-fuel can be easily manufactured within the same specification limits used for UO_2-fuel. A slightly modified ex-gel process from the HTR-technology as a chemical conversion is combined with the standard pelletizing, sintering and grinding process which is in commercial use for LWR-fuel fabrication. The development of $(Th,Pu)O_2$ fuel by means of Ce as simulator material is in good progress.

- Thermal and mechanical design of fuel rods containing thoria-based fuel can be done as well as with standard UO_2 fuel pins. The performance predictions made for the irradiation tests have been validated so far by the irradiation results.
- A favourable strategy to avoid the need of early reprocessing and to strive for worthmentioning savings in U-ore might be the use of $(Th,Pu)O_2$ fuel in a "once through-put away cycle" with extended burnups. Problems encountered with long term storage of spent Thoria-based fuel are very similar to that known from urania-fuel. It should not pose any extra problems.
- Thoria fuel may in the HEU-cycle require reprocessing to realize substantial savings in fuel cycle costs. Reprocessing of spent LWR-fuel with $(Th,U)O_2$-fuel is principally feasible using reprocessing schemes derived form experience gained in the context with HTR work and in combination with a chop-leach technique using the Thorex solvent.

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