

**NUCLEBRÁS**

Empresas Nucleares Brasileiras SA

**THE KFA/NUCLEBRÁS R+D PROGRAM ON  
TH-UTILIZATION IN PWR**

**NUCLEBRÁS/CDTN-441/82**

**July 1982**

**CENTRO DE DESENVOLVIMENTO DA TECNOLOGIA NUCLEAR**

**CAIXA POSTAL, 1941 - 30.000 - BELO HORIZONTE - BRASIL**



**KERNFORSCHUNGSANLAGE JÜLICH**  
GESELLSCHAFT MIT BESCHRÄNKTER HAFTUNG

**NUCLEBRÁS**

**Empresas Nucleares Brasileiras SA**

**THE KFA/NUCLEBRÁS R+D PROGRAM ON  
TH-UTILIZATION IN PWR**

<b>R. B. Pinheiro</b>	<b>NUCLEBRÁS</b>
<b>E. P. Andrade</b>	<b>NUCLEBRÁS</b>
<b>P. E. Cardoso</b>	<b>NUCLEBRÁS</b>
<b>V. Maly</b>	<b>KFA</b>
<b>W. Doerr</b>	<b>KWU</b>
<b>E. Wehner</b>	<b>NUKEM</b>

**Paper presented at the International  
ENS/ANS Conference  
New Directions in Nuclear Energy with  
Emphasis on Fuel Cycles  
held in Brussels, Belgium,  
28th-30th, April 1982**

**NUCLEBRÁS/CDTN-441/82  
Belo Horizonte - Brazil  
July 1982**

## CONTENTS

	Page
INTRODUCTION . . . . .	1
STATUS OF WORK . . . . .	2
<i>Strategy and Nuclear Core Design Calculations</i> . . . . .	2
<i>Fuel Fabrication</i> . . . . .	4
<i>Fuel Performance Prediction and Qualification</i> . . . . .	6
<i>Reprocessing Studies</i> . . . . .	7
CONCLUSIONS . . . . .	8
REFERENCES . . . . .	8

## THE KFA/NUCLEBRÄS R+D PROGRAM ON TH-UTILIZATION IN PWR

### INTRODUCTION

The utilization of thorium fuel in thermal reactors, due to the high  $\eta$  value of U-233, brings about better resource utilization compared with conventional uranium fuel. This particularly holds for advanced reactor types specifically designed for thorium application, but in light water reactors (LWRs) also, a substantial advantage is anticipated for a closed fuel cycle. The closing of the fuel cycle is a prerequisite for any long-term use of thorium in thermal reactors. The quickest and easiest way to demonstrate the feasibility of the complete fuel cycle is to prove this on first generation commercial reactors.

To utilize the available know-how and research capacities, a joint cooperative program was initiated between KFA and NUCLEBRÄS, with the participation of KWU and NUKEM on the German side. The objectives of the program, which has run since mid-1979, are as follows:

1. to analyze and evaluate thorium utilization in pressurized water reactors (PWRs)
2. to design the PWR fuel element and core for the thorium fuel cycle
3. to manufacture, test, and qualify Th/U and Th/Pu fuel elements under operating conditions
4. to study the closing of thorium fuel cycle by reprocessing of spent thorium-containing PWR fuel elements.

The project utilizes as much as possible existing technologies and available know-how for core design, fuel element fabrication, and reprocessing. Therefore, the existing techniques and equipment for the fuel cycle in high-temperature reactors (HTRs), i.e., manufacturing and reprocessing of fuel, as well as for the fuel cycle

in LWRs, i.e., fabrication and head-end treatment, will be applied. Advanced methods and processes (pellet fabrication from fuel kernels) will render an essential potential for employment. In the long term, the program could be extended to incorporate advanced methods comprising all steps of the fuel cycle, such as the fuel design, manufacture, and disposal.

#### STATUS OF WORK

The first activity comprised a review of the accessible literature about thorium utilization in LWRs (Ref.1). The evaluation has shown that a good base of knowledge is available for fuel characterization methods and, to a lesser extent, for the general behaviour of Th/U mixed-oxide fuel under irradiation. No statements can be made with regard to the properties depending directly on the manufacturing process, such as fission gas release, pellet-cladding interaction (PCI), accident behaviour, etc.

*Strategy and Nuclear Core Design Calculations.* Investigations on the thorium fuel cycles in PWRs have been performed with the same theoretical methods as are used for the U/Pu fuel. An outstanding task in the initial program phase was the modification and validation of standard PWR-related codes for calculation of thorium-based fuel.

Testing was done by analysis of Brookhaven National Laboratory exponential experiments.<sup>2,3</sup> When comparing the calculated and measured ratio of epithermal to subthermal capture in Th-232, we see that the results of the Th-FASER code agree within the uncertainty limits, especially in the region of moderator-to-fuel ratio of main interest. The calculation of  $k_{eff}$  with measured buckling showed an underprediction of 1 to 2%. We follow the opinion of Ref.3 that this is caused mainly by experimental data and by zero-dimensional calculations. Further improvements require explicit calculation of resonance interferences. Work in this direction should be done in parallel with nuclear design work using Th-FASER. The present version of Th-FASER has been used to generate two-group libraries for MEDIUM core calculations.

The three-batch Th/HEU once-through and self-generating recycle (segregated and mixed) reload schemes for a KWU 1300-MW(e) standard PWR have been investigated. Calculations for a four-batch core have also been carried out. The overall core calculations have confirmed that no modifications need to be done for thorium fuel when incorporated in a standard PWR fuel element. For the Th/Pu cycle, only the once-through reload scheme in a three-batch full Th/Pu core has been investigated up to now. Tables I and II summarize the information about the fuel cycles. First results show that a full Th/Pu core can be realized with standard PWR fuel elements. Preliminary calculations related to the safety analysis of both Th/HEU and Th/Pu, based in reactivity balance calculations, have shown sufficient control margin.

TABLE I  
Characteristics of Once-Through Fuel Cycles

		Th/HEU-Fuel 3 batch core	Th/Pu-Fuel 3 batch core
Enrichment of first core	w/o U-235 or w/o $Pu_{fiss}$	2.9 (69 FAs) <sup>a</sup> 3.6 (68 FAs) 4.2 (64 FAs)	2.8 (65 FAs) 3.6 (64 FAs) 4.4 (64 FAs)
	w/o U-234 or w/o $Pu_{fiss}$	4.2 (64 FAs)	4.4 (64 FAs)
	Boron concentration First core (BOC) Equilibrium core (BOC)	ppm 1461 1177	2754 1374
Cycle length	First core	330	317
	Equilibrium core	290	267

<sup>a</sup>FA = fuel assembly.

TABLE II  
Fuel Composition of Burnup-Equivalent Once-Through and First Recycling Generation Fuel Assemblies (FA's)

Th-HEU, Once-through Fuel			Th-U, Segregated Recycling Fuel		Th-U, Mixed Recycling Fuel	
	Loading w/o	Unloading w/o	Loading w/o	Unloading w/o	Loading w/o	Unloading w/o
Th 232	95.48	95.34	94.82	96.28	95.85	95.98
U 233	-	1.46	1.89	1.69	1.24	1.68
U 234	-	0.17	0.23	0.33	0.15	0.33
U 235	4.20	1.15	1.86	0.83	2.68	0.83
U 236	-	0.55	0.79	0.81	0.52	0.81
Sp 237	-	0.04	-	0.00	-	0.00
U 238	0.32	0.28	0.42	0.34	0.38	0.34
Reload batch n = 44, 4.2 w/o U235 (HEU)			Reload batch: 44 FA of 3.75 w/o U <sub>235</sub> rec. + 20 of 4.2 w/o U235 (HEU)		Reload batch n = 44, 3.905 w/o U <sub>235</sub>	
Burn up: 35.5 MWd/kg <sub>HM</sub>			Burn up: 33.9 MWd/kg <sub>HM</sub>		Burn up: 34.6 MWd/kg <sub>HM</sub>	
Th-Pu-fuel cycle			Th-Pa/U <sub>rec</sub> fuel without adding Pu			
Th/Pu fuel, Pu extracted from U-FA			make up extracted from U-FA			
	Loading w/o	Unloading w/o	Loading w/o	Unloading w/o		
Th 232	93.93	95.82	93.51	95.11		
U 233 + Pa 233	-	1.32	1.99	2.18		
U 235	-	0.20	0.26	0.34		
U 236	-	0.82	0.87	0.88		
U 238	-	-	-	-		
Pu 239	3.65	0.64	0.90	0.10		
Pu 240	1.41	1.12	1.66	0.75		
Pu 241	0.75	0.74	1.09	0.60		
Pu 242	0.25	0.44	0.67	0.88		
Burn up 34.6 MWd/kg <sub>HM</sub>			Burn up: 33.8 MWd/kg <sub>HM</sub>			

**Fuel fabrication.** For the fabrication of (Th,U)<sub>2</sub>O<sub>7</sub> fuel pellets, a combination of two fully developed fabrication routes is used: manufacturing of (Th,U)<sub>2</sub>O<sub>7</sub> microspheres by the method of external gelation, and pellet fabrication by the method of direct pelletizing.

The ex-gel conversion has been developed and is now used for the production of HTR fuel.<sup>4</sup> The direct pelletizing as an economically and technically advanced process is utilized in the manufacturing of uranium-based LWR fuel.<sup>5</sup> Both processes have a high degree of standardization and show a broad basis of production experience. The combination of both is used for Th-based LWR fuel production according to the flowsheet in Fig. 1.

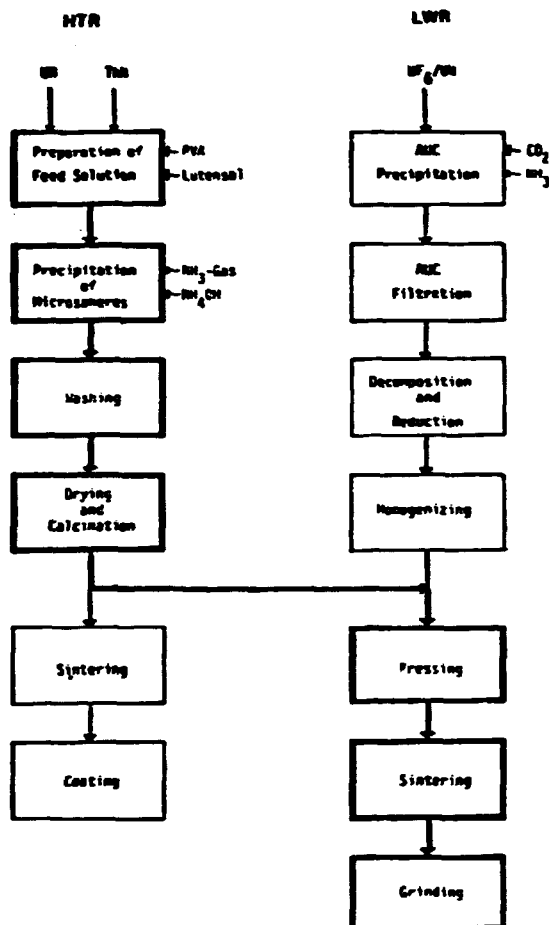


Fig. 1. Combination of available technologies for the manufacturing of mixed-oxide fuel.

The ex-gel fabrication leads to  $(Th,U)O_2$  microspheres, some properties of which are very advantageous for the direct pelletizing, as was shown in laboratory-scale investigations:

1. The two oxides are in a coprecipitated state; that means that there is no disturbing influence of the formation of solid solution during sintering, as is observed sometimes in mechanically mixed oxides.
2. Form, size, and morphology of the microspheres guarantee an easy handling (excellent flowability, for example).
3. The sinterability of the microspheres is very good.
4. The dust generation during processing is low.



To reach the required pellet quality, the adaptation of some of the microsphere's properties and, to a lower extent, of the pelletizing parameters also is necessary. This relates mainly to the adjustment of a suitable pellet microstructure, which requires a lowering of the generally high mechanical strength of the microspheres. By optimizing the heat treatment after washing, a sufficiently low mechanical strength was obtained. In addition, an adaptation of the precipitation conditions has been performed to assure a low content of residual organic material in the press-fit microspheres; beyond that, a special low-temperature treatment before drying has been developed for the same reason.

With respect to the pressing of microspheres, no essential changes of the highly standardized LWR pressing process were found to be necessary. A good lubrication of the die wells has to be provided for. The use of lubrication additives admixed to the kernels has a beneficial effect on the green and sintered density, but is not adequate for a good microstructure. Higher pressing forces and, consequently, higher green densities than in  $UO_2$  production have been found to be suitable for sufficient sintered density and good microstructure. Because of the high sinterability of the kernels, reduced sintering temperature of  $\sim 1200$  to  $1400^\circ C$  are sufficient. As a sintering atmosphere, reducing, inert, and even oxidizing gases, e.g., air, can be used. This is due to the excellent phase stability of  $(Th,U)O_2$  up to 50 mol%  $UO_2$  and at high temperature. These investigations resulted in the laboratory scale fabrication of  $(Th,U)O_2$  pellets that fulfill all LWR-specific requirements.

*Fuel Performance Prediction and Qualification.* For the prediction of the irradiation behaviour, a consistent data set for  $(Th,U)O_2$  has been developed on the basis of a literature review. Most of the data are available, or can be calculated from the values of pure  $ThO_2$  and  $UO_2$ . Physical and thermal properties of  $(Th,5\%U)O_2$  solid solutions were developed for adapting them to the CARO computer code. Models have also been developed for  $ThO_2$  and  $UO_2$  elastic moduli. The dimensional behaviour of  $(Th,5\%U)O_2$  fuel has been calculated on the basis of the densification model described

in Ref.6. The results are as follows:

1. The swelling rate of  $(\text{Th,U})\text{O}_2$ , is ~ 50% higher than that of pure  $\text{UO}_2$ .
2. The prediction of densification behaviour is not yet very reliable, but there is an indication of a smaller densification rate in  $(\text{Th,U})\text{O}_2$  than in pure  $\text{UO}_2$ .

These results confirm the former qualitative statement that the pore structure of the ex-kernel should be optimized. CARO code calculations on  $(\text{Th,U})\text{O}_2$  fuel show the following:

1. The maximum temperature of  $(\text{Th,5\%U})\text{O}_2$  PWR fuel is expected to be more distant from the melting point ( $3250^\circ\text{C}$ ) than that of  $\text{UO}_2$  fuel ( $2800^\circ\text{C}$ ).
2.  $(\text{Th,5\%U})\text{O}_2$  fuel rods have less stored energy than  $\text{UO}_2$  rods under corresponding conditions.

An in-pile fuel qualification will be performed by an irradiation test of six  $(\text{Th,U})\text{O}_2$  fuel rods, fabricated under pilot plant conditions, in FRJ-2 in Jülich. Start of irradiation is envisaged for the spring of 1983.

*Reprocessing Studies.* A literature search of all aspects of the back end of the thorium-uranium fuel cycle has been completed. The progressing evaluation of the literature indicates lack of knowledge in some areas, particularly in the head end. Cold laboratory tests in dissolution and solvent extraction are proceeding.

The dissolution experiments with cold  $(\text{Th,U})\text{O}_2$  pellets from an early development stage have shown a high dissolution rate of this fuel in Thorex solution. The dissolution of Zircaloy cladding tubes by Thorex solution has been identified as a main specific problem of reprocessing  $(\text{Th,U})\text{O}_2$  LWR fuel, since the expected zirconium concentration in the feed solution is about three times higher than of the  $\text{UO}_2$  fuel.

## CONCLUSIONS

The cooperative Brazilian/German thorium utilization in PWRs program creates a technological basis for industrial application of thorium fuel cycles in PWRs. By choosing an advanced fuel fabrication route that contains several features favorable for refabrication purposes, a spinoff for other cycles as well can be expected.

By 1983, a statement toward technological feasibility of Th/U ex-kernel fuel will be available. After that, an investigation of more detailed aspects connected with the reactor-related fuel performance and investigation of technological feasibility of Th/Pu fuel, as well as hot reprocessing experiments, will follow. The final goal is a fuel element test in a currently running PWR.

## REFERENCES

1. NUCLEBRÁS/CDTN 424/80, KFA-HBK-IB-2/80.
2. H.H.WINDSON, W.J.TUNNEY, and G.A.PRICE, Nucl. Sci.Eng., 42, 150 (1970).
3. EPRI-NP 359, Electric Power Research Institute.
4. M.KADNER, and J.BAIER, Kerntechnik, 18, 413 (1976).
5. H.ASSMANN and M.BECKER, Trans. Am. Nucl. Soc., 31, 147 (1979).
6. H.ASSMANN and H.STEHLE, Nucl.Eng. Des., 48, 1, 49 (1978).