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PROGRAM of RESEARCH and DEVELOPMENT on the THORIUM UTILIZATION in PWRs

FINAL REPORT for PHASE 1
(1979 – 1983)

MAY 1984

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The major results of the Phase 1 (1979 - 1983) of the program "Th-Utilization in PWRs" 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 pellet pressing out of ex-gel microspheres has been developed on laboratory scale. The $(Th,U)O_2$ test fuel produced satisfies PWR-specifications and is currently undergoing irradiation testing. 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. Based on the results of the Phase 1, recommendation for program continuation has been formulated.

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The report presents a joint activity of the program partners KFA, KWU, NUCLEBRÁS/CDTN and NUKEM. The main contributors have been for chapters 0, 1, 3, and 4 Dr.V.Maly, KFA, and Dr.R.B.Pinheiro, NUCLEBRAS, for 2.1 Dr.M.Peehs, KWU, 2.2 Dr.J.G.Schlosser and Dr. H.Finnemann, both KWU and Mr.E.P.Andrade and Dr.F.A.N.Carneiro, both NUCLEBRAS, 2.3 Dr.W.Dörr, KWU, 2.4 Dr.E.Wehner, NUKEM, 2.5 Mr.F.S.Lameiras, NUCLEBRAS, 2.6 Mr.K.Reichardt, KFA, and 2.7 Dr.E.Zimmer, KFA, and Miss M.L.Soares, NUCLEBRAS. Chapter 5 has been jointly formulated during the concluding 8. Program Management Meeting in Nov.1983. Many others have participated by providing materials or comments.

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0. SUMMARY

0.1 The Program

The utilization of thoria fuel in thermal reactors can result in higher conversion ratios and in better resource utilization compared with the uranium fuel in closed fuel cycle. However, before considering the use of these advantageous features a proof of the technological feasibility is required.

Within the frames of the "Governmental Agreement on Cooperation in the Field of Science and Technology" between Brazil and F.R. Germany (1969) and the "Memorandum of Understanding between Kernforschungsanlage Jülich GmbH-KFA and Empresas Nucleares Brasileiras S.A.-NUCLEBRÁS" (1978), the cooperative R&D program on "Thorium Utilization in Pressurized Water Reactors", between NUCLEBRÁS/CDTN on the Brazilian side and KFA with the participation of Kraftwerk Union A.G.-KWU and NUKEM GmbH on the German side, aims since mid 1979:

- to analyse and prove the Th utilization in PWR;
- to design the PWR fuel element and core for the Th-fuel cycle;
- to manufacture, test and qualify Th/U and Th/Pu fuel elements under operating conditions;
- to study the closing of Th-fuel cycle by reprocessing of spent Th-containing PWR fuel elements.

Three program phases have been foreseen:

Phase 1 (1979 - 1983) where methods and technologies were adapted to PWR thoria fuels;

Phase 2 where the $(Th,U)O_2$ fuel behaviour shall be demonstrated in a current PWR; and

Phase 3, devoted to the demonstration of the $(Th,Pu)O_2$ fuel.

Demonstration of the closing of the fuel cycle is, nevertheless,

beyond the program scope.

The existing technology and available know-how for fuel element and nuclear core design, fuel element fabrication and reprocess-

beyond the program scope.

The existing technology and available know-how for fuel element and nuclear core design, fuel element fabrication and reprocessing are utilized by the program, as is the case with fuel manufacturing and reprocessing, from HTR, and with fuel fabrication and head-end treatment, from LWR.

The program has been funded by KFA and NUCLEBRÁS, directed by a joint Management and supervised by Coordinating Committee, both formed by NUCLEBRÁS and KFA with the participation of the German industry partners. Subdivided into 13 tasks with different sub-tasks, Phase 1 reached nearly all planned results while stretched from the original 3 to 4 1/2 years by budget restrictions. The aspects of the transfer of technology have been duly considered and resulted in an increased level of cooperation.

0.2 Status of Knowledge - $(Th,U)O_2$ Use in Power Reactors

The goal of the international literature survey was to determine the status of irradiation experience with $(Th,U)O_2$-containing PWR fuel elements. As far as fabrication processes are concerned, the various fuel element fabrication methods can use fuel in the form of powder, claylike material, angular particles and microspheres prepared by arc-fusing, standard pellet from powder, and e: techniques. The fuel is filled into the tubes by different routes. Particle fuel is prepared by dry or wet chemical route.

With regard to irradiation testing, in-pile behaviour of $(Th,U)O_2$-containing fuel rods fabricated by pelletizing, vipac or sphere-pac, have been investigated in PWR with favorable first results.

As an overall conclusion it was established that the project can use to a large extent the existing technologies and available know-how for fuel element fabrication and reprocessing.

0.3 Strategy Calculations

This task aimed at analysing the incorporation of Th-fuelled PWRs in the nuclear-electric demand forecast, based on data from the nuclear core design calculations and making use of the KWU fuel cycle cost and strategy analysis code FUKOMA.

Originally written for utilization in the U/Pu fuel cycle, the strategy calculation part of FUKOMA had to be improved to accomodate calculations including reactors operating in the Th-fuel cycle.

An important conclusion reached is the confirmation that substantial resource savings can be obtained in the case of applying closed Th-fuel cycles to standard PWR.

0.4 Nuclear Core Design

The work has concentrated on the utilization of thorium-highly enriched uranium (Th/HEU) and thorium-plutonium (Th/Pu) in an unmodified 1,300 MWe PWR. For this purpose the KWU computer codes MEDIUM-2.2 and FASER-3 were employed directly or with some modifications (Th-FASER) to accomodate thorium fuels.

Several options of Th-fuelled PWR operating in once-through and recycling modes were analysed. (In the particular case of Th/Pu cycle, Pu comes from the reprocessed fuel of a standard PWR operating in the uranium once-through mode). The calculations consisted in the determination - for a desired cycle length - of some reactor parameters for the initial and reload cores: fuel enrichments, core configurations, isotopic compositions, power form-factors, reactivity coefficients and shutdown margins.

As an overall conclusion from the nuclear design it has been stated that the introduction of standard PWR fuel assemblies, Th/HEU- or Th/Pu fuelled, in unmodified PWR presents no technical problems precluding the adoption of any of the options analysed.

Further work will be needed to analyse some detailed aspects of the core design, such as the transition from uranium to thorium cycles, modified fuel assemblies (with different rod diameter and lattice pitch), as well as aspects of accident analysis.

0.5 **$(Th,U)O_2$ - Fuel Production and Quality Assurance**

The goal of this activity was the development of a production process for Th/U-PWR-fuel, together with a system of quality control, and the transfer of both to NUCLEBRÁS for laboratory-scale manufacturing.

For fuel production, a combination of two existing fabrication processes has been realized: $(Th,U)O_2$-microsphere production by external gelation, as is used in THTR fuel technology, and the direct pelletizing process (pressing and sintering) as the commc way for U-base LWR fuel manufacturing from UO_2-ex-AUC powder. The adaptation of both processes to each other could be performed mainly by adjusting the microspheres properties to the requirements of direct pelletizing and less by changing the pressing and sintering procedures.

The status of the development work permitted to establish a suitable laboratory scale production line. For an irradiation test in the FRJ-2 research reactor $(Th,U)O_2$ pellets and rodlets have been manufactured and all procedures for quality control have been settled and applied.

The transfer of the available know-how to NUCLEBRÁS resulted in the implementation of a lab-scale production line. This has successfully been proven by a parallel fuel manufacturing under the same conditions as in the F.R.G. irradiation test fuel production. Further on, NUCLEBRÁS was able to perform own R&D work in the last part of Phase 1 of the Program.

0.6 Irradiation Test Rod Production

The target of the fuel fabrication was to provide a fuel rod with properties representative of the latest $(Th,U)O_2$ fuel development program results.

As a prerun to the irradiation test batches, kernel batches were manufactured, pelletized and sintered under specified conditions, and used for characterization with almost all results within the tolerance limits. Then the irradiation test fuel was produced under the standard conditions.

For cladding material shortened Zircaloy-4 rods were selected, with fabrication parameters and dimensions correspondent to typical PWR rods. A list of quality requirements was compiled as a fundamental condition for fuelling and final treatment of these rods. For fabrication TIG welding was used.

Three prerun rods were fabricated with $(Th,U)O_2$ pellets from the prerun pellet fabrication; eight test rods were then manufactured. Examinations were carried out and documented; additional quality control examinations, including destructive characterization methods applied to parallel samples, produced results documented within the required quality.

0.7 Design and Performance Prediction of Test Fuel Rods

This activity comprised a literature survey on the relevant properties of $(Th,U)O_2$, a model development related to the behaviour of the mixed thorium-uranium oxide under irradiation, the implementation of this model to the CARO-D3 fuel rod computer code and the calculations with regard to the design of the 95% Th, 5% U mixed oxide fuel rods for the irradiation testing in the FRJ-2 reactor.

In the literature survey relevant properties of $(Th,U)O_2$ were treated: theoretical density, thermal conductivity and expansion, melting point and molar thermal capacity. In the model development, the behaviour of the mixed oxide concerning densification, swelling, fission gas release, restructuring and relocation have been considered. CARO-D3, a computer code for design and performance prediction of UO_2, UO_2/PuO_2 and UO_2/Gd_2O_3 fuel rods, was then implemented to accomodate the $(Th,5\%U)O_2$ fuel, incorporating models previously developed. Calculations with the new version Th-CARO code were then performed in order to design fuel rods for the irradiation tests. Preliminary calculations for zero burn-up and for irradiation under the FRJ-2 conditions showed results within some expected limitations and helped to set some further boundary conditions.

Regarding the test fuel rod, general criteria guided the best-estimate and design calculations. Main results established that:

- in view of the Th-CARO code calibration both test rods should be instrumented in the irradiation capsule with thermocouple;
- the densification and amounts of fission gas released are the most important questions to be answered by first tests;
- the test rod power level should be 200-300 W/cm with no ramp and the maximum burnup should be 15 MWd/kg U+Th.

Some sensitivity calculations were also performed with the Th-CARO code, in order to study the influence of parameters subjected to uncertainties. The main result of these calculations states that the uncertainty in thermal conductivity is the most important parameter concerning the fuel rod performance prediction.

0.8 **Irradiation Testing**

Three irradiation experiments will be performed with two test fuel rods each in a boiling water loop of the research reactor FRJ-2 at Jülich, in order to test the behaviour of the newly developed fuel, to demonstrate its qualification and to measure its basic properties. The fuel rod design, fabrication, performance prediction, etc., is a common effort of all parties involved in the Program. The irradiation concludes the Program Phase 1, but overlaps with the work of the Phase 2A.

The aim of the first 3 irradiations with reduced burn-up is to get information about the suitability and the properties of the new fuel, to support the irradiation in power reactors and to estimate its safety risk. The following features shall be studied: residual and fission gas release and pressure build up, restructuring of the fuel, dependence of fuel temperatures on power and burn-up, fuel-clad interaction and material transport in the fuel.

Each fuel rod contains 25 pellets of Th/HEU oxide of PWR-standard dimensions with Zircaloy-4 cladding, in total 275 mm of active length. Both test rods are instrumented and fitted one above the other into the irradiation rig at KFA-ZAT. The lower rod is positioned symmetrically in the neutron flux profile, while the upper rod meets a decreasing flux from its bottom to top.

For irradiation, the rig is put into a vertical channel of the reactor core and connected to the boiling water loop LV9. The power produced by the test fuel rods is measured by means of the cooling water heating and flow; fuel temperatures, pressure and other irradiation data are continuously processed by a computer.

The three irradiations are planned to take place in series of 3 to 9 reactor operation periods each, performing 55, 200 and 130 days, respectively. Results of the post-irradiation examinations (PIE) concerning the first irradiation experiment should be available before starting the third one.

Pre-irradiations examinations have been done. On November 25, 1983 the first irradiation experiment started and first results have been obtained.

0.9 Reprocessing Studies

Cold dissolution experiments have shown that in the dissolution of $(Th,U)O_2$ fuels by Thorex solution, the Zircaloy hull 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.

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 radioactivity of reprocessed U-233 fuel from power reactors on the one side and the experiences gained so far with the THOREX process variants on the other side lead to the proposal of a single-cycle THOREX process. Problems with crud formation during re-extraction can be safely avoided if this step is carried out first as separate thorium re-extraction (partitioning) with 0.5 M HNO_3 strip solution followed by uranium stripping with 0.01 M HNO_3.

Pulse columns should be preferred as extraction apparatus because this equipment produces less TBP degradation products due to the shorter contact time. In addition, pulsed columns are less sensitive against process disturbances by any crud formation. For extraction a continuous organic phase, for re-extraction a continuous aqueous phase operation in pulsed columns should be selected.