

## SOME PECULIARITIES OF THORIUM DIOXIDE-BASE FUEL PRODUCTION AT NSC KIPT\*

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**Abstract.** The paper describes the status of work on pyrocarbon-binder spherical fuel elements with thorium dioxide-base fuel for HTGR. Basic flow charts of manufacturing fuel microspheres, coated particles and spherical fuel elements are described. Results of investigations into the main characteristics of fuel elements and their constituents, including their operation under reactor irradiation conditions, are discussed. Some special features of the technology of pyrocarbon-bound (U,Th)O<sub>2</sub> - base spherical FE are presented and reports the results from studies of their main characteristics.

### INTRODUCTION

Over a few decades, the development of high-temperature gas-cooled reactors (HTGR) has been an active field of research activities in nuclear power. These reactors are advantageously distinguished from reactors of other types by their ability to generate simultaneously both electrical and thermal energy, and also by high safety, an economical fuel cycle, a high efficiency (~ 40%), etc. The HTGR can be operated with both uranium and mixed U-Pu or U-Th fuel cycles or their combinations, and the uranium-thorium cycle appears to be most economical. The decisive argument in favour of the thorium cycle is the possibility of long-term providing the nuclear energetics on the whole with necessary breeding material. The combined use of uranium and thorium cycles assures the provision of fuel resources at moderate and stable costs.

In HTGR designs developed in the former Soviet Union and FRG, the use was made of the principle of pebble-bed core with graphite fuel elements, 60 mm in diameter, in the central part of which (kernel) the fuel is dispersed as coated particles (CP), while the periphery region (element jacket) is free of the fuel. The CP are the spherical particles of thorium and uranium carbides or oxides coated with protective layers of dense pyrocarbon and silicon carbide. The advantages of such fuel elements (FE) lie in their high radiation resistance and good fission product retention that enable the attainment of high nuclear fuel burnup levels (up to 100 000 MW·24h/t and over) at a high operating temperature.

In the technology of spherical uranium-graphite fuel element production one can recognize the following three basic processes: (i) fuel microsphere (FM) production, (ii) coated fuel particle production, (iii) spherical FE production.

In the world practice there are three institutions (NUKEM - Germany, NSC KIPT - Ukraine [1], R&PA "Luch"- Russia [2]) known as the main developers of the spherical uranium-graphite FE processes. The first two processes are based on pressing the product billets by the known graphite-production electrode methods. The NSC KIPT technology has no foreign analogs as, instead of pressing, it makes use of the procedure of product billet molding followed by product densification with pyrocarbon deposited from gaseous hydrocarbons on

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\* 1998 meeting.

heated surfaces [1]. The NSC KIPT spherical FE have different design modifications, where fuel of different types ( $\text{UO}_2$ ,  $(\text{U,Th})\text{O}_2$ , UC, UCN, etc.) can be used.

## FUEL MICROSPHERES

By now, a list of basic requirements on FM for spherical HTGR FE has been elaborated [3]. In particular, the FM must be uniform in size (500 - 50  $\mu\text{m}$ ), and their sphericity coefficient ( $d_{\text{max}}/d_{\text{min}}$ ) should not exceed 1.05. The total free volume in both the FM and the buffer CP layer is in many respects responsible for the admissible fuel burnup and operating temperature of the fuel. It has been established by theory and experiment that the total porosity must be about 2 to 4 % per 1 % of heavy nuclei burnup at temperatures between 1300 and 1500 °C [4]. To provide the volume for collecting gaseous fission products (GFP) and solid fission products (SFP) in the FM, it appears more preferable, in our opinion, to follow the way of reducing the FM density rather than increasing the buffer CP layer thickness. Therefore, the FM density has been chosen to be about 85 % of theoretical density (TD), this is ensured at stages of "raw" billet manufacture and heat treatment of FM.

To make FM, the NSC KIPT team has developed the method of mechanical spheroidization of fuel billets made from plastified masses [5]. Though this method ranks below the sol-gel process in productivity, yet it is distinguished for its adaptability at the stage of experimental development of FM with different fuel compositions as the basis, and the process of FM production (to the sintering stage), as such, is wasteless.

The essence of the method consists in manufacturing and spinning cylindrical fuel billets from plastified masses until perfect microspheres are produced, which are then sintered at high temperature in vacuum.

Powders of uranium dioxide (enriched in uranium-235 up to 21, 36, 90 %) and thorium dioxide (natural) were used in experiments. The thorium-to-uranium ratios were 3:1 and 9:1. The required powder mixture was prepared in the planetary-type centrifugal mill. Specially made metallic thorium balls, 12 mm in diameter, were used as mixing and milling elements. The prepared powder mixture was mixed with 12 wt. % plasticizer based on paraffinum and petrolatum (65:35) at 70 °C in a mixer.

A special device was used to make from the plastified mass cylindrically-shaped sized billets with the H/D ratio of about 1.0 to 1.2. These billets were spinned in the "Spheroidizer" facility until spherical particles were produced. The nonsphericity of "raw" particles was about 1.02, the variation in size was less than 50  $\mu\text{m}$ .

The heat treatment of the obtained fuel kernels was performed in two stages. At the first stage, the plasticizer was distilled off in vacuum. To retain the shape, the "raw" fuel kernels were heated in the aluminum oxide powder filling to a temperature of about 400 °C. At the second stage, the fuel kernels, separated from the filling, were sintered in the rotating container in a vacuum furnace at a temperature of 2000 °C. The density of microspherical fuel kernels after sintering was found to be 75-85 % TD. The fuel kernel  $(\text{Th,U})\text{O}_2$  structure is shown in Fig. 1. During the process of sintering, the chemical composition of mixed  $(\text{Th,U})\text{O}_2$  kernels remained the same.

Table I gives some characteristics of pilot batches of microspherical  $(\text{Th,U})\text{O}_2$  particles.

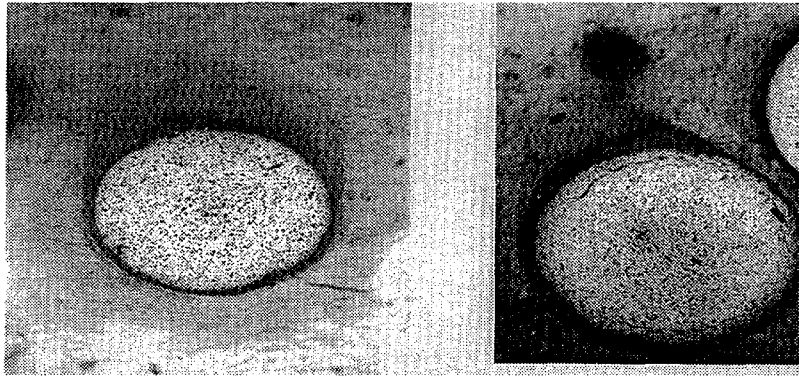


Figure. 1 Structure of fuel kernels  $(Th,U)O_2$  ( $\times 100$ ).

Table I. Some characteristics of pilot batches of microspherical  $(Th,U)O_2$  particles.

1	Batch number	Sintering conditions		Diameter, mkm	Density		Oxygen Coefficient
		Temper, K	Time, h		g/cm <sup>3</sup>	% TD	
1.	69-36-1-84	2280	0,7	450-650	9,72	85	1,99
2.	70-21-1-87	2280	0,5	450-650	7,55	75	-
3.	01-90-1-87	2273	0,5	580-640	7,8	77	-
4.	02-90-1-87	2273	0,5	800-850	8,0	80	1,99
5.	03-90-1-87	2273	0,5	620-680	7,86	78	2,00

## COATED FUEL PARTICLES

To deposit several protective coatings onto FM, the well-known "boiling layer" method is used at NSC KIPT. The technology devised to produce CP differs from analogs abroad by the type of gases used and by the conditions of protective layer deposition [6]. In particular, instead of inner and outer dense PyC layers, here the combined (PyC+SiC) coatings of  $\sim 2.4\text{g/cm}^3$  are used. This substitution has allowed us to reduce the GFP yield from CP irradiated in the free-fill state by factors of 5 to 10.

As the process of CP production was developed, various CP designs were created:

- type I - PyC-SiC-PyC,
- type II - (PyC+SiC)-SiC-PyC,
- type III - (PyC+SiC)-SiC-(PyC+SiC),
- type II\* (pilot) - (PyC+SiC)-SiC.

## SPHERICAL FUEL ELEMENTS

To perform a combination of reactor tests of  $(Th,U)O_2$  spherical FE, the last ones were fabricated by the process of volumetric gas-phase densification of porous bodies with pyrocarbon in the form of model spherical fuel elements (45 mm in diameter) [1].

Table II. Reactor test results for uranium-thorium fuel

№	CP batch number	Kernel material	Enrichment in $^{235}\text{U}$	Kernel density $\text{g/cm}^3$	Kernel diameter, mkm	Coating material	Coating density, $\text{g/cm}^3$	Coating thickness (mkm)	Reactor, channel	Irradiation temperature ( $^{\circ}\text{C}$ )	Neutrons fluence $E > 0,1 \text{ MeV}$ , $\times 10^{20} \text{ cm}^{-2}$	Burnup, % fima	R/B in $^{88}\text{Kr}$	R/B in $^{133}\text{Xe}$
1	2	3	4	5	6	7	8	9	10	11	12	13	14	15
1.	21-10-X-87	(3Th,1U)O <sub>2</sub>	21	7,55	550±100	PyC PyC PyC+SiC SiC	1,1 1,5 2,4 3,16	42 10 56 70	SM-2, №15	1250 1500	0,8 1,4	0-2,1 2,1-3,5	8,1·10 <sup>-6</sup> 1,0·10 <sup>-4</sup>	1,1·10 <sup>-5</sup> 1,0·10 <sup>-4</sup>
2.	36-12-X-84	(3Th,1U)O <sub>2</sub>	36	9,72	550±100	PyC PyC PyC+SiC SiC PyC+SiC	1,1 1,5 2,4 3,18 2,4	35 7 70 140 120	MR-2 Karat 6	1100-1200 1350-1600 1400-1600	1,45 1,27 1,22	13,9 12,0 13,4		
3.	36-13-X-84	(3Th,1U)O <sub>2</sub>	36	9,72	550±100	PyC PyC PyC SiC PyC	1,1 1,5 1,8 3,18 1,8	35 7 60 100 70	RBT-6, №2	1250	1,1	3,0	8,7·10 <sup>-6</sup>	1,42·10 <sup>-5</sup>
4.	* 21-10-X-87	(3Th,1U)O <sub>2</sub>	21	7,55	550±100	PyC PyC PyC+SiC SiC	1,1 1,5 2,4 3,16	42 10 56 70	SM-2 BKS-1	1250	2,1	0-4,0 5,0-8,0 8,0-9,8	1,2·10 <sup>-6</sup> 1,7·10 <sup>-6</sup> 1,0·10 <sup>-4</sup>	2,1·10 <sup>-6</sup> 3,3·10 <sup>-6</sup> 1,0·10 <sup>-4</sup>
5.	* 90T-1-X-88	(9Th,1U)O <sub>2</sub>	90	7,8	610±30	PyC PyC PyC+SiC SiC PyC+SiC	1,1 1,5 2,4 3,16 2,4	58 18 63 67 50	SM-2 №13	1250		>3,0	0,9·10 <sup>-6</sup>	-

\* model spherical fuel elements

## REACTOR TEST RESULTS

The reactor test program for CP based on mixed (Th,U)O<sub>2</sub> fuel was a part of a wide-scale program of experimental development of uranium-graphite pyrocarbon-bound FE for HTGR. On performing reactor tests of CP and spherical FE with (Th,U)O<sub>2</sub> fuel, the primary consideration was given to the following issues:

- effect of the CP design on the GFP yield;
- effect of irradiation temperature on the serviceability of CP;
- behaviour of CP during failure of the outer protective layer under irradiation both in the free-fill state and as constituents of the graphite spherical FE matrix.

To simulate the failure of the outer protective coating, a pilot batch of CP (21-10-X-87) was specially prepared without the mentioned coating. To investigate the radiation resistance of microspherical (Th,U)O<sub>2</sub> fuel, a few batches of CP enriched in uranium-235 to 21, 36 and 90 % were manufactured. The thorium-to-uranium ratios were 3:1 and 9:1. When making CP, various designs were checked: type I (batch 36-13-X-84), type II\* (batch 21-10-X-87), type III (batches 36-12-X-84 and 90T-1-X-88).

As mentioned above, the CP were reactor-tested in the free-fill state and as constituents of model spherical FE. The CP were generally irradiated at a temperature of 1250 °C. However, several radiation-resistance tests of the (Th,U)O<sub>2</sub> fuel under development were also performed at temperatures between 1500 and 1600 °C.

The reactor test values for CP with this fuel are listed in Table II. It is seen from the table that the rate of GFP from CP of the mentioned batches is at the same level as that shown by similar CP designs but with the uranium dioxide fuel [6]. In free-fill tests of CP (batch 21-10-X-87) without an outer pyrocarbon layer, irradiation was performed at an elevated temperature of 1250 °C to 2.1 % fima, and then, with a jump-like rise in temperature up to 1500 °C, - to 3.5 % fima. Immediately after the rise in temperature the rate of GFP release increased by nearly an order of magnitude (from  $8.1 \cdot 10^{-6}$  to  $1.0 \cdot 10^{-4}$  (Kr-88) and remained the same until the experiment completion. This behaviour of type II\* design CP has led us to the conclusion about impossibility of using such CP at elevated irradiation temperatures ( $T \geq 1500$  °C). Yet, these CP can be used under standard irradiation conditions when they enter into the composition of pyrocarbon-bound FE, since the 20-30 mm thick pyrocarbon films deposited as a result of pyrodensification can partly fulfil the functions of the outer protective pyrocarbon layer.

The model spherical FE manufactured on the basis of batch 21-1-X-87 CP were irradiated at 1250 °C to 9.8 % fima (fast neutron fluence of  $2.1 \times 10^{20}$  cm<sup>-2</sup>). The CP retained high service ability up to the design burnup value (for type VGM- and VG-400-type reactors), but after the excess of which some part of CP lost their hermiticity.

Studies were made of free-fill CP for the vitality of uranium-thorium oxide microspherical fuel (CP of batch 36-12-X-84) under conditions of design abnormal rise in temperature (up to 1600 °C) for operating conditions of VGM- and VG-400-type reactors. On achieving the fuel burnups of (12.0-13.4) % fima ( $T_{\text{irr}} = 1350-1600$  °C) and 13.9 % fima ( $T_{\text{irr}} = 1100-1200$  °C), the CP retained their serviceability. The undertaken metallographic examinations have confirmed the integrity of protective coatings.

So, the behaviour under irradiation of the uranium-thorium microspherical fuel considered here is virtually the same as the behaviour of  $UO_2$  fuel under the same reactor test conditions. It has been established by experiments that the  $(Th,U)O_2$  fuel entering into the composition of CP retains its normal operation up to a temperature of 1600 °C, and the pyrocarbon binder of the matrix GSP-graphite further increases the CP hermiticity.

## CONCLUSION

The present results of prereactor and reactor tests give evidence for high radiation resistance and performance characteristics of CP and FE comprising the  $(Th,U)O_2$  fuel and manufactured by the NSC KIPT technology.

At present, the joint LLNL (USA) - NSC KIPT (Ukraine) project is under way to develop the concept of underground nuclear reactor with a nondischarged core throughout the service life (TIW-reactor). The NSC KIPT was commissioned to analyse and select different-purpose materials (fuel and structural) as candidate materials of the TIW reactor core.

On elaborating the underground nuclear reactor concept, the important scientific and technical challenge is to develop thorium-base fuel materials serviceable as a nuclear fuel. In this connection, when making choice for the TIW reactor we shall take into account the successful experience of experimental development of microspherical  $(Th,U)O_2$  fuel for HTGR.

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