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## **FABRICATION, PROPERTIES AND IRRADIATION PERFORMANCE OF MOLDED BLOCK FUEL ELEMENTS FOR HTGRs**

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### **ABSTRACT**

The molded block fuel element (FE) also called monolith is a molded body, consisting of a substantially isotropic highly crystalline graphite matrix, fuel regions within the same matrix and cooling channels. The fuel regions contain the fuel in the form of coated particles which are well bonded to the remaining graphite matrix, so that both parts of the block form a monolithic structure. The monolith meets the requirements for the very high temperature reactors attaining helium outlet temperatures above 1000°C. To fabricate the molded block FEs demonstration plant was erected and put into operation. The equipment worked without malfunction. The produced block FEs meet the specifications of GA machined block FEs. All specimens and block segments irradiated at temperature up to 1600°C and max. fast fluence  $E > 0,1 \text{ MeV of } 11 \times 10^{21} \text{ n/cm}^2$  show perfect behaviour without any damage.

**Keywords:** HTR Molded Block Fuel Element, Monolith, A3 Graphite Matrix, Iso Granulate, Portable Steel Die, BISO or TRISO Coated Particles, Access Uranium or Thorium, Ring Gap Homogenizer

### **DESCRIPTION**

Following the fabrication technique originally developed for molded fuel spheres the molded block fuel element (FE) was introduced and brought to technical maturity. The development was brought to a state characterized by reproducible fabrication of block FEs with required properties providing the base for

economical fabrication, including the irradiation testing and Post Irradiation Examinations (PIE).

The molded block FE also called monolith is a molded body, consisting of a substantially isotropic highly crystalline graphite matrix, fuel regions within the same matrix and cooling channels. The fuel regions contain the fuel in the form of coated fuel particles which are embedded in the graphite matrix. It is essential that the fuel regions are well bonded to the remaining graphite matrix so that both parts of the block, that is the fuel containing graphite matrix and the fuel free regions form a monolithic structure.

The monolith has a great potential:

- Since no machining of the graphite is required the fabrication is facilitated and the usual graphite losses are avoided.
- The thermal performance is considerably improved. Under the present operating condition the fuel temperature is 200 – 250°C lower due to elimination of the gap between the regions containing fuel and free of fuel and due to increase of the thermal conductivity of the fuel regions.<sup>1</sup>
- In addition the coated fuel particles should stand even higher ratings since they make excellent thermal contact with the surrounding fine grained and highly dense graphite matrix material and are better spread within the FE. In consequence thereof the core power level can be considerably increased.

The monolith is fabricated in two steps using a combined cold-hot molding process:

In the first step graphite powder and resin are wet mixed and then dried. The resinated graphite matrix powder thus obtained is isostatically consolidated into spheres in a rubber die at room temperature under a high forming pressure of about  $100 \text{ MN/m}^2$

The graphite spheres are crushed to granules from which the 0.3-3mm fraction is obtained by screening. The average granule consists of about a million isotropically arranged particles of the original graphite powder.

In the second step the granulated graphite material and coated fuel particles are molded to a block in a steel die at  $180^\circ\text{C}$  at low forming pressure of about  $12 \text{ MN/m}^2$ . Subsequently the block is heat treated, in two stages, first up to  $800^\circ\text{C}$  to carbonise the resin with an argon purge and then to  $1900^\circ\text{C}$  in vacuum.<sup>2,3,4</sup>

The graphite matrix powder of A3 composition has been used and this is a mixture of 64 weight per cent natural graphite powder FP, Company Kropfmühl, 16% graphitised petroleum coke powder KRB, Company Ringsdorf and 20 % phenol formaldehyde resin 4911, Company Bakelite. The A3 graphite has been irradiated in several test reactors up to the very high fast fluence of  $2.2 \times 10^{22} \text{ n/cm}^2$ ,  $E > 0.1 \text{ MeV}$ . The irradiation tests were very successful and revealed a relatively low and nearly isotropic shrinkage behaviour.<sup>5,6</sup>

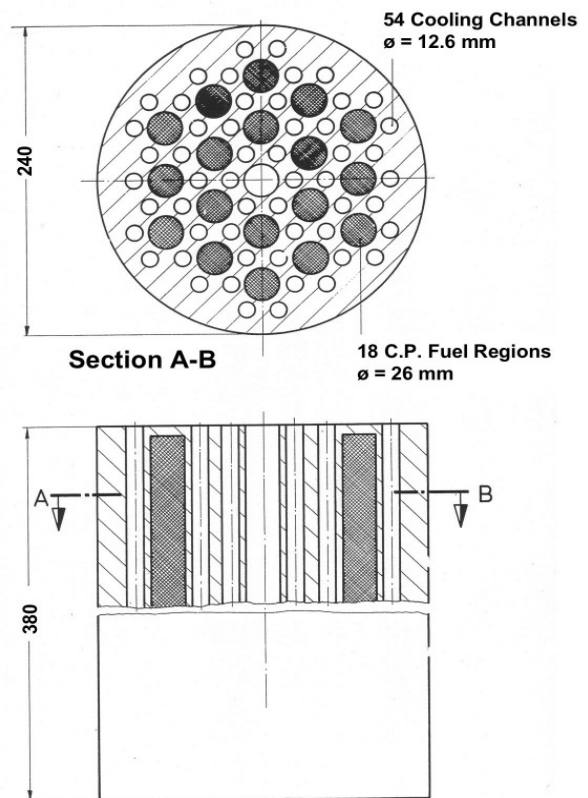
The monolith was first fabricated based on the Dragon FE development (hollow rod and 18 pin tubular block design) with reduced size and of inverse design. The reduced block size was chosen for following reasons:<sup>7,8</sup>

- to keep the development effort at a reasonable level in this early stage
- to produce a block size with dimensions suitable for irradiation testing in the Peach Bottom and Dragon Reactors and finally
- to obtain the required design criteria to set up the demonstration plant suitable for economical fabrication of fuel size block FE, meeting GA requirements.

Figure 1 shows the molded block FE of inverse design. In table 1 design data of different HTR-FE are compared with molded block FE of inverse design and reduced size 1:2.

After a relatively short development period the demonstration plant was erected and put into operation. The main feature of the new plant is a portable steel die, which is moving from one to other location where the following successive steps are carried out:

- filling of the die with granulated graphite material and premolding of the block with a 70 Mpa triple action hydraulic press



**Fig. 1 Molded Block FE of Inverse Design and Reduced Size 1:2**

- loading of the pre-molded block with fuel
- heating up to a temperature of  $180^\circ\text{C}$
- final molding using a 250 Mpa double action hydraulic press
- ejection of the block, using a 70 Mpa single action hydraulic press and
- heat treatment.<sup>9</sup>

The fabrication line has been successfully proved as presented in Figure 2. No difficulties occurred. The equipment worked without malfunctions. In conclusion to the termination of the monolith development a series of 7 block FE of original size and 8-rows design was fabricated and the blocks were characterized.<sup>10</sup> The fuel block is a hexagonal prism with 360 mm width across flats, 793 mm height and a total volume of 86 l, containing 13,3 kg thorium and 0,77 kg uranium in form of BISO and TRISO coated fuel particles.

The 8-row design was selected as reference block FE for the planned 1160 MWe power reactor. Figure 3 shows the flow diagram for fabrication of molded block FEs.

After the final annealing at  $1950^\circ\text{C}$  all 7 block FE were free of cracks, no defects were detected visually. Figure 4 shows 8-rows block FE.

A test plan for characterization of the 6 block FE produced was set up with accordance to GA specifications for machined block FEs. The blocks were first measured and than dismantled in order to

Design Features Fuel Element Type	Volume of c.p. containing regions (%) $V_r/V_e \times 100$	Volume of cooling (%) "Voidige" $V_c/V_e \times 100$	Volume of structural graphite $V_g/V_e \times 100$	Specific cooling surface cm <sup>2</sup> /liter
DRAGON				
- Hollow Rod	11	18	-	221
- Block - FE				
- Tubular Block - FE	13	17,5	-	348
GA Font St. Vrain 10 Row Block FE	23	18	59*	460
HKG THTR Molded Fuel Sphere	34	40**	60 (34 + 26)	600
Molded Block FE with Inverse Design and Reduced Size	26	18	82	560

$V_e$  = Volume of FE

$V_r$  = Volume of c. p. regions

^ pick up hole unconsidered

$V_c$  = Volume of cooling channels

$V_g$  = Volume of structural graphite

\*\* sphere packing density

Table 1: Design Data comparison of different HTR FEs with Molded Block FE of inverse Design and reduced size 1:2

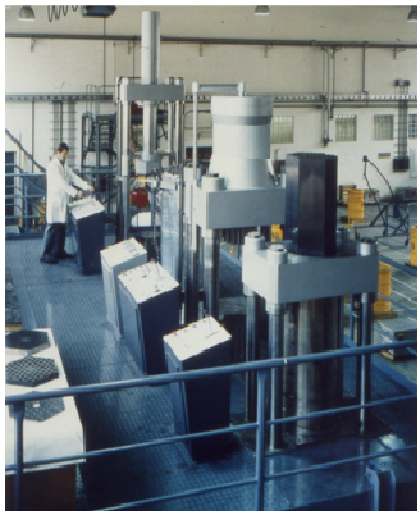


Fig. 2 Demonstration plant for Fabrication of HTR Molded FEs

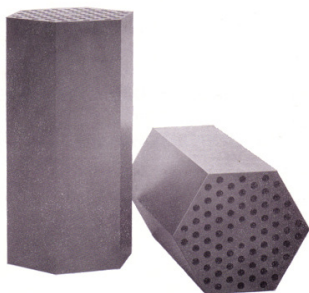


Fig. 4 HTR Molded Block Fuel Element after Heat Treatment

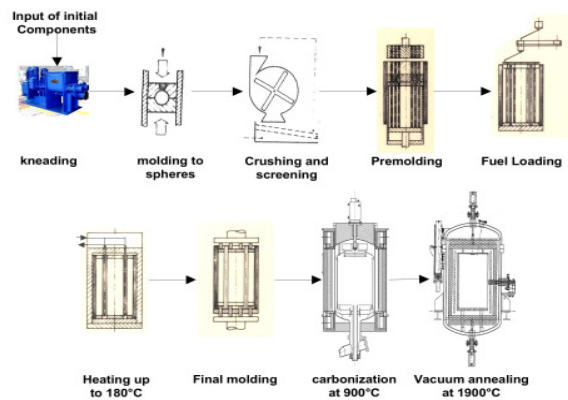


Fig. 3 Flow Diagram for Fabrication of Molded Block FE

determine the physical and chemical properties. To examine the dimensional stability of the blocks following properties were measured: Width over flats and diameter of coolant holes. The results are represented in table 2.

In order to determine physical properties test samples were taken as well from the top, middle and bottom of the blocks as from inner and outer blocks area.

The survey of physical properties for all six molded block FEs is compiled in table 3.

The results revealed the monolith is almost without any properties gradients.

Properties Measurement	Width over Flats (mm)	Diameter of Coolant Holes (mm)
Target Value	359,9 ± 0,4	21,03 <sup>+0,08</sup> <sub>-0,18</sub>
$\bar{x}$	360,03	21,03
X max	359,6	21,07
X min	359,6	20,88
S	0,19	0,03
n	30	198

**Table 2 Dimension Stability of 8 Row Molded Block FE after Heat Treatment**

Properties Measurements	$\bar{x}$	s	n
Density (g/cm <sup>3</sup> )	1,73	0,01	718
3 Point Bending Strength (MN/m <sup>2</sup> )			
axial	14,6	1,59	356
radial	16,8	1,78	354
Tensile Strength (MN/m <sup>2</sup> )			
axial	6,9	0,79	142
radial	7,31	0,86	57
Young Modulus (MN/m <sup>2</sup> ) x 10 <sup>2</sup>			
axial	6,40	0,5	358
radial	11,0	0,3	357
CTE 20 - 500° C (° K) x 10 <sup>-6</sup>			
axial	3,66	0,35	120
radial	2,55	0,23	120
Heat Conduct (W/cm <sup>2</sup> K)			
axial	0,55	0,03	325
radial	0,80	0,03	329
Special Electric Resist. (m.Ohm.cm)			
axial	2,20	0,05	356
radial	1,48	0,04	359
Corrosion at 1000°C (mg/cm <sup>2</sup> h)	1,31	0,19	30
Merit Factor* (kW/m)			
axial	10,2	-	-
radial	10,8	-	-

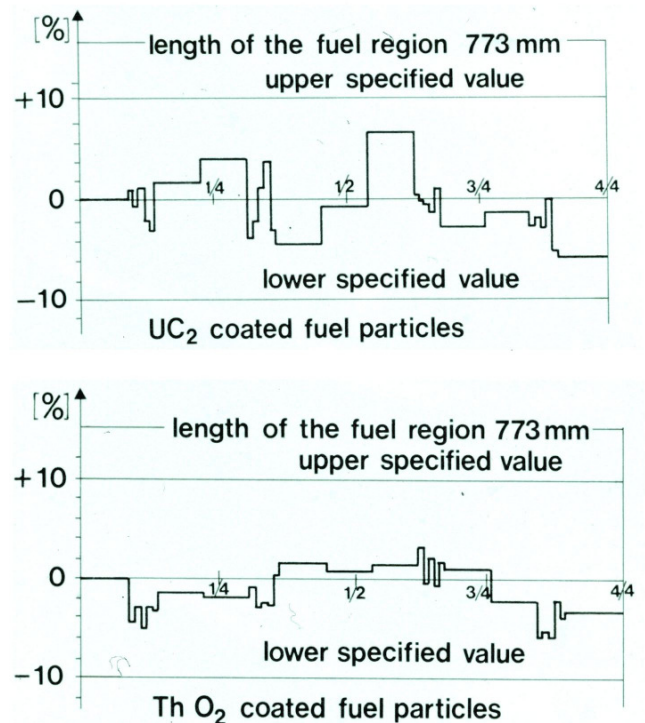
\*Merit Factor =  $\frac{\text{Tensile Strength} \times \text{Heat Conduct at } 1000^{\circ}\text{C}}{\text{CTE} \times \text{Young Modulus}}$

**Table 3 Physical Properties Survey of Six 8-Row Molded Block FEs**

For determining the free (access) uranium and thorium 5 samples with 20 mm diameter and 60 mm height were taken from each block FE and electrolytically disintegrated. After disintegration the coated particles and graphite were leached with nitric acid and analyzed for uranium and thorium. Each sample contained 0,25 g uranium and 4,16 g thorium. The mean values measured on 30 samples are  $50 \times 10^{-6}$  for uranium and  $43 \times 10^{-6}$  for thorium (specified value  $< 200 \times 10^{-6}$ )

For fuel loading of the premolded block a homogeneous mixture of overcoated coated fertile particles, overcoated coated fissile particles and isogranulate was loosely filled into 22 premoulded channels for fuel reception. In order to obtain the required homogeneous mixture a ring gap

homogenizer followed by portion splitter was introduced. The distribution of fissile and fertile coated particles over the total fuel channel length are represented in figure 5.



**Fig. 5 Distribution of Fissile and Fertile Coated Particles**

The Post Irradiation Examination (PIE) tests are divided into 3 groups:

- graphite matrix research tests
- model sample tests to investigate interaction behaviour of graphite matrix and fuel containing regions and
- tests of representative segments machined out of the molded blocks with the required layout.

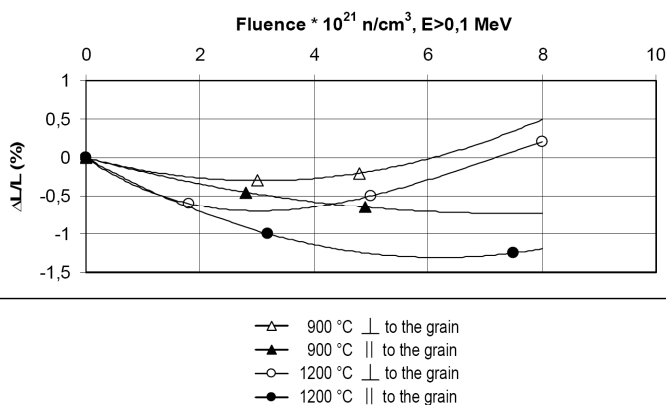
The irradiation tests were performed in HFR Petten, FRJ Jülich, DR Dragon, BR2 Moll, R 2 Studsvik and PBR Peach Bottom Reactors. The survey of the tests is compiled in table 4

Properties	Temperature (°C)	Fluence ( $\times 10^{21}/\text{cm}^2$ E > 0,1 MeV)
<b>Measurements</b>		
<b>Graphite Matrix Samples</b>		
HFR – M 17 – 23	400 - 1400	8,0
FRJ1 – MF 1	500 - 1000	1,0
DR – C1	950 - 1400	0,4
<b>Model Samples</b>		
DR – S 6/7	1200	2,4
BR2 – P 15	1300	11,0
R2 – M 5	1200	5,9
<b>Block Segments</b>		
DR – B1, 2, 3	1400	3,4
DR – Met VI 1, 2	1250	2,1
PBR – B <sup>1</sup>	1600	2,5

**Table 4 Compilation of Irradiation Tests**

The dimensional changes of A3 graphite matrix samples irradiated in the HFR Petten are described in figure 6

In particular it has to be emphasised the results of 16 model BR2 – P15 samples irradiated in the BR2 reactor Moll (diameter 20 mm, length 30 mm, coated particles volume fraction in the fuel region 20 %). The temperature ranged between 800 and 1300°C and the fast fluence up to  $1,1 \times 10^{22}$  n/cm<sup>2</sup>, E > 0,1 MeV. The samples shrunk radially max. 1,8 % and swelled axially by a maximum of about 1,2 %. Although the axial turn-around point of the dimensional changes was exceeded the breaking load increases slightly from 2560 N before to 2855 N after irradiation (mean value).



**Fig. 6 Dimensional Changes of A3 Graphite vs. Fast Fluence**

The Post Irradiation Examination (PIE) results of the block segments irradiated in the Peach Bottom reactor are described in GA-report No. 13699. The stress analysis of the segments yielded tensile stresses up to 226 % of the ultimate tensile strength during irradiation and up to 428 % at shut down. Nevertheless the segments exhibit no crack formation.

Figure 7 represents a compilation of successfully irradiated molded block FE samples and segments. Figure 8 represents irradiation column of Dragon segments and fig. 9 Dragon reactor test FE



**Fig. 7 Compilation of Successfully Irradiated Block FE Samples and Segments**



Fig. 8 Irradiation Column of Dragon Segments



Fig. 9 Dragon Reactor Test Element

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