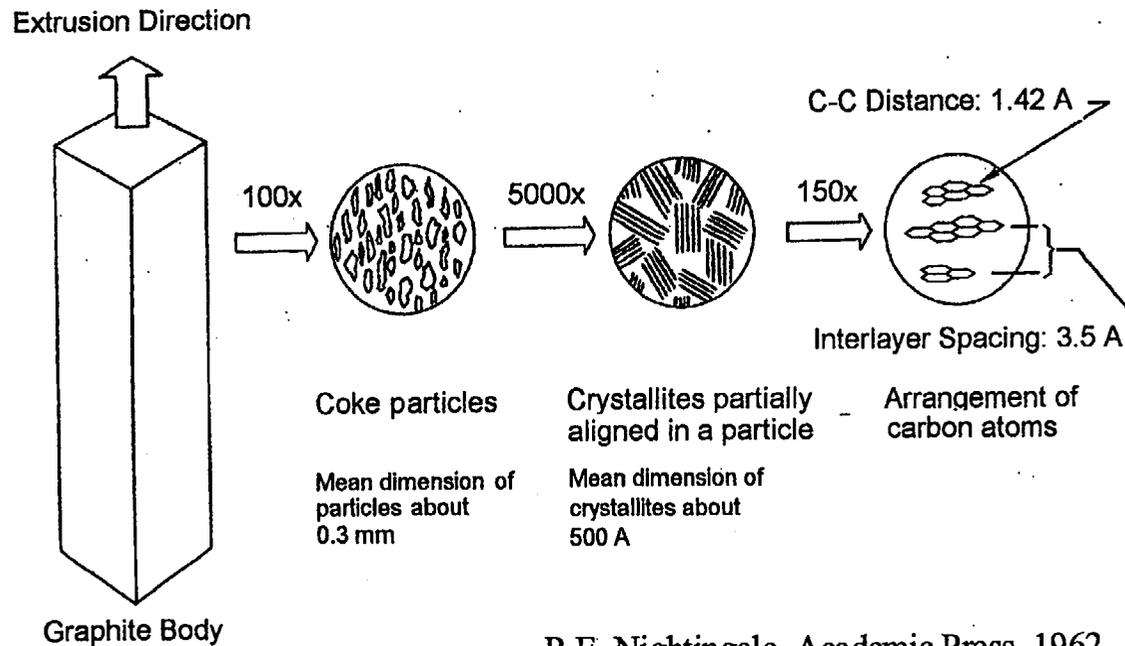


D.E. Carlson  
25.07.01 #19  
Jülich

# Safety Aspects of HTR Technology

G. Haag: *Nuclear Graphite for the HTR - Research, Development, and Industrial Production*

## The substructures of polycrystalline graphite



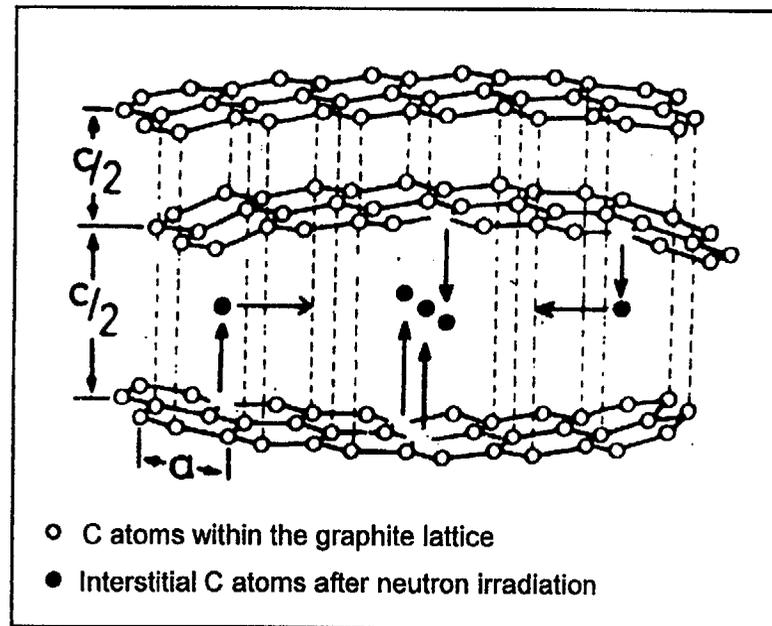
R.E. Nightingale, Academic Press, 1962

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Research, Development, and Industrial Production*

## Radiation Damage in Graphite



Aus E. Fitzer: Graphit als Reaktorwerkstoff, Haus der Technik, 1967 (?)

# **Safety Aspects of HTR Technology**

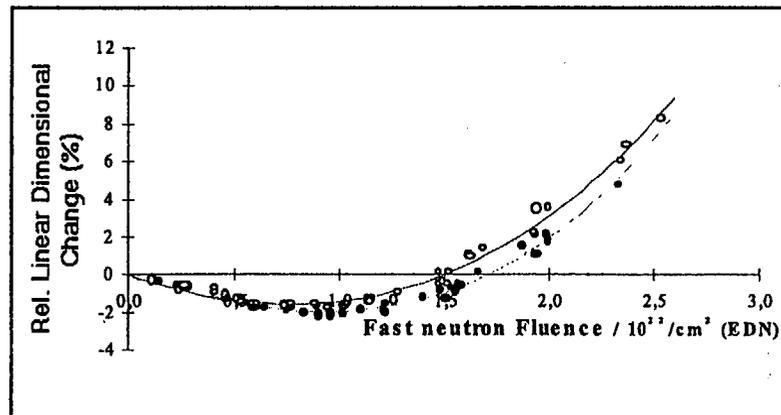
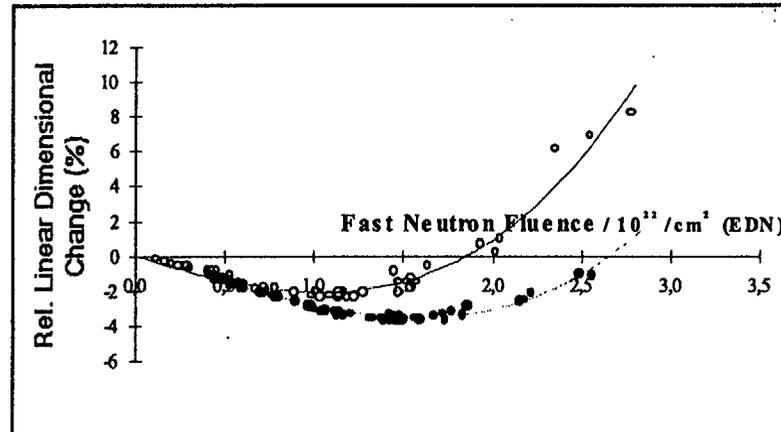
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The number of C-atoms in graphite crystallites displaced by a single 1 to 2 MeV neutron is of the order of 20 000.

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## Irradiation Induced Dimensional Changes



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## Influence of Coke and Forming Technique on Important Properties of Reactor Graphites

Grade	ATR-2E	ASR-1RS	ASR-2RS	ASR-1RG
Coke	Special Pitch Coke	Ordinary Pitch Coke Sec. Coke T.	Ordinary Pitch Coke Sec. Coke T.	Ordinary Pitch Coke
Forming	Extrusion	Vibration	Vibration	Vibration
App. Density (g/cm <sup>3</sup> )	1.80	1.82	1.87	1.78
Tens. Strength <i>par.</i> (N/mm <sup>2</sup> )	12.6	18.3	19.5	13.0
<i>perp.</i>	12.4	18.3	18.5	11.6
Anisotropy	1.12	1.05	1.02	1.15

# Safety Aspects of HTR Technology

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## *Statements I*

- Nuclear graphite for non exchangeable core components must be nearly isotropic - but not isostatically moulded.
- Special coke processing and careful vibrational moulding yield the best graphite grades with respect to isotropy, strength, and homogeneity.
- The expected lifetime of graphitic core components has to be verified by stress analysis using reliable irradiation data.
- Today, none of the former widely tested graphites is still available.

# **Safety Aspects of HTR Technology**

G. Haag: *Nuclear Graphite for the HTR -  
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## ***Statements II***

- Graphite for the PBMR reflector should be produced on a best guess basis using still existing procedures and experience.
- Data for stress analysis calculations should be deduced from similar materials tested in former irradiation programmes.
- Therefore, an international database with data from former nuclear graphite test programmes (US, UK, Japan, Germany, France) should be supported by possible users.
- For future HTR projects, development and irradiation testing of new graphites should be resumed as soon as possible.

## DEVELOPMENT OF REACTOR GRAPHITE \*

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The German graphite development programme for High Temperature Reactors has been based on the assumption that reactor graphite for core components with lifetime fluences of up to  $4 \times 10^{22}$  neutrons per  $\text{cm}^2$  (EDN) at  $400^\circ\text{C}$  can be manufactured from regular pitch coke. The use of secondary coke and vibrational moulding techniques have allowed production of materials with very small anisotropy, high strength, and high purity which are the most important properties of reactor graphite. A variety of graphite grades has been tested in fast neutron irradiation experiments. The results show that suitable graphites for modern High Temperature Reactors with spherical fuel elements are available.

### 1. Introduction

Graphite is an almost perfect reactor material. In 1942, Enrico Fermi already used graphite as moderator in the first nuclear reactor fashioned by the hand of man. In the course of nuclear reactor development, the physical and chemical properties of graphite have been responsible for many important advances in nuclear reactor technology.

It is obvious that on the way to modern High Temperature Reactors (HTR) providing temperatures of about  $1000^\circ\text{C}$  severe technical problems had to be solved, but in no case was graphite with its good mechanical and excellent thermal properties the reason for these problems. Only the future reactor concepts HTR-500, HTR-100 and to some extent the Modular HTR make use of graphite in a way that limiting factors have to be considered. This was the reason for the development of new isotropic graphites and for irradiation programmes to investigate the influence of the Wigner effect (i.e. fast neutron radiation damage in the graphite

crystal structure) on the physical properties of graphite at very high neutron fluences.

Most of the nuclear graphites have been produced by the Acheson process which for several decades had been used for the production of furnace electrodes [1]: A coke filler is mixed with thermoplastic hydrocarbon binder, pressed to form a "green" artifact and then in two steps is subjected to a more or less complex heat treatment. The first step consists of the baking process at temperatures of about  $1000^\circ\text{C}$ , converting the binder into almost pure carbon, while in the second step the baked material is graphitized by electric resistance-heating to a final temperature of  $2600^\circ\text{C}$  or higher. In some cases where higher density is required, the porosity resulting from the raw materials and the baking process is decreased by impregnation with pitch or tar prior to graphitization.

The fast neutron irradiation behaviour of polycrystalline graphite had been studied for many years in the frame of the Advanced Gas-cooled Reactor programme and the OECD Dragon Reactor Project in Britain, the Experimental Gas-Cooled Reactor (EGCR) program and the High-Temperature Gas-cooled Reactor (HTGR) program in the USA and the AVR and THTR Pebble Bed Reactor programmes in the Federal Republic of

\* Dedicated to Prof. H. Nickel on the occasion of his 60th birthday.

Germany. During the early 1970s it became clear that only isotropic graphites might be able to meet the needs of future pebble bed High Temperature Reactors.

The most promising way of manufacturing isotropic graphites had been based on Gilsonite coke as filler, a naturally occurring bitumen mined in Utah, USA, which consists of almost spherical grains. However, for some reasons the longterm availability of Gilsonite coke had to be called into question and the first oil crisis in 1973 led to the decision to use only domestic raw materials for German reactor graphites.

## 2. HTR requirements

The German development programme for reactor graphite is based on the cooperation of the companies Hochttemperatur-Reaktorbau GmbH (HRB) and Gesellschaft für Hochttemperaturreaktor-Technik GmbH (GHT)/Interatom GmbH as nuclear reactor constructing industry, Sigri GmbH and Ringsdorff-Werke GmbH manufacturing graphite and the Kernforschungsanlage Jülich GmbH (KFA).

As an example for future High Temperature Reactors, fig. 1 shows schematically the core construction of a HTR-500 pebble-bed reactor. The different requirements with respect to neutron fluence, mechanical load, chemical impact or block size led to different goals of the graphite development programme.

### 2.1. Reflector components

Unexchangeable components for the high- and low-fluence zones are called reflector components. Their main purpose is to reflect and moderate the neutrons escaping from the reactor core.

Because the reflecting power depends on the number of carbon atoms per  $\text{cm}^3$ , the density of reflector graphite should not be less than  $1.70 \text{ g/cm}^3$ .

With respect to neutron physics, the overall neutron-capture cross section is required to be smaller than 5 mbarn. Consequently, the content of neutron-absorbing chemical elements such as Gd, B, Sm, and Eu, has to be kept small. With respect to corrosion by impurities from the cooling gas such as  $\text{O}_2$ ,  $\text{H}_2\text{O}$  and  $\text{CO}_2$ , catalytically acting chemical elements such as Fe, Ca, Sr and Ba are important. Experience shows that all this can be summarized by an upper limit for the ash content of nuclear graphite of about 600 ppm.

For the use of graphite as structural reactor material, the mechanical stability – primarily the tensile strength – has turned out to be most important. As a result of the Wigner effect and of temperature gradients, internal

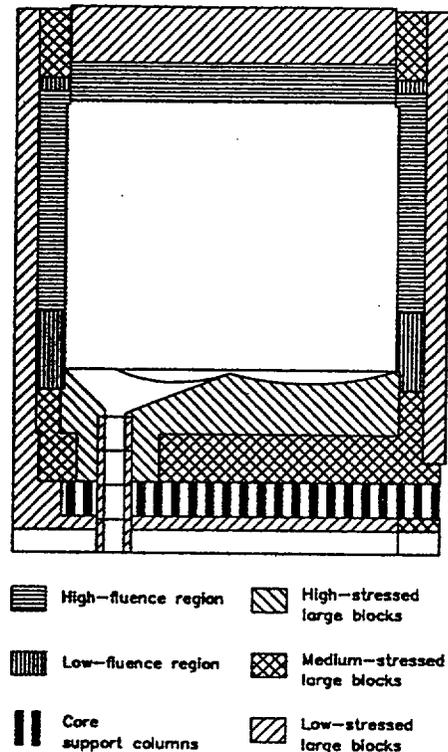


Fig. 1. Vertical section of a pebble-bed HTR (schematic diagram).

stresses are created in the components which might be damaged severely if the stresses exceed the strength. Therefore, some other physical properties are supposed to satisfy limiting conditions such as dynamic Young's modulus not to exceed  $12 \text{ kN/mm}^2$ , thermal conductivity higher than  $90 \text{ W/mK}$  at room temperature, and coefficient of linear thermal expansion (CTE) smaller than  $6 \times 10^{-6}/\text{K}$  from 20 to  $500^\circ\text{C}$ .

The anisotropy factor of graphite is defined as the ratio of the linear thermal expansion coefficients in the two main crystallographic directions. For the construction of reactor components only isotropic graphites with anisotropy factors from 1 to 1.05 are stable enough against fast neutron irradiation damage. A compromise had to be found between small CTE-values to be achieved using anisotropic coke and isotropic products based on coke with high CTE.

### 2.2. Core support construction and outer parts of the reflector

Graphite components belonging to the core support construction or to the outer parts of the reflector (see

large block size areas in fig. 1) are almost entirely protected against fast neutrons by the bottom reflector, the top reflector and the inner side reflector. Therefore, the design of these components does not depend on any irradiation behaviour. The strength requirements depend on the special function of the components but normally are of minor importance compared to the inner reflector components. However, development and production of core support blocks with dimensions of  $500 \times 500 \times 2000 \text{ mm}^3$  was a major problem. Because the temperatures of these components under operation are particularly high, good chemical purity was necessary to achieve low corrosion rates.

The core support columns are also part of the support construction (see fig. 1). They must have very high strength, in particular as they are exposed to cooling gas streams of different temperatures leading to thermal stresses. Core support columns are also in danger of corrosion and therefore are made from graphite with small ash content.

### 3. Developing reflector graphite for High Temperature Reactors

The HTR graphite development programme was originally based on two main aspects:

*Development of new cokes* with isotropic structure and isometric grain made from coal tar pitch to produce reactor graphite using conventional methods (e.g. forming by extrusion).

*Development of new methods* for the production of isotropic graphites from commercially available anisometric pitch coke.

However, the demand for raw materials for nuclear purposes has been estimated to be too small to make every effort to develop new cokes. Actually, there was only one special pitch coke available to produce the ATR-2E graphite which exhibited good physical properties and excellent irradiation behaviour without modifying the conventional production process. This experience led to the conclusion that the development of new production techniques using ordinary pitch coke might be successful.

#### 3.1. Secondary coke technique

Normally, the industry produces graphite from well graphitizing cokes with a more or less pronounced layer structure. These cokes fracture parallel to the layer planes when they are ground, leaving anisometric grains which may be needle-like in the extreme. Depending on

the pressing technique, anisometric grains can show preferred orientation, which makes the final product anisotropic.

The preferred orientation of the grains can be avoided using a particular preproduction technique [2]: The precursor coke with 0.12 mm maximum grain size is mixed with standard coal tar pitch binder using a fast mixer. Then, the hot mixture is compressed by vibration moulding (see section 3.2.) yielding large blocks to be baked at temperatures above  $1100^\circ\text{C}$  and then ground to a maximum grain size of 1 mm. Thus, an isotropic coke aggregate is obtained with high bulk density, excellent mechanical properties, low contents of ash and volatiles, and nearly spherical grains.

This secondary coke is used exactly like conventional filler; it is mixed with coal tar pitch binder in a fast mixer, with 20% ground isotropic nuclear graphite added (see section 3.3.) to improve the baking and graphitization behaviour.

#### 3.2. Vibrational moulding

In principle, the preproduct technique can be used in combination with all forming methods. Nevertheless, reflector blocks for future High Temperature Reactors are so large, that forming by moulding or by extrusion had to be given up for cost reasons and the vibrational moulding technique [3] has been developed. The equipment and the procedure are shown in fig. 2. The vibration moulding machine consists of a vibration table, a mould, a covering weight with a guide rod and a vacuum device.

The hot green mixture is poured into the mould directly from the mixer. Compression of the mixture results from vertical vibrations of the mould against the

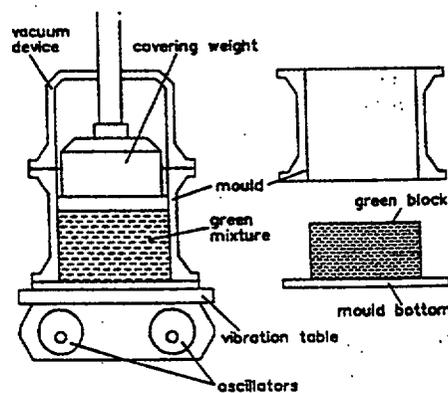


Fig. 2. Schematic diagram of the vibrational moulding process.

inert mass of the covering weight. Evacuating the mould before and during densification improves the final density and the homogeneity of the product.

The optimization of the vibrational moulding process had to take several parameters into account: The best *vibration frequency* is dependent on the composition of the green mixture and of the height of the block; the optimum frequency ranges from 20 to 35 cps. The optimum *amplitude* increases from 1.5 to 4 mm with increasing height of the block. Amplitudes below the optimum lead to smaller densities, higher amplitudes favour crack formation. The *covering weight* depends on the composition of the green mixture, the height and the height to cross section ratio of the block. The optimum pressure ranges from 150 to 500 g/cm<sup>2</sup>. The *vibrating time* is less than 1 min.

By carefully tuning the frequency and amplitude, a stable counterphase oscillation of the vibrating table and the covering weight is adjusted, which provides steady progress of compression and good homogeneity of the blocks.

### 3.3. Improving the graphitization behaviour

In the initial stage of the development of isotropic reactor graphites based on coal tar pitch coke, many graphite blocks cracked during graphitization. At that time, experiences concerning the expansion/shrinkage behaviour of carbon materials were limited to anisotropic grades (e.g. from the examination of puffing effect), whereas the behaviour during graphitization of isotropic carbons based on pitch cokes had not yet been investigated in detail.

After observing the dimensional behaviour of different baked carbon grades in a high temperature dilatometer from 100 to 2400 °C, it became clear that carbons based on petroleum-needle coke behave different from those based on secondary coke. Fig. 3 shows that these materials behave similarly at temperatures below the baking temperature of 900 °C where they both expand linearly, but at higher temperatures the secondary coke carbon reverses its dimensional behaviour, whereas the needle-coke material continues expanding – slowly from 900 to 1700 °C and again faster above 1700 °C. The pronounced shrinking at temperatures above the baking temperature was found to be characteristic for carbon materials based on isometric coke and, when combined with the higher Young's modulus of isotropic graphite, is believed to cause the poor graphitization behaviour.

Numerous test series have been performed to investigate the influence of various production parameters. As can be seen from fig. 3, adding graphite to the

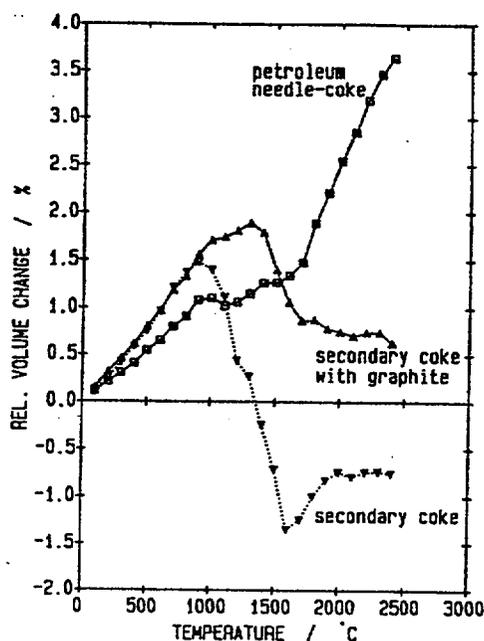


Fig. 3. Dimensional behaviour of baked carbon based on different fillers in the temperature range from 100 to 2400 °C.

secondary coke filler markedly reduced the shrinkage during the graphitization process. Another positive influence on the dimensional behaviour has been found from impregnating and rebaking at higher temperature [4]. Therefore, to prevent isotropic carbon artifacts from cracking the following steps have been established:

- adding about 15 to 20% ground graphite of the same grade to the final mixture (which does not change the properties of the product),
- increasing the maximum baking temperature from 900 to 1100 °C (which improves the properties of the product).

### 3.4. Reference graphites

A wide range of different graphite grades have been irradiation tested (see section 4). Finally, the decision was made in favour of the graphite grade ASR-1RS. Its smaller anisotropy, larger block sizes and appreciably higher strength had been decisive.

As there is only one impregnation necessary to achieve such good properties, it is possible to improve this graphite again by a second impregnation. This leads to the grade ASR-2RS to be used in all cases where the

Table 1  
Properties of German graphites for the reflector of High Temperature Reactors

Property <sup>a)</sup>	Graphite grade and year of production			Improved ASR-1RS 1979	Demonstration grades		
	Reference grades				ASR-1RS 1986	ASR-2RS 1986	ASR-1RG 1986
	ASR-1RS 1975	ASR-2RS 1981	ASR-1RG 1981				
Ash content (ppm)	390	40	130	490	640	620	570
Density (g/cm <sup>3</sup> )	1.78	1.87	1.79	1.81	1.84	1.88	1.79
Lin. therm. expansion (  )	4.70	3.92	3.50	4.22	3.62	3.62	3.15
coefficient (10 <sup>-6</sup> /K) (⊥)	4.87	4.12	3.95	4.42	3.85	3.84	3.72
Anisotropy factor	1.04	1.05	1.13	1.05	1.06	1.06	1.18
Young's modulus (  )	9.9	10.5	8.7	10.2	10.1	11.1	8.7
(kN/mm <sup>2</sup> ) (⊥)	9.2	10.1	7.7	9.8	9.9	10.7	7.6
Bending strength (  )	26.0	28.5	19.4	26.9	24.7	27.8	16.1
(N/mm <sup>2</sup> ) (⊥)	23.0	28.2	17.1	26.4	25.8	28.6	15.5
Compress. strength (  )	67.1	79.8	47.0	66.5	62.5	73.4	39.8
(N/mm <sup>2</sup> ) (⊥)	63.1	80.0	47.8	66.5	64.8	75.7	41.5
Tensile strength (  )	14.9	19.1	12.0	17.9	17.2	18.8	10.6
(N/mm <sup>2</sup> ) (⊥)	13.5	18.1	10.9	18.1	17.5	19.6	10.1
Thermal conductivity (  )	125	146	154	134	155	162	162
(W/mK) (⊥)	125	142	136	130	149	157	141

<sup>a)</sup> (||) = parallel, (⊥) = perpendicular to grain orientation.

highest possible strength is required.

Some parts of the core structure of a pebble-bed HTR (especially in the so-called HTR-Modul reactor) are exposed to only small or medium neutron fluences. For this purpose it has been suggested to use a cheaper graphite made directly from ordinary pitch coke by vibrational moulding with only one impregnation. This material exhibiting higher anisotropy and smaller strength is called ASR-1RG. Some physical properties of all reference grades are listed in table 1.

The development of isotropic reactor graphite with optimized properties raises the problem of how the properties change from batch to batch or when the production technique is scaled up. As an example, table 1 shows that the transition from ASR-1RS(1975) produced in laboratory scale, to ASR-1RS(1979) produced in preproduction scale, resulted in remarkable property changes. This led to the decision that, at the end of the development programme, so-called demonstration batches of all reference grades had to be produced in production scale, i.e. in the order of 30 to 40 full size blocks. The question, if the observed variations in some of the physical properties are significant to the design of graphitic reactor components, remains upon until the irradiation testing is done.

### 3.5. Developing isostatically moulded graphites

Based on the experience that reactor graphites for core components must be isotropic, the isostatic moulding technique has to be taken into account when, in the early 1970s, graphites made from Gilsonite coke had to be replaced. At that time, the Ringsdorf-Werke company had a lot of experience with that moulding process. However, the block dimensions had to be increased by a factor of 2 to 3 for reactor purposes and at that time there was only slight experience of the neutron irradiation behaviour of isostatically moulded graphite.

These conditions seemed to be somehow contradictory since isostatic moulding requires small grain size of the filler but high binder content. Consequently, the risk of graphite blocks cracking during the baking process increases with increasing block size and also a higher binder content, in general, leads to a decrease of dimensional stability under irradiation.

In the first stage, different cokes, for example regular petroleum coke and pitch coke, had been taken into consideration which showed that there might be some preference for petroleum coke. However, this development has also been influenced by the decision to use domestic raw materials exclusively.

Thus, effort has been concentrated on graphite grade V483 and several changes of the production technique have been investigated to improve the baking behaviour of blocks as large as  $\varnothing 370 \times 900$  and  $370 \times 300 \times 900$  mm<sup>3</sup>.

The irradiation testing has been done primarily in the HFIR reactor at Oak Ridge, directly comparing all the concurrent graphites. Therefore, neither temperature regulation was necessary nor flux effects (if existing) had to be considered. It turned out that V483 graphite exhibited faster and higher initial shrinkage due to its relatively high binder content. After the dimensional changes reversed, rapid expansion occurred, which obviously would lead to higher stresses in graphitic reactor components. This was the reason for giving up the V483 material. Being a fine grain graphite its fracture behaviour is expected to be worse than that of coarse grain graphites and the relatively high strength was not high enough to compensate this disadvantage.

### 3.6. Developing graphite for core support columns

Originally, graphite for core support columns had been produced with the same mixture V483 as was used for developing graphite for reflector components. However, the required block dimensions and physical properties were different, and other steps had to be taken to realize the desired product. With the available press, it was not easy to achieve the block dimensions of  $\varnothing 155$

$\times 1980$  mm<sup>3</sup> after machining and, at the same time, to get high strength and high corrosion resistance.

The first reasonable result was the graphite grade V483T; its properties are listed in table 2. However, it turned out that using still smaller filler grain significantly improved values for strength could be obtained and simultaneously, an important reduction in ash content was achieved. This improved grade is referred to as V483T2 in table 2.

This stage of development was reached in 1982. In the meantime, no further pebble-bed reactor has been built. However, the best way to conserve this knowledge is to continue developing graphites for future applications and to guarantee long-term availability. This led the Ringsdorf-Werke to the decision to use other raw materials, and to find out how to reproduce or even to improve the properties of graphite for core support columns.

The data of grades V483T5 and V483T6 in table 2 show that these efforts have been very successful. The good bending and compressive strength values of V483T2 have again been increased by about 30% (T5) and 40% (T6) and the ash content has been decreased by more than a factor of 2 in both grades.

## 4. Irradiation testing

Irradiation induced stresses resulting from the Wigner effect are most important for the selection of graphite for application at high fast neutron fluences. The graphite lattice is fundamentally damaged in a reactor environment by collision of high-energy neutrons with carbon atoms in the lattice. The carbon atoms are displaced to interstitial positions, leaving behind vacant sites in the layer planes. Some of the vacancies and interstitial atoms are immediately annealed by recombination, but those remaining may concentrate, depending upon the conditions of neutron fluence and irradiation temperature, and form larger clusters.

Since the German HTR concepts are based on unexchangeable reflector components, lifetime fluences are particularly high ranging from 3 to  $4 \times 10^{22}$  neutrons/cm<sup>2</sup> on the so-called Equivalent-Dido-Nickel (EDN) scale. Therefore, irradiation testing of HTR core materials is done in material test reactors such as the High Flux Isotope Reactor (HFIR) at Oak Ridge National Laboratory or the High Flux Reactor (HFR) at Petten Joint Research Center, where significant neutron fluences are accumulated at high neutron flux.

Table 2  
Properties of graphite V483 for core support columns

Property <sup>a)</sup>	Grade				
	T	T2	T5	T6	
Ash content (ppm)	240	70	13	32	
Density (g/cm <sup>3</sup> )	1.76	1.78	1.78	1.77	
CTE (20–500 °C) (10 <sup>-6</sup> /K)	(  )	3.57	3.59	3.83	3.93
	(⊥)	4.01	3.87	4.18	4.48
Anisotropy factor	1.12	1.08	1.09	1.14	
Youngs' modulus (kN/mm <sup>2</sup> )	(  )	9.5	9.5	11.1	11.3
	(⊥)	8.4	9.1	10.7	10.9
Bending strength (N/mm <sup>2</sup> )	(  )	28.6	38.9	51.6	55.3
	(⊥)	25.0	37.7	47.3	51.3
Compressive strength (N/mm <sup>2</sup> )	(  )	59.9	77.8	103	111
	(⊥)	62.1	79.9		
Tensile strength (N/mm <sup>2</sup> )	(  )	17.2	25.0		
	(⊥)	15.5	24.5		
Thermal conductivity (W/m K)	(  )	143	111	94	86
	(⊥)	134	106	87	79

<sup>a)</sup> (||) = parallel, (⊥) = perpendicular to grain orientation.

As the irradiation temperature in HFIR experiments is calculated before and verified after the irradiation (monitoring and controlling equipment are not available during irradiation), in the German research programme HFIR data are only used to compare the irradiation behaviour of different graphite grades, whereas design data are created in the High Flux Reactor (HFR) Petten at neutron fluences about 4 times smaller, but at well controlled temperatures. Capsules equipped with thermocouples and containing about 90 to 120 specimens are kept at constant temperature by gas mixture and by shifting the capsule vertically as fuel is burned and control rods are raised.

The first batch of ASR-1RS graphite was produced in 1975 when the graphite grade ATR-2E (made from special pitch coke) was still under consideration and had already proven its good irradiation behaviour. To determine whether using the secondary coke technique and vibrational moulding is a better way to produce reactor graphite, and to focus the development pro-

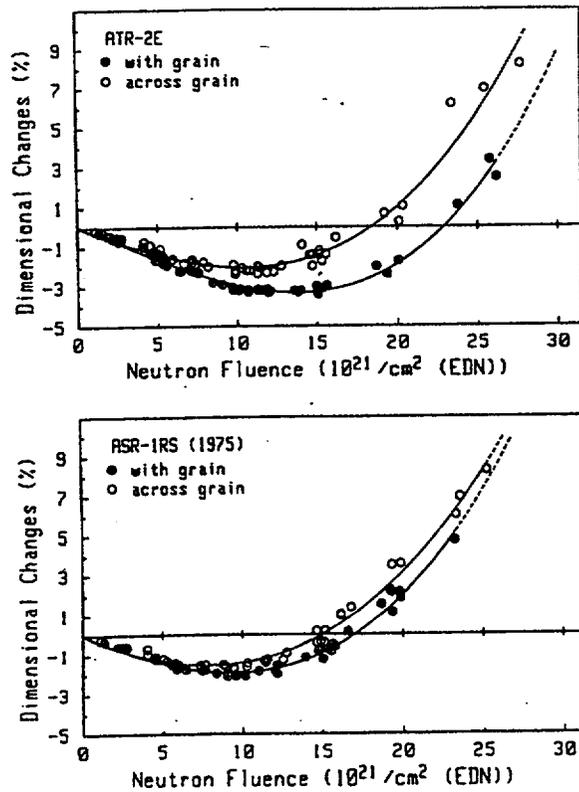


Fig. 4. Irradiation behaviour of extruded ATR-2E graphite at 500°C in comparison with the reference grade ASR-1RS (1975).

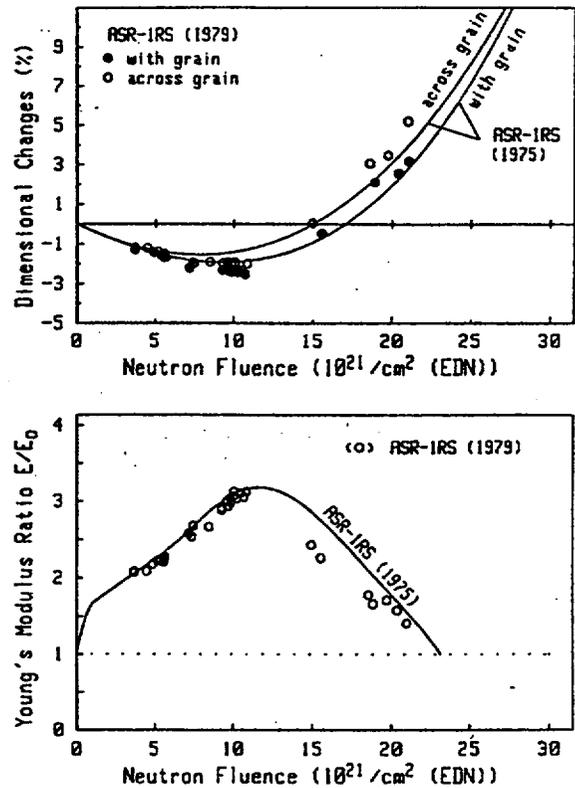


Fig. 5. Irradiation behaviour of ASR-1RS (1979) graphite at 500°C in comparison with the reference grade ASR-1RS (1975).

gramme on the most promising materials, it was necessary to get to know the irradiation behaviour of ASR-1RS as soon as possible. Therefore, all candidate graphites have been irradiated in three steps in HFIR. After accumulation of  $2 \times 10^{22}$  neutrons/cm<sup>2</sup> (EDN) (which is equivalent to about one year effective irradiation time) significant differences between the candidate graphites became visible. With regard to its very small anisotropy and good strength, ASR-1RS graphite was defined as a reference material for the highly loaded parts of the reflector.

Later, when the irradiation results from HFR Petten experiments became available, this decision was confirmed. Fig. 4 shows that ATR-2E graphite exhibits higher maximum shrinkage and higher anisotropy than ASR-1RS (both leading to higher irradiation induced stresses), but assumes the original length at a higher fluence in both directions. However, considerably larger blocks are manufactured by the vibrational moulding technique compared to extruding. In all cases where the

critical stresses occur in the across grain direction when the extruded ATR-2E material is used, the vibrated ASR-1RS graphite can be used in the with grain direction, and from fig. 4, it can be seen that the dimensional changes of ATR-2E across grain and of ASR-1RS with grain are almost the same.

In HFR Petten, ASR-1RS (1975) graphite has accumulated a maximum neutron fluence of about  $3 \times 10^{22}$  neutrons/cm<sup>2</sup> (EDN) in about 10 years. Its changes of linear dimensions, Young's modulus, thermal conductivity, and coefficient of thermal expansion (CTE) by irradiation are known.

In the meantime, other grades (see table 1) have been developed and the question was raised whether their more or less different physical properties would lead to a different irradiation behaviour. However, the irradiation testing of all these later produced grades is not yet complete. Nevertheless, it looks as if their behaviour is

not too much different from ASR-1RS (1975). This result is somewhat unexpected; before irradiation, ASR-1RS (1979) exhibits higher density, smaller CTE, and higher strength compared to the reference graphite but shows almost the same relative changes during irradiation (fig. 5).

The same impression is given by the ASR-2RS data in fig. 6. Even a second impregnation, yielding a bulk density as high as 1.87 g/cm<sup>3</sup>, does not affect either the dimensional or the Young's modulus behaviour significantly.

If these results can be confirmed and are also valid at temperatures different from 500°C, it could be concluded that the irradiation behaviour of graphite can be easily reproduced from batch to batch if only the same raw materials are used. Moreover, the irradiation testing of the graphite ASR-1RG (manufactured with regular pitch coke) will show if the secondary coke technique is essential for good irradiation behaviour.

For reactor designers these results may be reassuring. However, the question "How can the irradiation behaviour of reactor graphite be predicted from its physical properties?" remains as open as it was almost twenty years ago when G.B. Engle and W.P. Eatherly emphasized [5]: "The mechanisms of irradiation damage and crystallite changes and the relationships between crystallite and bulk dimensional changes have not been developed to the point where dimensional and volumetric changes of reactor graphites can be predicted accurately from pre-irradiation properties or structural features."

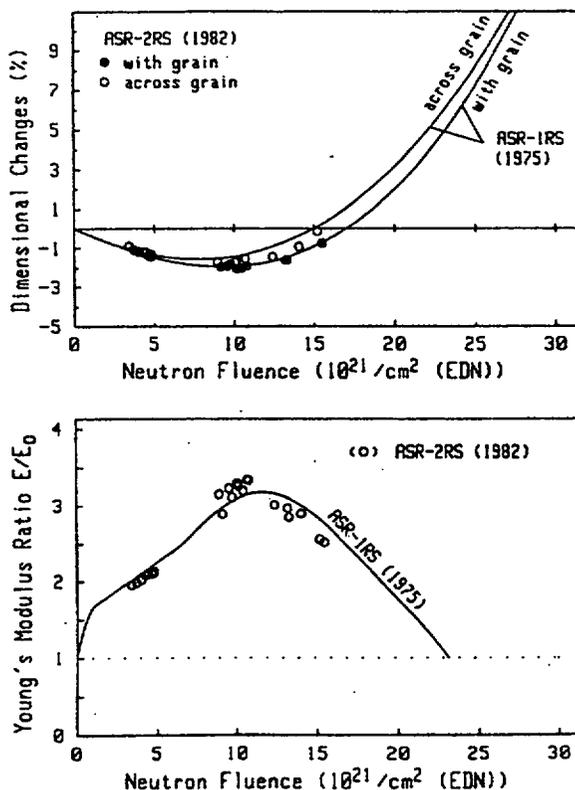


Fig. 6. Irradiation behaviour of ASR-2RS (1981) graphite at 500°C in comparison with the reference grade ASR-1RS (1975).

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