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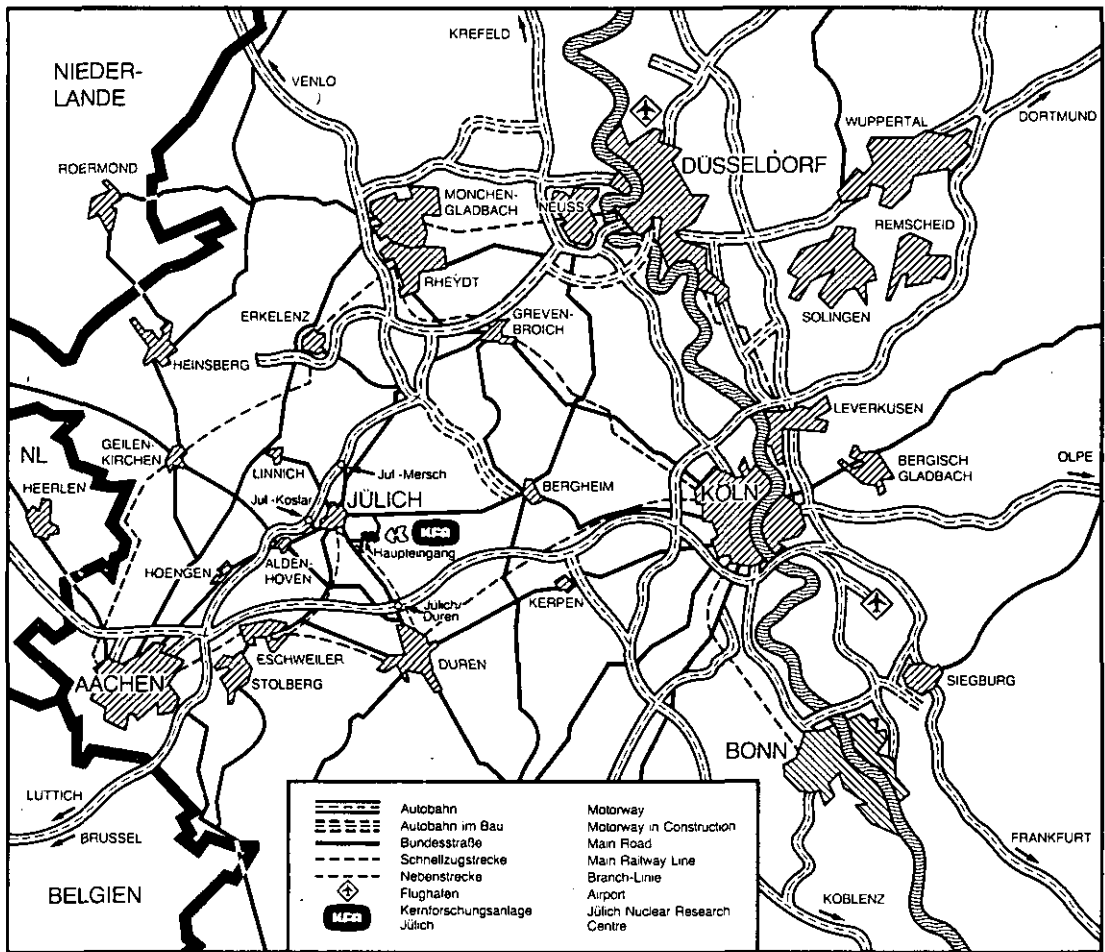
**Contributions for the
International Carbon Conference**

CARBONE 84

**Papers to be read at the Conference
in Bordeaux
July 2nd - 6th, 1984**

compiled by
W. Delle

**Jül - Conf - 49
Juni 1984
ISSN 0344-5798**



Als Manuskript gedruckt

Berichte der Kernforschungsanlage Jülich – Jül - Conf - 49

Zu beziehen durch: ZENTRALBIBLIOTHEK der Kernforschungsanlage Jülich GmbH

Postfach 1913 · D-5170 Jülich (Bundesrepublik Deutschland)

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**Contributions for the
International Carbon Conference**

CARBONE 84

compiled by

W. Delle

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BEITRÄGE ZUR INTERNATIONALEN
KONFERENZ IN BORDEAUX
CARBONE 84

OBER KOHLENSTOFF UND GRAPHIT
VOM 2. - 6. Juli 1984

ZUSAMMENGESTELLT VON
W. DELLE

KURZFASSUNG

Dieser Bericht enthält die Zusammenstellung von Beiträgen der KFA Jülich GmbH für die International Carbon Conference CARBONE 84, die in Bordeaux/Frankreich abgehalten wird. In den Beiträgen werden Fragen der Herstellung sowie der technischen und nuklearen Anwendung kohlenstoffhaltiger Materialien behandelt.

CONTRIBUTIONS FOR THE
INTERNATIONAL CARBON CONFERENCE
CARBONE 84.

ON
CARBON and GRAPHITE
JULY 2nd - 6th, 1984

COMPILED BY
W. DELLE

ABSTRACT

This report is the compilation of some papers prepared by KFA Jülich GmbH for the International Carbon Conference CARBONE 84, which will be held at Bordeaux/France. The presentations deal with objectives of manufacture as well as technical and nuclear applications of carbeneous materials.

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1. Einleitung

Die Groupe Francais d' Etude des Carbones (G.F.E.C.) veranstaltet im Jahre 1984 erstmals eine internationale Kohlenstofftagung. Sie fügt sich damit in die bisher von der Society of Chemical Industry in Großbritannien und vom Arbeitskreis Kohlenstoff der Deutschen Keramischen Gesellschaft in der Bundesrepublik Deutschland regelmäßig veranstalteten Kohlenstoffkonferenzen ein. Die europäischen Konferenzen finden alle 2 Jahre jeweils zwischen den amerikanischen Konferenzen statt, die alle 2 Jahre durchgeführt werden. Tagungsort für die CARBON'82 war London; die CARBON'86 ist für Baden-Baden vorgesehen.

Folgende Arbeitsgebiete werden auf der CARBONE 84 behandelt:

- Struktur und Textur
- Oberfläche und Reaktivität
- Physikalische Eigenschaften
- Kohlenstofffasern
- Kohlenstoffverbundwerkstoffe
- Intercalation
- Chemischer Aufbau und Mesophasenbildung
- Nukleare Anwendungen

Im Institut für Reaktorkeramik der KFA Jülich wird seit vielen Jahren auf dem Gebiet des Kohlenstoffs gearbeitet. Waren es zunächst grundsätzliche Fragen über die Strahlenschädigung von Graphit bei der nuklearen Anwendung, so hat sich der Schwerpunkt nunmehr auf Fragen des Verhaltens unter extremen Bedingungen, d.h. nach Akkumulation sehr hoher Neutronenfluenzen bei Temperaturen bis etwa 1000°C, verlagert. Auch Möglichkeiten zur Erweiterung der Materialpalette für die graphitische Matrix in kugelförmigen Brennelementen werden geprüft, indem Neuentwicklungen der Industrie im Materialtest-Reaktor erprobt und die Ergebnisse ausgewertet werden.

In den letzten Jahren wurden, aufbauend auf Erfahrungen aus der nuklearen Anwendung, neue kohlenstoffhaltige Materialien entwickelt, die als Gußformen in der Glas- und Metallindustrie besonders gut verwendet werden können. Dieses sog. Coat-Mix-Material kann mit hoher durchgehender Porosität hergestellt werden, was seine Imprägnierung mit flüssigen Medien erlaubt. Es

bietet damit Möglichkeiten zur Herstellung von Siliziumkarbid, auch mit definierten Überschüssen an Kohlenstoff und Silizium, wie sie z.B. bei der Verwendung als Gleitringdichtungen für das Targetrad der geplanten Spallations-Neutronenquelle (SNQ) in Erwägung gezogen werden. Auch kohlenstoff-imprägnierter Graphit, eine andere Variante der SNQ-Gleitringmaterialien, kann mit Hilfe des Coat-Mix-Verfahrens entwickelt werden.

Der vorliegende Bericht zeigt, wie - ausgehend von der Anwendung von Kohlenstoffmaterialien für den Hochtemperatur-Reaktor - diese Werkstoffe in der KFA nunmehr für weitere fortschrittliche Technologien entwickelt und erprobt werden. Er läßt erkennen, daß der Kohlenstoff bei der Neuorientierung der KFA auf zukünftige Aufgabenstellungen einen wichtigen Platz einnimmt.

Der Bericht enthält die Beiträge, die von Mitarbeitern der KFA maßgeblich erarbeitet wurden und auf der CARBONE 84 von ihnen vorgetragen werden.

PROPERTIES AND APPLICATIONS OF COAT-MIX MATERIALS

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General description

In developing new materials for reactor technology, a new process for the uniform coating of filler grains with synthetic resins has been developed in the Institute of Reactor Materials at the KFA. New, versatile materials have been obtained with this so-called coat-mix process. Materials fabricated by the coat-mix (CM-) process have been successfully tested in many industrial applications. These materials are of special significance in the glass industry, for metal casting, and high temperature filters. The following features are to be emphasized:

Homogeneous and isotropic grain structure; high permeability for gases and liquids; excellent reproduction accuracy, both in moulding the pattern and also the subsequent casting; no smoothing agent is necessary; outstanding resistance to thermal shocks; high temperature resistance; long tool life.

Parameter study

In order to become acquainted with the range of properties of CM-materials a parameter study has been made, using petroleum and pitch coke as the filler material and phenolic resin as the binder. The conditions of the production of test specimens have been reported earlier*. After the coking step each test specimen was cut into two parts, one part was characterized and the other HT-treated and then characterized.

The following properties have been determined:

Powder:

BET-surface before and after the coating process; bulk density; compacted apparent density; screen analysis.

Test specimen green:

height of the test specimen after pressing; apparent density.

Test specimen after cooking (800°C):

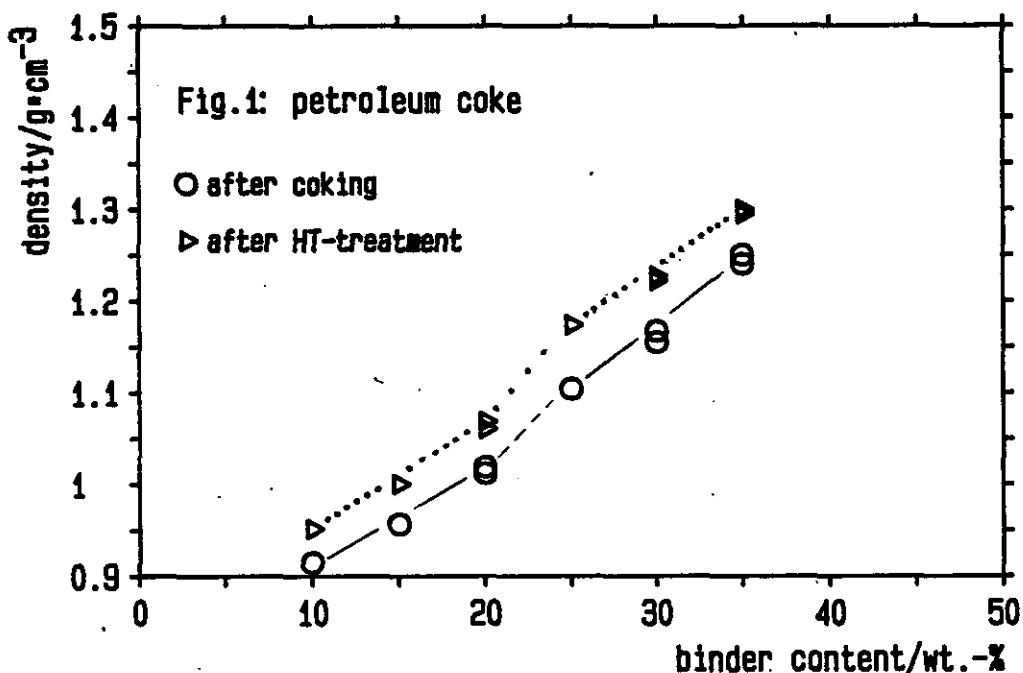
Shrinkage; density; total porosity; open porosity; air permeability; CTE, thermal expansion up to 500°C; thermal conductivity; electrical conductivity; Youngs modulus; compressive strength; bending strength.

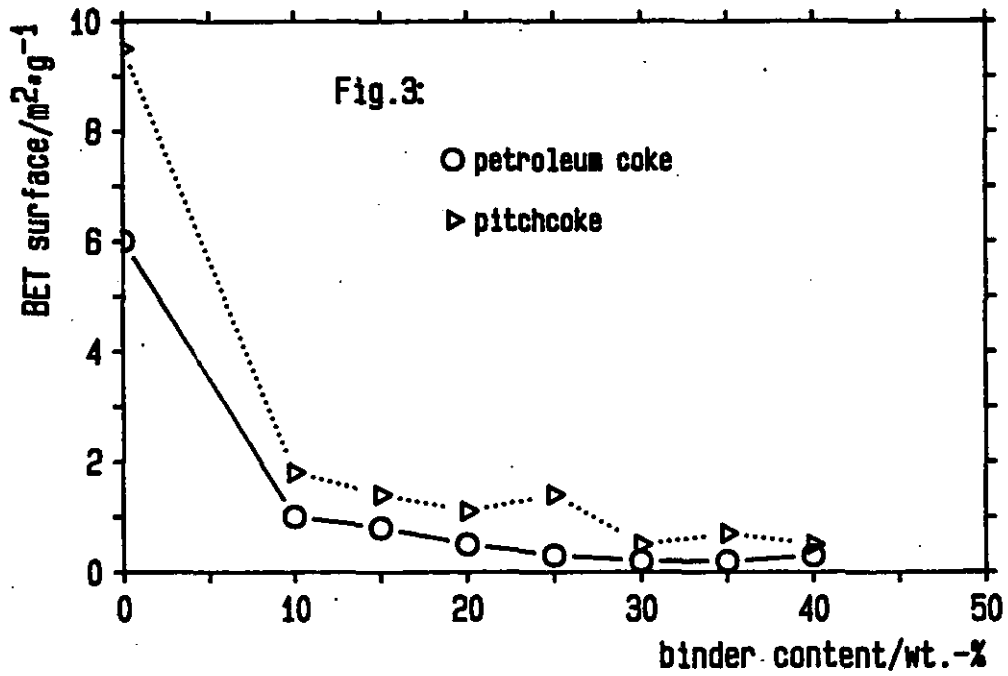
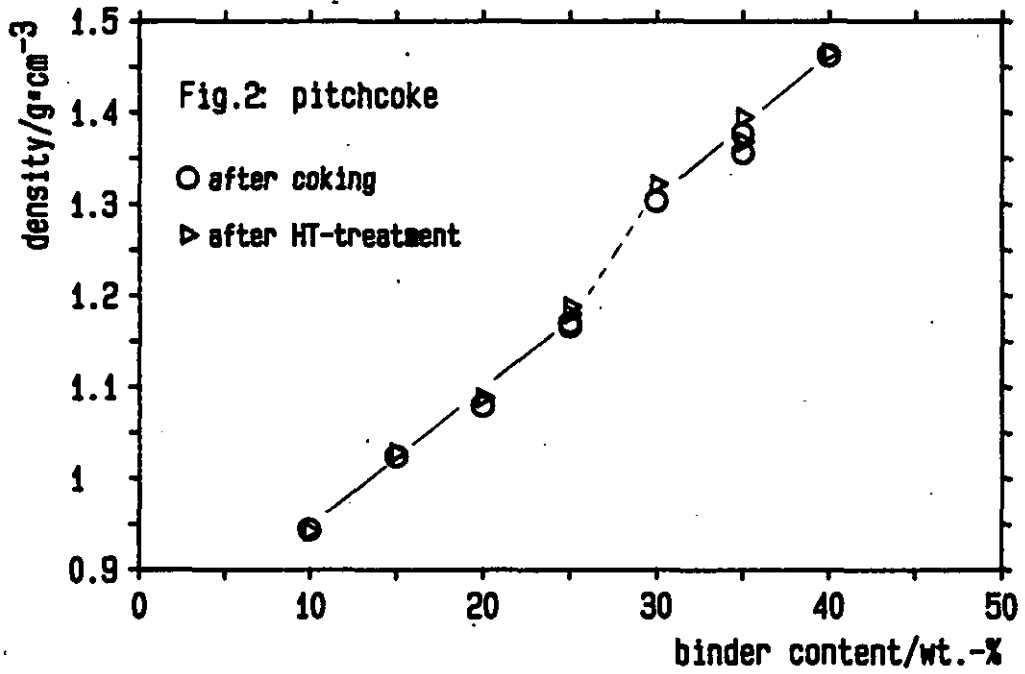
Test specimen after HT-treatment (1600°C, vacuum):

Characterization like after coking and CTE, thermal expansion up to 1100°C

In general, in case of most of the properties the characterization data show a steady dependence on the binder content between 10 wt.-% and 40 wt.-%, for both of the used filler types. Examples are is given in Fig. 1 and Fig. 2, showing the density of the specimen after coking and HT-treatment. The effectivity of the coating process can be deduced from the results shown in Fig. 3.

* Extended Abstracts of the 16th Biennial Conf. on Carbon,





CARBON MATERIALS FOR SLIDING RING SEALS

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1. Introduction

The use of sliding ring seals in pump systems of nuclear power stations as well as in the target support and drive unit of the planned German Spallation Neutron Source¹⁾ (SNQ*) require suitable ring material combinations. Besides silicon carbide with excesses of silicon and/or carbon, special sliding carbons with various impregnations (i.e. C or Sb) are under consideration for application^{2,3)}.

The two special sliding carbons (Sb or C impregnated) mainly consist of carbonized, but ungraphitized carbon, containing parts of graphitized carbon which are responsible for the self-lubricating properties. As expected, their thermal conductivity is very low and their anisotropy, determined from thermal expansion (α), amounts to about $\alpha_{\perp} / \alpha_{\parallel}$ 1,2.

The carbon materials distinctly differ from each other by porosity as well as surface roughness being higher in the case of the so-called all-carbon product; accordingly, the latter exhibits more reservoir for water storage.

In order to provide back-up materials particularly with a view to satisfactory irradiation behaviour, developments are being performed at KFA Jülich⁴⁾.

The principle of sliding seals is based upon the pressing of two plane ring surfaces against each other and on the coincident rotation of one of them against the other, the latter being stationary (Fig. 1).

* from German: Spallations-Neutronenquelle

The two sliding seals differing by the kind of friction are:

- type I with mixed (solid/liquid) friction for sealing of the cooling water from the internal vacuum space (1)
- type II with technical dry (solid/solid) friction for retention of the water vapour, being in the internal vacuum space, from the external one (2).

2. Materials and investigations

A few combinations of ceramic materials can be taken into consideration as candidates for the seals material as shown in Tab. 2.

From physical data measurements and from microstructure investigations the following can be said about the materials Si-SiC, carbon (Sb) and carbon (C). The high density ($\rho = 3.08 \text{ g/cm}^3$; $\rho_{th.} = 3.21 \text{ g/cm}^3$) Si-SiC consists of selfbonded (α - and β -phase) SiC as the structural part, and of the metallic Si-phase, filling the skeleton pores. The SiC grains of different sizes show a good isotropic distribution. Both the ceramic as well as the metallic materials components are of very high purity. All this indicates a good quality of the sliding ring of SNQ dimensions.

The two special sliding carbons (Sb or C impregnated) mainly consist of carbonized, but ungraphitized carbon, containing parts of graphitized carbon which are responsible for the self-lubricating properties. As expected, their thermal conductivity is very low and the anisotropy of their thermal expansion amounts to about $\alpha_{\perp}/\alpha_{\parallel} = 1.2$. The carbon materials distinctly differ from each other in porosity as well as in surface roughness which is higher for the all-carbon product carbon (C); accordingly, the latter exhibits more space for water storage.

For the combination Si-SiC/carbon (Sb), experience is available from nuclear reactor power stations. Problems resulted from the Sb isotopes in the cooling water (2,3), but the SNQ conditions are in part more favourable for the use of this material. Standard tests on sliding ring seals consisting of the combinations

Si-SiC/carbon (C impregnated)
Si-SiC-C/carbon (C impregnated)
Si-SiC-C/carbon (Sb impregnated)

performed by the Pacific company, Dortmund showed that the first combination seems to be most favourable for the seal of type I, whereas the others might be used for type II.

For the materials investigations, sliding rings of SNQ dimensions were partitioned into 5 equal segments of identical sample sets for different purposes. Machining of SiC materials was done by the electrical discharge machining (EDM). Additional investigations are being carried out with integral sliding rings (e.g. determinations of the mechanical strength) at the Pacific company.

3. Irradiation testing

During SNQ operation, the sliding seals will be exposed to high energy spallation neutrons. The operation conditions relative to the running time, the temperature and the fast neutron fluence (with reference to the different target materials) are shown in Tab. 1.

Table 3 shows the programme intended for irradiation testing of the sliding ring materials. The major objective of the experiments is to establish a data base for the materials after fast neutron exposure, completed by investigations of irradiation induced changes in the microstructure of the materials.

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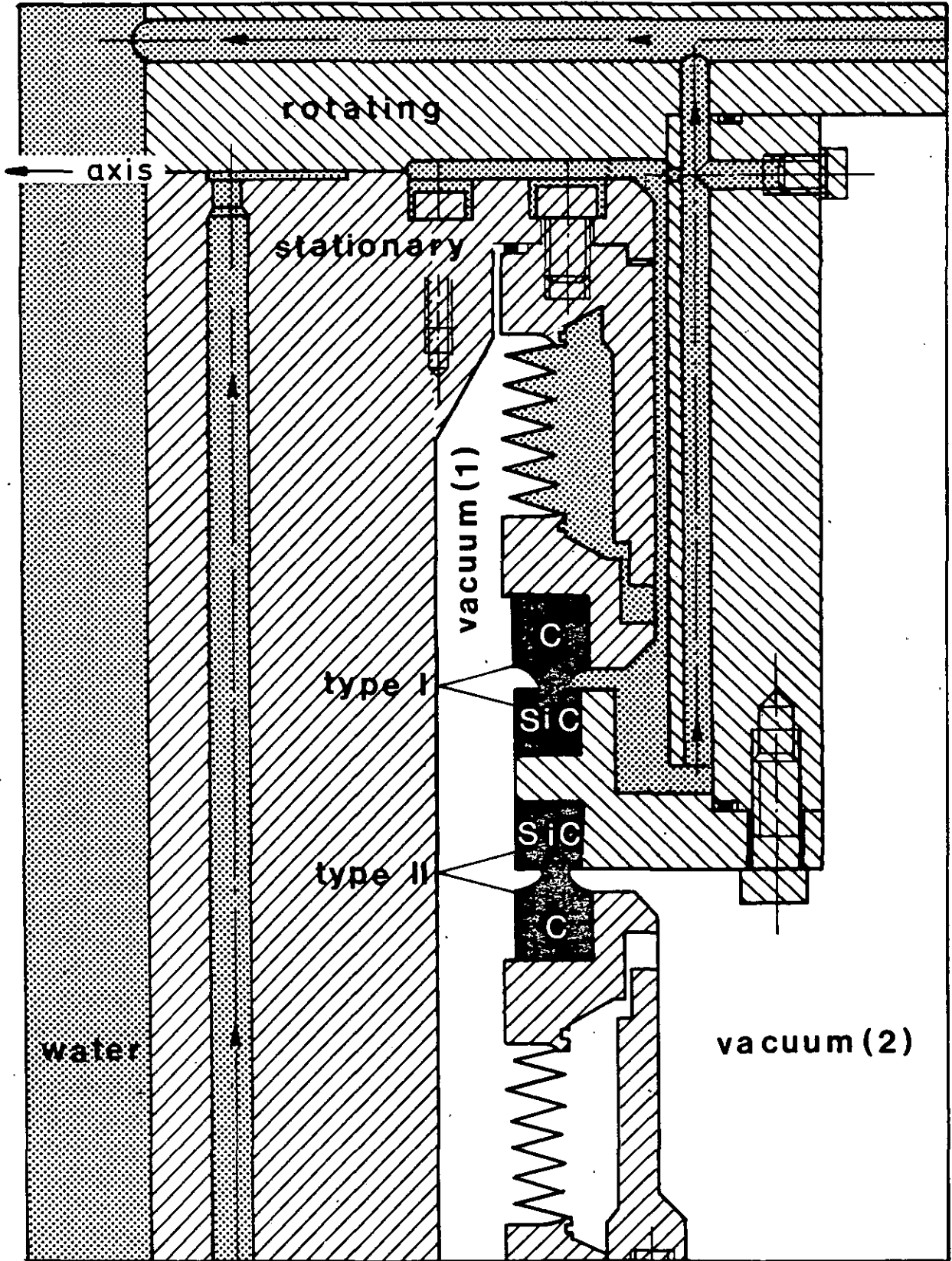


Fig. 1: Arrangement of the sliding ring seals in the target support and drive unit of the SNQ

<ul style="list-style-type: none"> ● time of operation : 12000 h ● max. operating temperature : 70°C (type I); 200°C (type II) ● accumulated fast neutron fluence ($\text{cm}^{-2} E > 0,1 \text{ MeV}$) detailed with reference to the target materials and the steps of development for SNQ: 			
SNQ step of development	Target material		
	Pb	W	^{238}U
SNQ - I	$\sim 5 \cdot 10^{18}$		$\sim 1 \cdot 10^{19}$
SNQ - II	$\sim 5 \cdot 10^{19}$		$\sim 1 \cdot 10^{20}$

Table 1: Operation conditions for SNQ sliding ring seals

Sliding ring seal	Material combination for sliding rings			
	proposed for the seal in the SNQ	being developed for the SNQ	used for seals in reactor power stations	provided for
mixed friction	Si-SiC/carbon C or Sb impregn. ¹	Si-SiC _{mod.} ² / Si-SiC _{mod.} ²	Si-SiC/carbon Sb impregn.	Si-SiC-C/ Si-SiC-C
			WC / carbon Sb impregn.	
dry friction	Si-SiC-C/carbon C or Sb impregn. ³	Si-Si-C _{mod.} ³ / carbon _{mod.} ³ C impregn.	—	—

¹ sliding carbon impregnated with carbon or Sb

² or Si-SiC-C_{mod.} / Si-SiC-C_{mod.}, both combinations on the basis of coat-mix material

³ on the basis of coat-mix material

Table 2: Material combinations for the sliding seals of type I and type II

Irradiation test		Irradiation data		Environ - ment	Capsule loading	
at/in	for SNQ seal of type	tempe - rature (°C)	fast neutron fluence (cm ⁻² E>0,1 MeV)		test samples	material or material combination
Proton accelerator LAMPF (Los Alamos)	I*	- 50	- 6 · 10 ¹⁹	air	samples machined from SNQ sliding rings	Si - SiC carbon (C) carbon (Sb)
			- 2 · 10 ²⁰			Si - SiC - C ¹ Si - SiC carbon (C) carbon (Sb)
	II**	- 120 - 150	- 2 · 10 ¹⁹	air		Si - SiC ² Si - SiC _{mod.} Si - SiC - C ³ Si - SiC - C _{mod.} carbon (C) carbon _{mod.} (C) carbon (Sb) steel
			- 6 · 10 ¹⁹			
Test reactors FRJ - 1 and HFR Petten	I	- 70	- 6 · 10 ¹⁹	Acionat		
			- 2 · 10 ²⁰			
	II	- 200	- 6 · 10 ¹⁹	inert gas		
			- 2 · 10 ²⁰			
Test reactor HFR Petten	I	- 70	- 2 · 10 ²⁰	Acionat	sliding ring component ⁴	1. Si - SiC/ carbon C impregn.
II	- 200	- 2 · 10 ²⁰	inert gas	1. Si - SiC - C/ carbon C impregn.		
						2. Si - SiC - C _{mod.} / carbon _{mod.} C impregn.

* with mixed friction ** with dry friction ¹ irradiation testing only possible in 1 experiment
² main material in the experiments for seal type I. ³ main material in the experiments for seal type II
⁴ sliding ring with steel holder (ring ϕ - 100 mm)

Table 3: Programme for irradiation testing of materials for SNQ sliding ring seals

WARM-MOULDED GRAPHITIC MATRIX FOR SPHERICAL HTR FUEL ELEMENTS

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The development and irradiation testing of graphitic matrix materials for cold-moulded HTR fuel elements have been completed successfully with the use of two approved materials for the fuel elements of the reactors AVR (matrices A3-3 and A3-27) and THTR (matrix A3-3).

In order to enable the manufacture of fuel elements with high heavy-metal loading, as well as to improve the fabrication technology, the NUKEM/HOBEG company developed a process for the warm-moulding of the elements (Fig. 1).

Furthermore a pitch coke graphite was used as a basic raw material for the graphitic matrix instead of the raw material mixture of natural graphite and petroleum coke graphite used before [1].

Tab. 1 shows the composition of the warm-moulded matrices W2-1 and W2-2 as well as of the cold-moulded fuel matrices A3-3 and A3-27 and also of 4 further variants of A3-3 designated as A3-18, A3-17, A3-16 and A3-21. These variants were manufactured with addition of hardener to the binder, this being adequate to the manufacture of W2-1 and W2-2. The relation between the warm-moulded matrices and the cold-moulded A3-3 variants concerning the binder coke content is demonstrated in Fig. 2. Additional relations exist with regard to the binder to hardener ratio and the filler to binder coke ratio. All these material parameters allow some comparisons of warm-moulded and cold-moulded matrices, thus contributing to a better understanding of the materials behaviour before and after neutron irradiation [2,3,4].

Comparing the material properties of the standard quality of the warm-moulded matrices (W2-1) with those of the cold-moulded fuel matrices A3-3(1950°C),

A3-3(1800°C) and A3-27 (Tab. 2), the first shows a substantially better strength behaviour. On the other hand, the corrosion rate of W2-1 is more unfavourable than that of A3-3(1950°C) and A3-27, although heat treated at the same temperature[1,2,5]. Moreover, moulding in a die, causes a higher quotient of thermal expansion coefficients ($\alpha_{\perp}/\alpha_{\parallel}$) for W2-1 and W2-2. Analogous to this, both warm-moulded matrices show slightly anisotropic dimensional behaviour under fast neutron exposure; furthermore they differ in shrinkage due to the different binder coke contents [1,6] (Fig. 3).

The thermal conductivity of W2-1 and W2-2 generally decreases under irradiation, at temperatures between 880 and 940°C approximating a saturation value of about 68 % of the pre-irradiation value (Fig. 4). Similar changes were determined for the cold-moulded matrices A3-3 and A3-27.

The results described were obtained within the framework of the HTR project "Hochtemperaturreaktor-Brennstoffkreislauf" (High-Temperature Reactor Fuel Cycle) involving the Gesellschaft für Hochtemperaturreaktor-Technik mbH, Hochtemperaturreaktor-Brennelement GmbH, Hochtemperatur-Reaktorbau GmbH, Kernforschungsanlage Jülich GmbH, NUKEM GmbH and Sigri Elektrographit GmbH/Ringsdorff-Werke GmbH. The project is sponsored by the "Bundesministerium für Forschung und Technologie" (Federal Ministry of Research and Technology) and by the State of North-Rhine/Westphalia.

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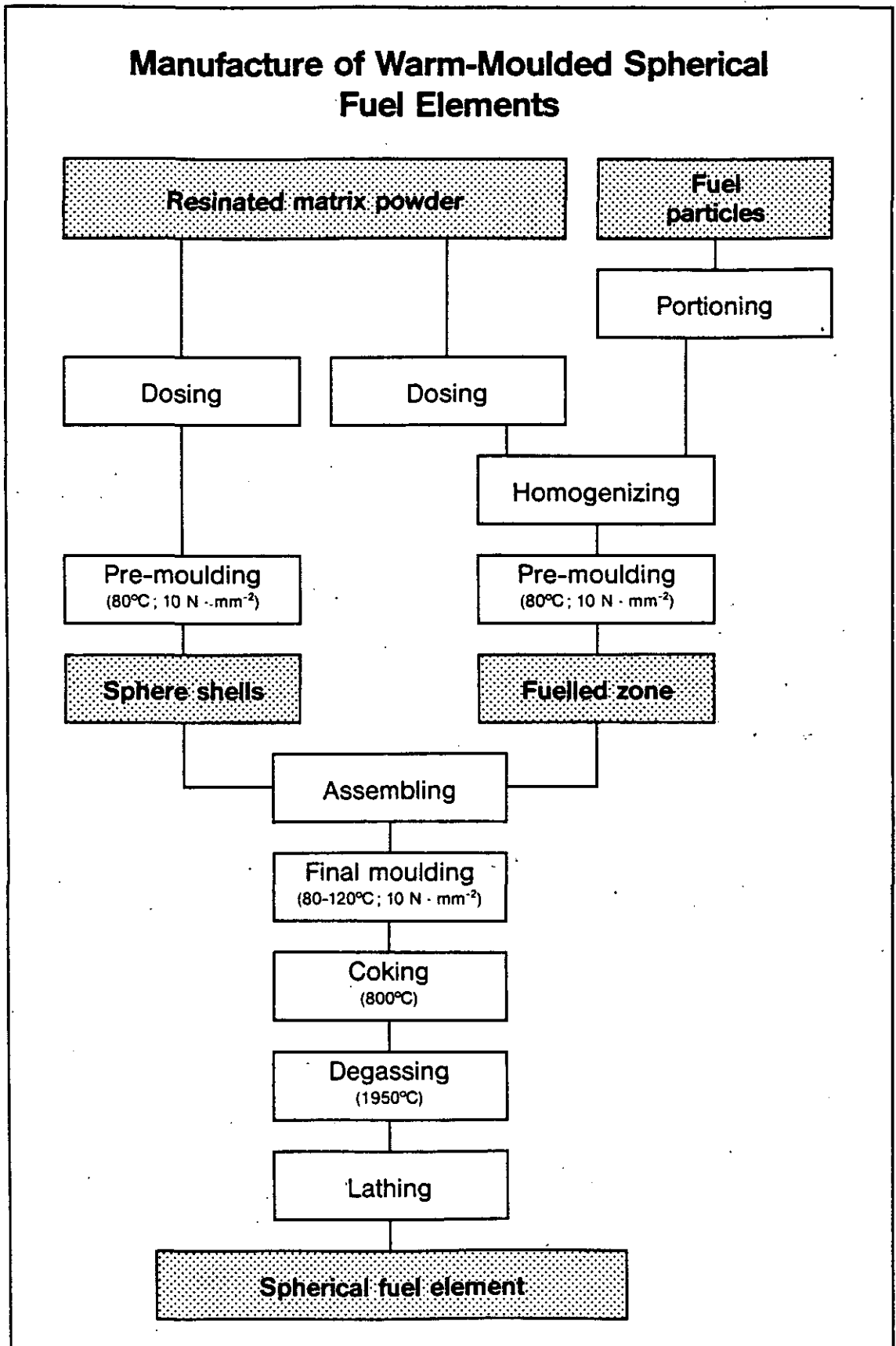


Fig. 1: Process for warm-moulding of spherical fuel elements

Graphitic matrix materials	Filler (wt. %)	Binder (wt. %)	Hardener (wt. %)**	Binder coke (wt. %)
warm-moulded matrices W2-1* W2-2	pitch coke	phenol formaldehyde	hexa-methylene-tetramine	9.8
	graphite			
cold-moulded fuel matrices A3-3 A3-27	84	16	5	11.0
	82	18	5	
	natural + petroleum graphite	phenol formaldehyde	phenol hexa-methylene-tetramine***	
	64	20	-	10.0
	62.4	-	22	11.0
cold-moulded A3-3 variants with hardener A3-18 A3-17 A3-16 A3-21	natural + petroleum graphite	phenol formaldehyde	hexa-methylene-tetramine	9.9
	64			
	16		2	11.3
	15.6		10	12.0
			20	11.9

* standard quality ** related to the binder content *** synthesized during matrix formation

Tab. 1: Composition of warm-moulded and cold-moulded matrices

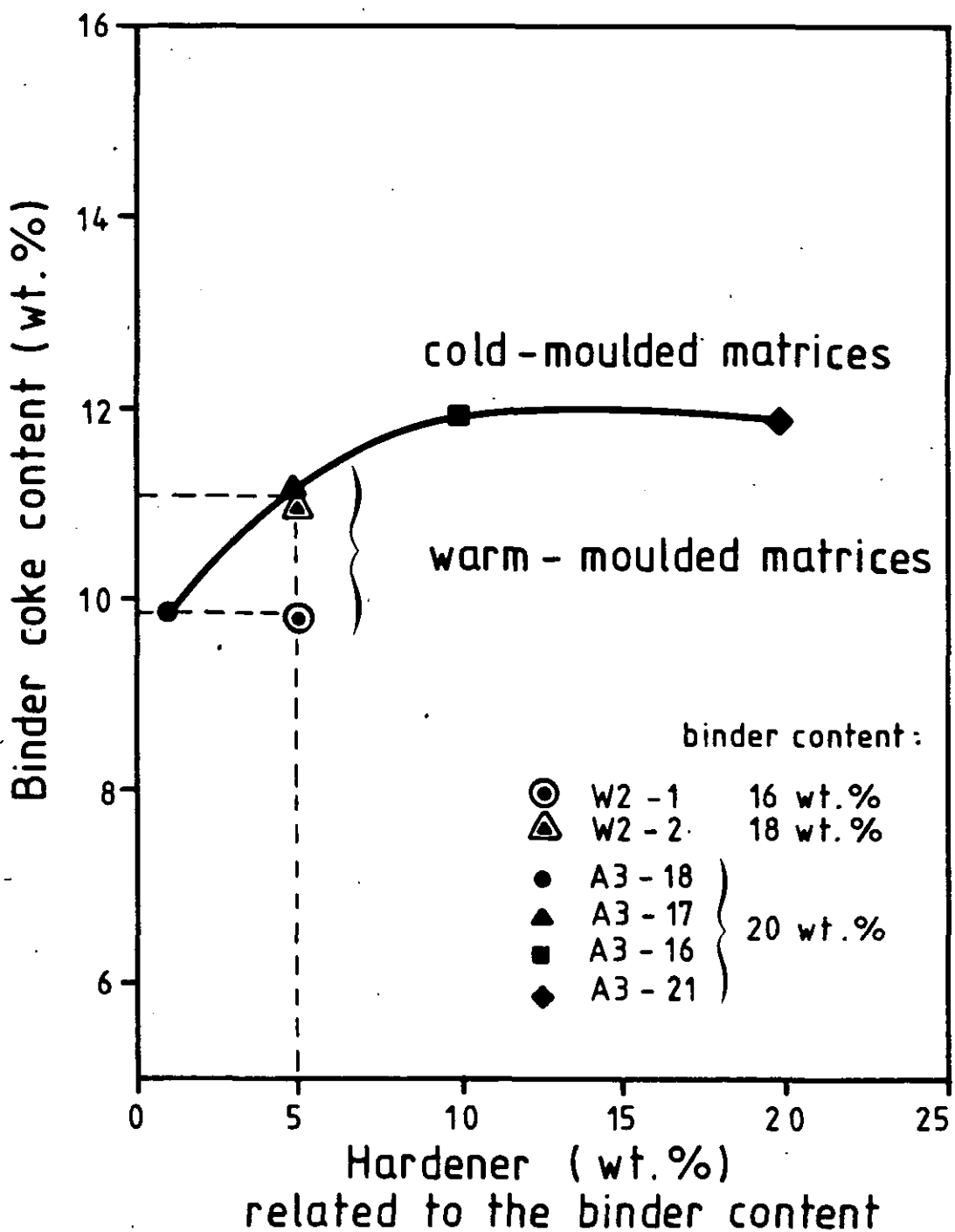


Fig. 2: Relation between warm-moulded and cold-moulded matrices

Material	High-temp. treatment (°C)	High-Application	Property								
			Young's modulus (kN · cm ⁻²) * I*	Density (g · cm ⁻³)	Coeff. of lin. therm. expansion 20 - 500°C I	Quotient of coeff. of therm. expansion α_I / α_{II}	Therm. conductivity at 1000 °C (W · cm ⁻¹ · K ⁻¹) I	Compressive strength (daN · cm ⁻²) I	Tensile strength (daN · cm ⁻²) I	Corrosion rate at 1000 °C in He of 1 bar with 1 vol. % H ₂ O (10h) (mg · cm ⁻² · h ⁻¹) I	
Warm-moulded matrices	1950	Development of warm-moulded fuel elements	1095	1.73	2.94	1.27	0.30	874	225	1.14	
			882	3.73	0.27	832	220				
Cold-moulded matrices	1950	AVR fuel elements, THTR Production Previous AVR fuel elements	1110	1.74	2.97	1.25	0.28	775	132	1.01	
			980	3.70	0.26	740	164				
Cold-moulded matrices	1800	AVR fuel elements, THTR Production Previous AVR fuel elements	1000	1.73	2.89	1.19	0.32	435	135	0.97	
			970	3.45	0.29	426	126				
Cold-moulded matrices	1950	AVR fuel elements	1020	1.70	2.80	1.07	0.28	382	104	1.19	
			991	2.92	0.27	376	105				
Cold-moulded matrices	1950	AVR fuel elements	1070	1.74	2.43	1.11	0.34	469	135	0.73	
			1020	2.69	0.32	464	130				

* parallel and perpendicular to the equatorial plane of the matrix sphere ** standard quality

Tab. 2: Material properties of warm-moulded and cold-moulded matrices

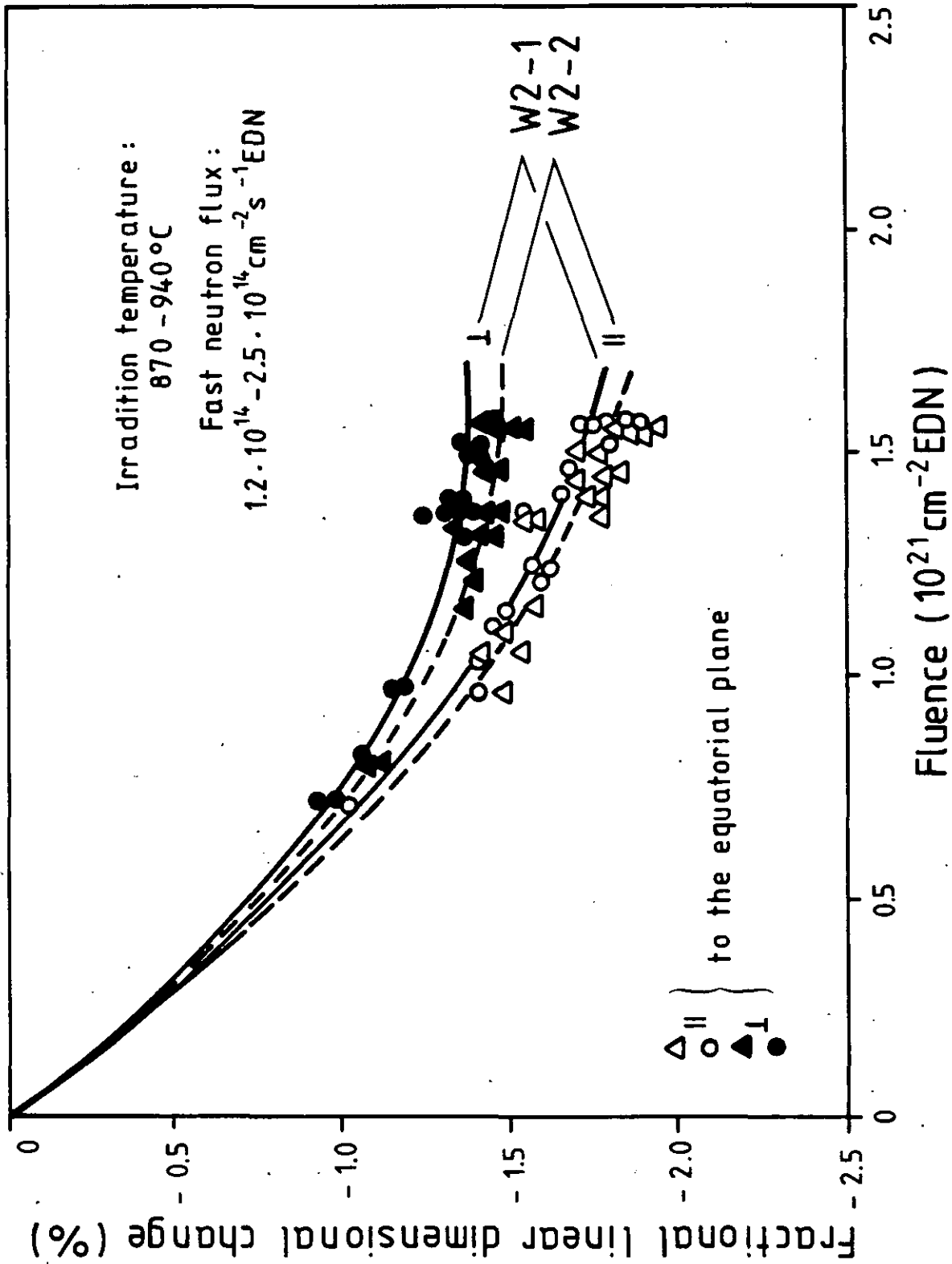


Fig. 3: Dimensional change of warm-moulded matrices versus fluence

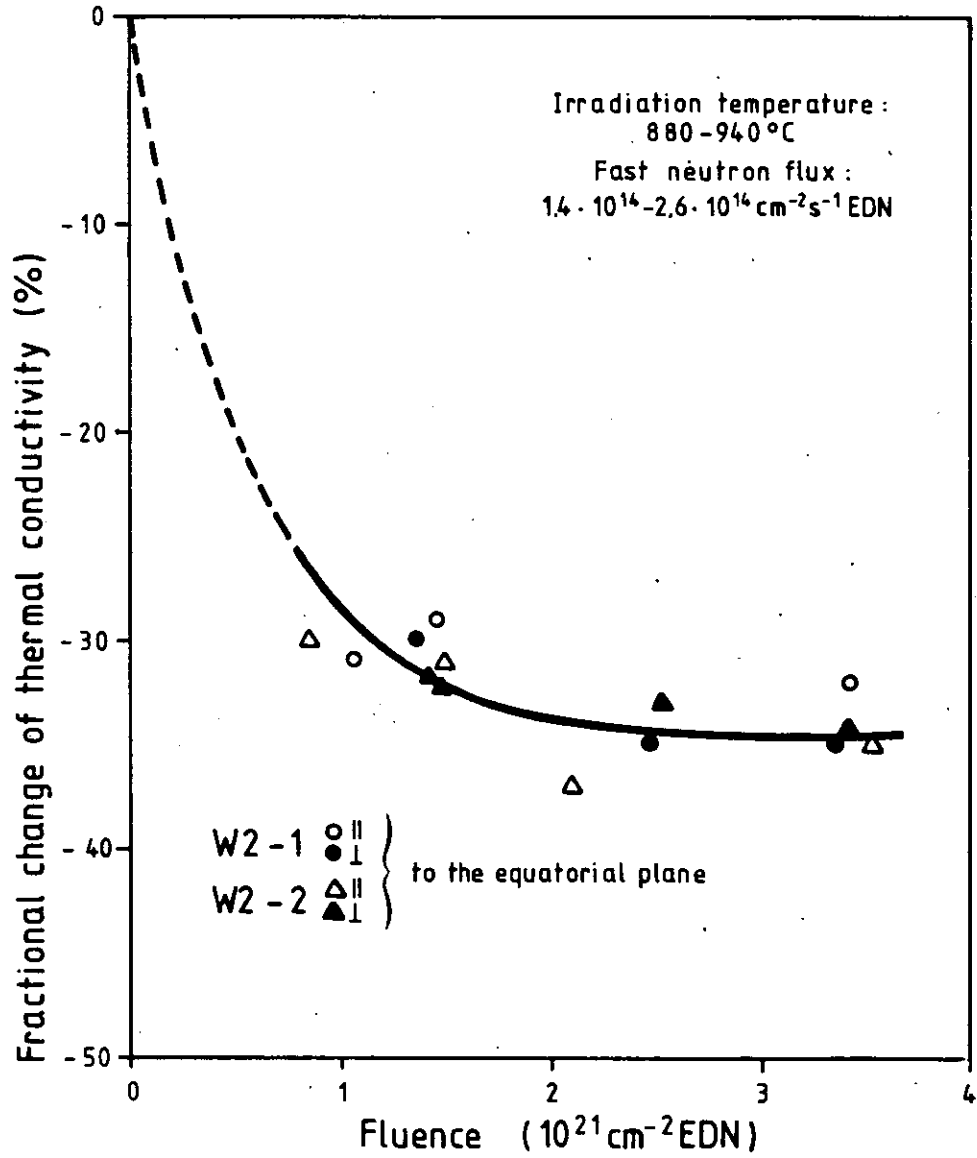


Fig. 4: Fractional change of thermal conductivity of warm-moulded matrices versus fluence

IRRADIATION INDUCED CREEP IN GRAPHITE WITH RESPECT TO THE FLUX EFFECT
AND THE HIGH FLUENCE BEHAVIOUR

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Irradiation induced creep of graphite has been investigated as part of a long term programme to qualify graphites for the pebble bed High Temperature Reactor. In accelerated irradiation creep tests, performed in the HFR Petten, in a fast neutron flux of about $2 \times 10^{14} \text{ cm}^{-2} \text{ s}^{-1}$ and at temperatures of 300 and 500°C, a fast neutron fluence in excess of $20 \times 10^{21} \text{ cm}^{-2}$ (EDN) has been attained so far. As a supplement to this, an analogous creep test was conducted in a fast neutron flux lower by a factor of four which is more typical for the service conditions in an HTR, with a maximum fast fluence of only $4 \times 10^{21} \text{ cm}^{-2}$ (EDN). This experiment was aimed at answering the question if, for equal fast fluence, enhanced irradiation creep and Wigner dimensional change would take place in a reduced fast neutron flux. This problem has more generally been addressed to as the "flux effect" or the "equivalent temperature concept" /1,2/.

The investigated graphite was a rather coarse grained, extruded pitch coke grade, manufactured by SIGRI Elektrographit GmbH as a side reflector material /3/.

Fig. 1 shows, as an example, the strain versus fluence behaviour for the compressive creep samples and unstressed references at 500°C, both the corrected measurement points and the curvilinear least squares fits. From diagrams like this, plots of irradiation creep strain ϵ_c and creep coefficient K versus fluence were derived as shown in Figs. 2 and 3 respectively. K is the creep strain rate with respect to fluence normalized to unit stress. Creep coefficients both tensile and compressive at 300 and 500°C first decrease with fluence when irradiation induced densification (Wigner shrinkage) and hardening (Young's modulus increase) take place. Thereafter,

the increase of K goes along with new pore generation, loss of strength and Young's modulus decrease. It may be assumed that when this occurs, creep mechanisms are being modified. This assumption is supported by the transversal creep strain behaviour suggesting a continuous decrease of Poisson's ratio in compressive creep from a value of about 0.5 to effectively zero, i.a. a change from volume conserving creep to void generating creep.

A comparison of the Wigner deformation, e.g. at 500°C in the low and high flux irradiations respectively, according to Fig. 4, gives no evidence of a significant flux effect. One might have expected this result from the calculation of equivalent temperatures with an effective activation energy of $Q = 1.2 \text{ eV}$ /1/ and consideration of the nearly temperature independent initial shrinkage rate in the range 500 to 700°C (high flux). The situation is somewhat different for the creep behaviour (Figs. 5 and 6). In both the 300 and 500°C irradiations, a remarkably higher primary creep strain at $4 \times 10^{20} \text{ cm}^{-2}$ /EDN) has been measured for the low flux conditions. Starting from this enhanced creep strain level, secondary creep for low flux tends to proceed with approximately the same rate as for high flux. Further consideration will be given to the understanding of the apparent sensitivity of primary creep strain rate to the damage flux during the period when the equilibrium primary defect structure in the graphite is being established.

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This work is being conducted within the framework of the Project "Hochtemperaturreaktor-Brennstoffkreislauf" (High Temperature Reactor Fuel Cycle) which includes the partners GHT GmbH, HOBEG mbH, HRB GmbH, KFA Jülich GmbH, NUKEM GmbH and SIGRI GmbH/Ringsdorf-Werke GmbH, and is financed by the Federal Ministry of Research and Technology and the State of Northrhine-Westfalia.

