PROPERTIES AND BEHAVIOUR UNDER IRRADIATION OF ISOTROPIC FINE GRAINED GRAPHITES

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Le Carbone Lorraine (LCL) has manufactured for a long time fine grained graphites with high properties. On the 1st table one notes the values of the properties of grades 3780, 2239, 5890 wich are commercial products.

To answer Nuclear Energy requirements LCL has prepared a fine grained isotropic material grade 7477 by isostatic molding. It's possible to obtain prismatic blocks useful for HTR and graphite elements for MSBR. In the last case, the fluence taken by the graphite can reach 4.10^{22} n/cm2 EDN* insteed of 5.10^{21} for the HTR. Consequently, it was interesting to prepare finer grained graphites than the classical nuclear products. The table 2 give the values of the properties of these graphites.

Grade 7477 is perfectly adequate for HTR. If we look at the irradiation properties of US materials and if we compare the properties before irradiation of the US materials and French very fine grained grades 8270 and S 1260, we can conclude that these two grades could be adequate for the MSBR.

IRRADIATION BEHAVIOUR

1 - HTR - The grades 3780 WEG, 5890 and 7477 were tested after irradiation by Kernforschungsanlage Jülich (1), Japan Atomic Energy Research Institute (2) and the French Atomic Energy Commission (3) at temperatures between 600°C and 1300°C with fluence riching 5.10^{21} n/cm2 EDN. Grade 7477 is the best : its dimensional change is generally < 1 % at temperatures between 600°C and 1100°C. The other grades are less interesting : contraction riching 1 % to 2 %.

The three figures (3, 4, 5) show the results obtained by the three laboratories and figure 6, all results compared with the same neutron unit ØFG :
ØFG ~ 1,85 EDN ~ 1,25 n > 0,18 MeV. It's important to note the sharp evolution of dimensional changes above 1100°C.

On the other side, KFA has studied the dimensional changes, versus doses and graphitization temperature of grade 2239 (fig. 7). (The temperature is given by the value of magneto-resistivity). The minimum dimensional change is given by the highest temperature of graphitization.

Finally Jaeri made corrosion's studies with water vapor and hydrogen at high temperatures. It showed that with H_2 the corrosion was very small with grade 7477.

2 - MSBR - Various grades of very fine grained graphite were irradiated in the French Reactor Osiris at temperatures of 800°C and 1300°C, with doses up to 2.10^{22} n/cm2 EDN. The results of irradiation are not get known, but we hope to have some of them for the conference.

With the aim of improving graphite's density and of reducing their porosity especially to have pore dimensions sufficiently low to have no penetration of fused salts, these graphites have been several times impregnated with a mixture of pitch and resin.

After impregnation, carbonisation and graphitization, the densities, porosities and largest pore dimensions have the values given on table 8.

These impregnated materials have also been irradiated in the same way.

COMMENTS

Grade 7477 is difficult to obtain in large pieces. Consequently, we developped a graphite with double texture.(4) The term "double texture" means a product obtained by molding aggregates of fine grained coke. This material is easier to bake and to graphitize than 7477 itself.

In the range of ultra fine grained graphite, it's necessary to note that it would be impossible to manufacture large pieces (like HTR blocks) but it's possible to make blocks or bars with at least one dimension less than two inches.

In that way, we, at LCL, continue to improve density and mechanical properties of graphites.

The better materials recently obtained in this way ; ultra fine grain, high mechanical properties and high density are shown in table 9. These better grades could be irradiated in 1978.

On the other hand, to improve density and to reduce porosity and pore dimensions, the study of impregnation continues on various grades of graphite with resins, pitch and chemical vapor deposition of carbon with vacuum and pressure technique.

As a conclusion Le Carbone Lorraine present materials which could be used for HTR and MSBR. Le Carbone Lorraine goes on developping this range of materials which corresponds better to it's capabilities than that of graphites used in gaz-graphite reactors.

Moreover, in cooperation with SERS, LCL hopes to make less expensive products at prices convenient for HTR. This is especially the case for the "double texture" graphite.

ACKNOWLEDGEMENT

We thank greatfully KFA, JAERI and CEA for the communication of the results obtained on our products after irradiation.

* 2.10^{22} turn around

(1) "Carbon 76" BADEN-BADEN - July 1976

(2) Private communication

(3) Note CEA-N-1903 - September 1976
P.MOTTET and G. MICAUD
"Graphite obtenu par pressage isostatique"
"Comportement sous irradiation neutronique"

(4) French patent n° EN 75-19678 - June 1975
J. FOURRE - J.P. GERVAIS - J. MAIRE
"Graphite à double texture"

grades / Characteristics	Unités	Tableau I			Tableau 2		
		3780	2239	5890	7477	S1260	8270
Density		1,60	1,80	1,82	1,75	1,65	1,76
Flexural strength RF	daN.cm^{2}	225	500	600	550	450	650
Thermal expansion 25 to 525°	10^{6}°C^{-1}	3	4	4,2	4,7	6,1	5
Porosity	%	24	10	8	12	25	13,8
Highest pore radius r	µ					0,3	0,8
Mean pore radius	µ					< 0,2	0,5

grades / Characteristics	Unités	Tableau 8			Tableau 9		
		7477 3H	8270 1H	S1260 1H	S1346	S1410	S1410 1H
Density		1,88	1,84	1,76	1,90	1,79	1,87
Flexural strength RF	daN.cm^{2}	690	650	590	800	750	800
Thermal expansion 25 to 525°	10^{-6}°C^{-1}	3,9	4,6	5,9	5,3	5,1	
Porosity	%	3,9	8,5	10,9	8,4	14,5	6,8
Highest pore radius r	µ	0,8	0,4	0,5	1,2	0,3	0,1
Mean pore radius	µ				1	0,2	0,06

1, 2, 3H means number of impregnations.

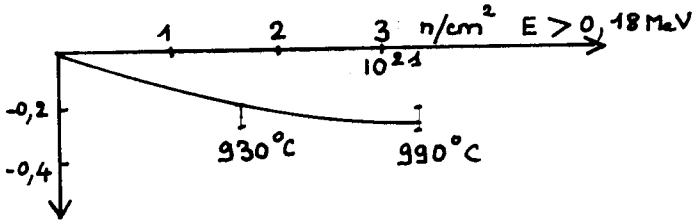

Fig. 3 - Dimensionals changes 7477 - JAERI

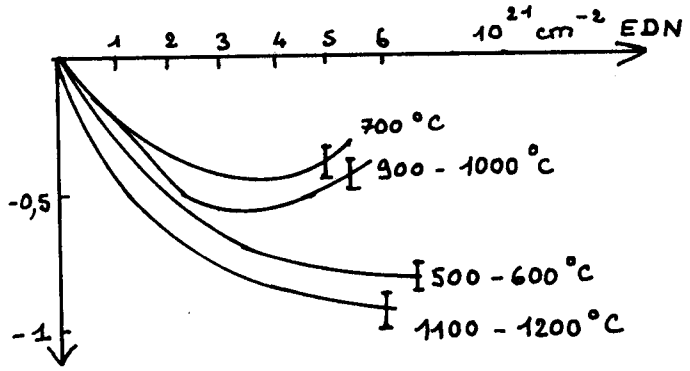

Fig. 4 - Dimensionals changes 7477 - KFA

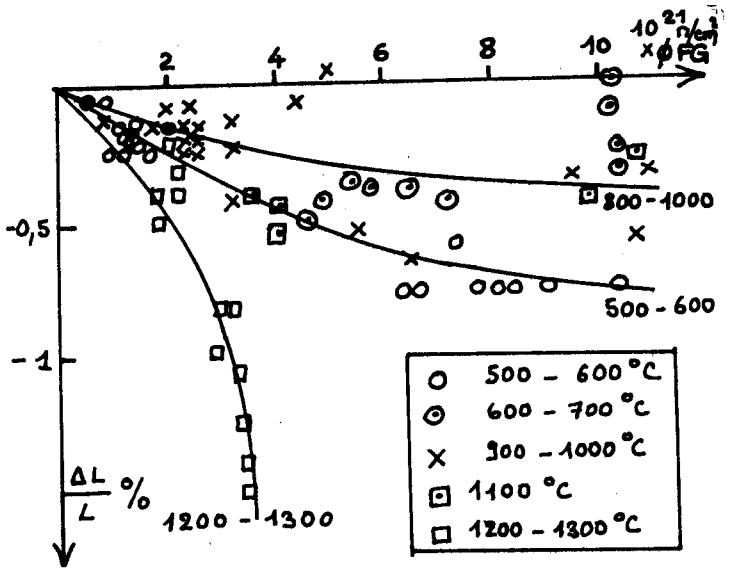

Fig. 5 - Dimensionals changes 7477 - CEA

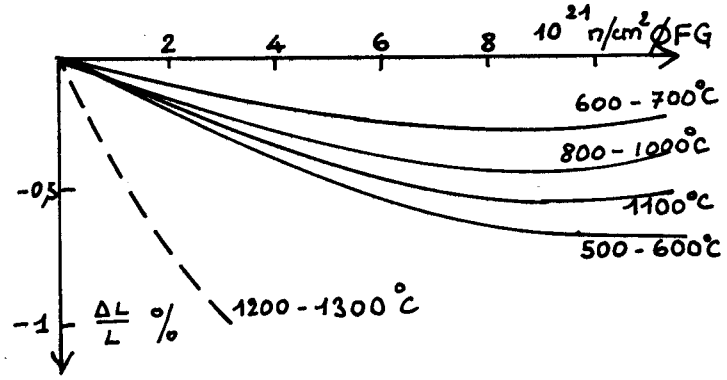

Fig. 6 - Dimensionals changes 7477

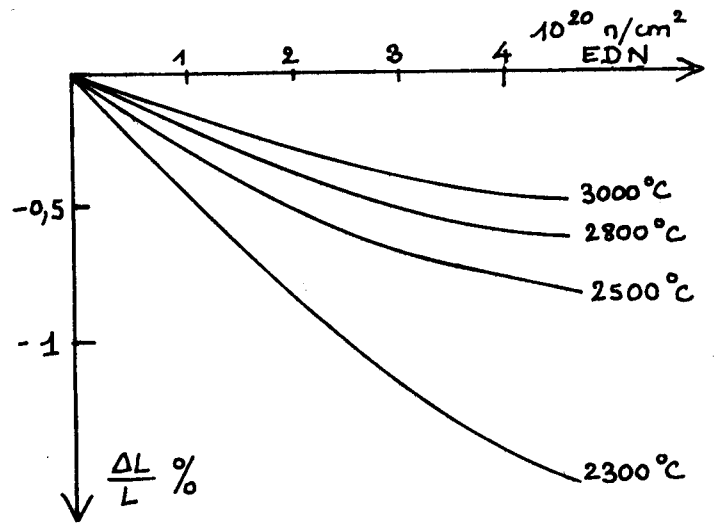

Fig. 7 - Dimensionals changes 2239 f(temperature of graphitization) KFA