ISOTROPIC NUCLEAR GRAPHITES
THE EFFECT OF NEUTRON IRRADIATION

J.LORE, A.BUSCAILLON
Groupe Péchiney Ugine Kuhlmann, Paris (France)

P.MOTTET, G.MICAUD
Commissariat à l'Energie Atomique, CEN Saclay (France)

1. INTRODUCTION

The search for nuclear graphites having a behaviour that is sufficiently stable and isotropic under neutron irradiation has led manufacturers to use isotropic cokes or to carry out isostatic pressing of a fine grain coke/pitch mixture. In the former case there is a difficulty in supplying coke while in the second case the manufacturing cost is rather high.

CORNUAULT et al /1/ have shown that the macroscopic properties of electro-graphites depend mainly on the nature of the coke used, characterized by its apparent dilatability α_a (axial expansion of a cylindrical specimen made under special conditions /2/ /3/ starting with finely crushed coke).

The principal industrial forming processes (extrusion or pressing), intended for the manufacture of graphite from carbonaceous mixtures, can be considered as uniaxial because they tend to orient the grains of coke in the same direction. There exist, however, multiaxial forming processes which cause the grains to be randomly oriented so that no preferred direction is apparent. Among the different multidirectional processes one can number isostatic pressing, extrusion with alternative pressing, and pre-extrusion after multiple crushings of a fine-grained cured product. It is thus possible to prepare macroscopically isotropic graphites from any coke and in particular, from cokes as anisotropic as needle cokes. An additional advantage of these isotropic graphites is that they have low coefficients of thermal expansion (CTE) which lead to lower thermal stresses.

2. EXPERIMENTAL PROCEDURES

Isotropic graphites made from several types of cokes (pitch and petroleum cokes) with an apparent dilatability ranging from 1.20 and 3.60 have been prepared by using multidirectional forming processes. All of these graphites have been fabricated from a mixture composed of typical coal tar pitch (KS = 92°C) and crushed coke. The maximum grain size of the coke is 1.6 mm except for the grade PZ 130 of which the maximum grain size is 0.4 mm. All of these products were cured at 1000°C and have been impregnated once or twice with pitch before graphitization at 2700°C and purification.

3. PHYSICAL AND MECHANICAL PROPERTIES OF THE PRODUCTS

The main characteristics of these products are shown on Table I. One may note that all the graphites are isotropic with respect to thermal expansion. They have lower CTE than graphites prepared from isotropic cokes like gilsocoke (CTE about 5 to 6.10^{-6} $°C^{-1}$). P_3JHAN and P_3JHA$_2$N graphites, prepared from pitch coke by normal extrusion, are less isotropic. Pitch impregnations carried out before graphitization enable one to obtain high densities and mechanical characteristics comparable to those of conventional graphites.

4. BEHAVIOUR UNDER NEUTRON IRRADIATION

The irradiation of these graphites have been carried out in the fast neutron reactor Rapsodie (Cadarache), between 800°C and 1400°C and at fast fluences up to 1.7 10^{22}n.cm^{-2} ϕFG*. In addition to the graphites noted above, two fine grain isotropic graphites (ref. S.1134 and S. 1365) manufactured by Le Carbone Lorraine Co (Table I) have been irradiated.

The dimensional changes as a function of fluence are shown on Figures 1 and 2 and lead to the following remarks :

- the product PZ 130, which is distinguished from PZ 140 by the grain size, exhibits greater anisotropy and larger axial dimensional change.
- the non impregnated products PZ 135, PZ 138 and PZ 141 undergo isotropic changes but expand at lower fluences and at higher rates than singly or doubly impregnated products.
- the graphite PZ 140 based on anisotropic petroleum coke has a more stable behaviour than PZ 143 graphite made from more isotropic pitch coke.
- the products S.1134 and S.1365, in spite of their finer texture, exhibit dimensional changes equivalent to the above graphites. However, their behaviour is not as good as other fine grained graphites /4/. One will note the better dimensional stability of the petroleum coke graphite in spite of its lower density.
- in general, these isotropic graphites are more stable than P_3JHAN and P_3JHA$_2$N except for the non impregnated products.

The CTE of the impregnated graphites do not change greatly in contrast to those of non impregnated graphites which undergo large variations.

The thermal conductivity of the impregnated graphites drops very rapidly as a function of fluence and then saturates at a level varying according to the state of initial graphitization. At fluences greater than 1.2 10^{22}n.cm^{-2} ϕFC, the thermal conductivity undergoes a second degradation which occurs at higher fluences if the graphite has a better stability.

* graphite damage fluence = 1.25 fluence E > 0.18 MeV = 1.85 EDN fluence

5. CONCLUSION

The use of multidirectional forming processes enables one to obtain isotropic graphites from anisotropic cokes. Under neutron irradiation these graphite exhibit isotropic behaviour. Their dimensional stability is **lower** than that of graphites based on isotropic cokes or made by isostatic pressing of fine grained coke/pitch mixtures.

Bibliography

/1/ P.CORNUAULT, F.DUCHAFFAUT, M.COSTE
Classement des cokes suivant leur dilatabilité apparente
Carbon Conf. Baden-Baden
preprints p 371, 1972

/2/ P.CORNUAULT, J.LORE, J.RAPPENEAU, M.YVARS, P.MOTTET
Changes in the properties of Nuclear Graphites obtained from a mixture of cokes
10th Biennial Conf. on Carbon
Bethleem, Penn., 1971

/3/ C.F.STOUT, M.JANES, J.A.BIEHL
Technical Documentary Report
WADD TR 61.72, vol XXXVI, p 18, Oct. 1963

/4/ P.MOTTET, G.MICAUD
Graphite obtenu par pressage isostatique
Comportement sous irradiation neutronique
CEA-N 1903, Sept. 1976

Table I - Physical and mechanical properties of isotropic graphites

Ref. graph.	Coke nature	α_a (1)	Forming Process (2)	Pitch Impreg.	Density g.cm^{-3}	CTE $\bar{\alpha}\,^{525}_{20}\,10^{-6}\,°C^{-1}$ axial	CTE radial	Anisotropy α_R/α_A	Resistivity (μΩ cm) axial	Resistivity radial	Tensile Strength daN. cm^{-2} (3) axial	Tensile Strength radial
P_3JHAN	Pitch coke	3.0	U	1	1.73	3.05	4.21	1.36	900	1185	135	95
P_3JHA_2N	Pitch coke	3.0	U	2	1.79	3.08	4.23	1.35	800	1060	175	110
PZ 130	Regular Petroleum coke	2.15	M	1	1.74	3.00	3.80	1.26	885	1070	205	150
PZ 135	Premium Petr. needle coke	1.20	M	0	1.61	2.40	2.65	1.10	1220	1140	-	95
PZ 138	Regular Petr.	2.15	M	0	1.57	3.20	2.90	0.90	1340	1140	-	100
PZ 140	Regular Petr.	2.15	M	2	1.75	3.25	2.95	0.91	875	780	150	110
PZ 141	Pitch coke	3.6	M	0	1.59	4.15	4.15	1	1650	1520	-	105
PZ 143	Pitch coke	3.6	M	2	1.81	4.90	4.65	0.95	1100	1040	165	150
S.1134	Petr. coke	-	S	1	1.65	3.12	3.12	1	1500	1500	-	-
S.1365	Pitch coke	-	S	1	1.79	5.06	5.28	1.04	1500	1500	-	-

(1) apparent dilatability of coke
(2) U = uniaxial forming process, M = multiaxial forming process, S = special forming process
(3) measured on "diabolo" specimens

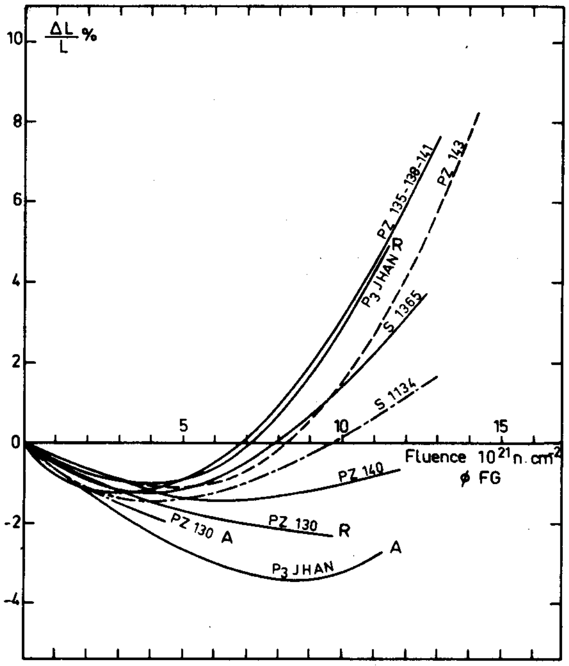

Fig 1. Dimensional changes at 1000°C

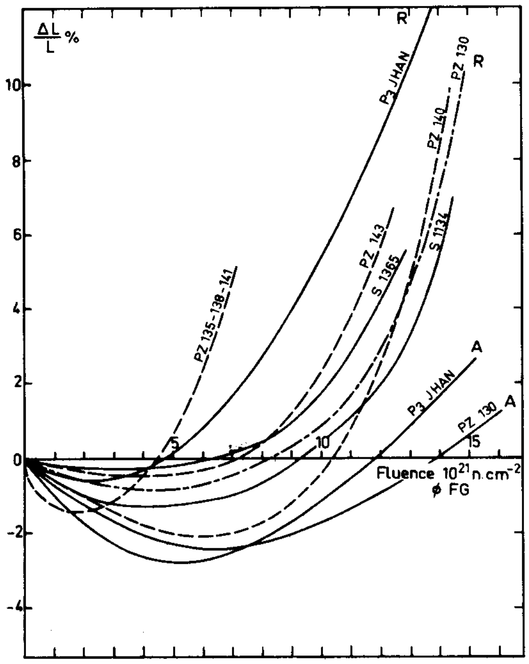

Fig 2. Dimensional changes at 1300°C