

## Preheat Treatment of Carbon Fibers for All Carbon Composites (CFRC)

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### 1. Introduction

It was shown that for CFRC composites highly heat treated carbon fibres type HM without surface treatment are required to achieve the translation of fibre strength up to 100% in the composite compared with only 30% in case of non surface treated HT fibres (1,2,3,4). Surface treated C-fibres behave even worse (see Fig.1) (5). Such fibres with reactive surface cause high-degree of shrinkage. Thus dangerous internal stresses are induced which reduce the final strength of the composite.

Some improvement was reported by post heat treatment of HT fibres up to 1970 K (6) but even then, only 35 % translation of the fibre strength can be calculated from the published data. Therefore, systematic experiments were started to study the effect of heat treatment of HT fibres with increasing temperatures up to 2570 K on the final mechanical properties of CFRC composites.

### 2. Experimental

**2.1 Fibre type.** Commercial carbon fibres type HT (HTA-7-12000 TOHO Besficht) were heat treated in argon at temperatures of 1870, 2270 and 2570 K alternatively for one hour with heating and cooling rates of 400 K/h. The commercial carbon fibre type HM (HM-12 SIGRI) was used for comparison. The original commercial HT fibre and HM fibre were pre-heated some seconds in nitrogen at 1170K to eliminate the commercial finish before preparation of the composite.

**2.2 Composite preparation.** With the original HT and HM fibres and the heat treated HT carbon fibres UD composites with 52 vol% fibre content were prepared by the wet winding technique with phenolic resin formaldehyde type novolak (BAKELITE No.DP 226) as matrix precursor. The carbonization was performed with a heating rate of 10 K/h up to 1270 K in argon atmosphere. The carbonized composites were densified up to 4 times by impregnation with the precursor resin and recarbonization up to 1270 K.

### 3. Results

**3.1 Fibre properties.** The effect of heat treatment of the fibres up to varied final temperatures is compiled in Table 1. The density increases from 1.77 gcm<sup>-3</sup> for the

Table 1. Carbon Fiber Properties

Fiber	Density (g/cm <sup>-3</sup> )	Tensile Strength (MPa)	Young's Modulus (GPa)	Elong. (%)
HT-7-12000	1.770	3075.8	217.9	1.41
1870 K	1.790	2693.9	276.6	0.97
2270 K	1.880	2601.0	343.1	0.76
2570 K	1.900	2778.3	363.1	0.74
HM-12	1.815	2521.5	315.9	0.79

untreated HT fibres to 1.90 gcm<sup>-3</sup> after heat treatment up to 2570 K and is higher than that of the commercial carbon fibres HM type. The same is observed for the YOUNG's modulus which increases from 217.9 GPa to 363.1 GPa again to a higher value of the commercial carbon fibres HM type which was used for comparison. The tensile strength shows a flat minimum after heat treatment up to 2770 K. These changes of density and mechanical properties are in principle in agreement with literature data and explained by the thermally activated reorganization of the fibre structure.

**3.2 Properties of the carbon/carbon composites during processing.** Fig. 2 gives the bulk densities of the C/C composites after first carbonization and after the subsequent impregnation recarbonization steps. It can clearly be seen that the degree of shrinkage expressed as bulk density after first carbonization is extremely high with HT fibre types. The lowest cross sectional shrinkage was found for the commercial HM fibre type. The post-heat treatment of the HT fibres in laboratory scale decreases the amount of shrinkage (see Table 2). The densification by impregnation/recarbonization is in agreement with literature data. But the highest density value was achieved after four densification cycles with the fibre which has been post-heat treated at highest temperatures in the laboratory.

The mechanical properties of all composites were measured after the final (fourth) densification step. Fig. 3 shows the flexural strength and the YOUNG's modulus. In agreement with our previous results, the carbon/carbon composite with HM type fibres has a higher final strength than that prepared from HT fibres. The strength increase is

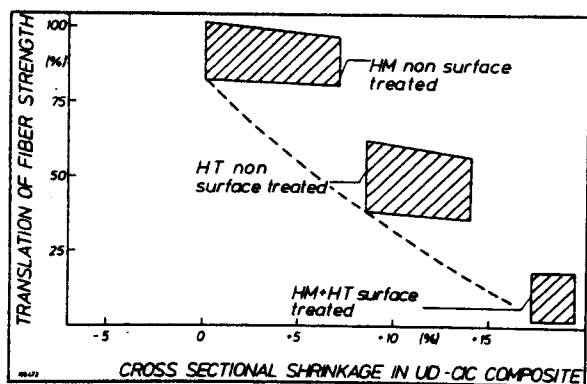


Fig. 1. Cross sectional shrinkage of the composites during the carbonization process reinforced with different fiber types.

Table 2. Cross Sectional Shrinkage of the Composite

HT-7-12000: 14.7%	2270 K: 8.2%	HM-12: 3.0%
1870 K: 13.4%	2570 K: 5.4%	

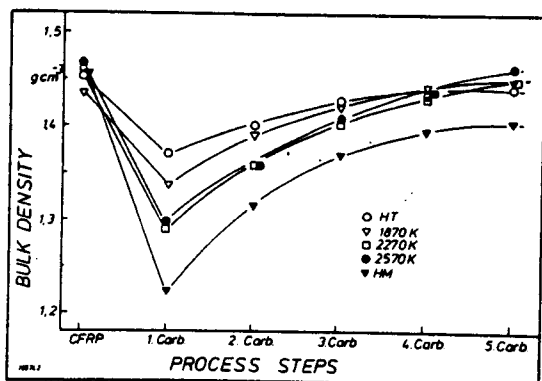


Fig. 2. Densities of the composites after each process step.

more expressed with the post-heat treated HT fibre types and this although the tensile strength of the pure fibres drops by this treatment as shown in Table 1.

Also the translation of fibre strength as shown in Fig. 4 is in agreement with our previous results. Commercial HM fibre type shows a translation which is twice of that of HT fibres. The absolute values of only 55 % translation in case of HM fibres is obviously caused by the precursor. 100 % translation is only found with pitch impregnation cycles. A stronger increase in translation of fibre strength was found with the HT fibres post-heat treated in laboratory scale. Maximum translation of 70 % is achieved by heat treatment around 2300 K.

The translation of the fibre modulus to the composite increases with temperature of post-heat treatment, too. It can be shown clearly however that in case of the used phenolic matrix precursor, the effect of heat treatment on the modulus is not so strong as in

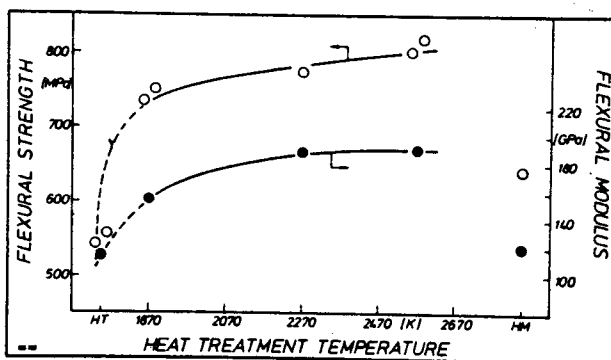


Fig. 3. Flexural strength and flexural modulus of the composites reinforced with different fibers after four densification steps.

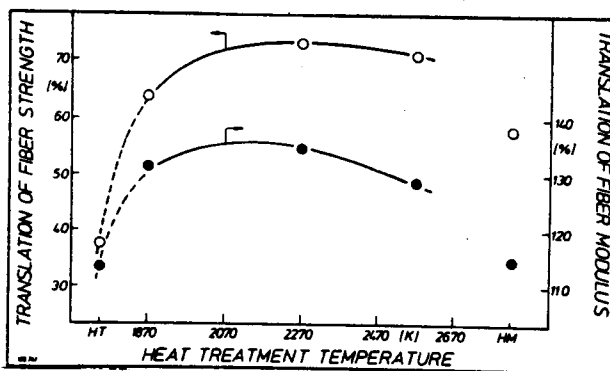


Fig. 4. Translation of fiber strength and fiber modulus in the composite reinforced with different fibers after four densification steps.

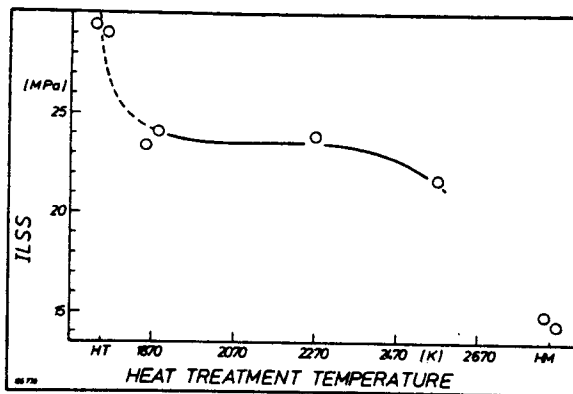


Fig. 5. ILSS of the composites reinforced with different fibers after four densification steps.

case of pitch based matrix carbon. Even with the high modulus fibres the translation of modulus of the composite is quite low as shown previously in literature (4). In case of special pitches which form mesophase and well graphitizing coke the YOUNG's modulus of matrix contributes in the same extent of magnitude to the modulus of composite (2).

Fig. 5 shows the ILSS measured by short beam

test. As known the adhesion between fibre and matrix is best with HT fibres and worse with HM type fibres. This is confirmed again in the medium range of post-heat treated HT fibres where we have an intermediate ILSS value which corresponds very clearly with the maximum in translation of fibre strength and modulus.

#### 4. Discussion

The systematic study of influence of the microstructure of carbon fibre surfaces in preparation of carbon/carbon composites by the liquid impregnation process has confirmed the experiences discussed in literature. Namely the adhesion between matrix precursor and the fibre should be low in order to avoid damages of the fibre during carbonization. In case there is a low adhesion between fibre and matrix, the binder precursor will shrink away from the fibre during first carbonization. In such a way, a formation of circumstantial slits between fibre and first binder coke are found which are ideal for densification steps by impregnation/recarbonization cycles. In case of a good primary adhesion between binder and fibre, the binder coke shrinks on the fibre but causing high degree of internal

stresses and very often also severe damages of the fibre itself. This damage can be seen from the low values of translation of fibre strength in the final composite.

The systematic study here shows however that post-heat treated fibres but not at a maximum temperature as commercial HM fibres are even more suitable for carbon/carbon composites. Additionally lower costs can be expected. For the HT fibres used in these experiments the optimum post-heat treated temperature is in the region between 2070 and 2170 K if phenolic binder precursor is used.

It is assumed but not yet experimentally confirmed, that these conclusions are also valid for multidirectional C/C composites.

#### 5. Literature

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