PREPARATION OF CARBON FIBERS FROM THE ISOLATED ISOTROPIC PHASE OF A THERMALLY TREATED COAL-TAR PITCH

V. Prada, C. Blanco, R. Santamaría, J. Bermejo and R. Menéndez

Instituto Nacional del Carbón, Apdo. 73, 33080-Oviedo (Spain)

Introduction

Carbon fibers are used in a wide range of applications. There are many types of carbon fibers, with different properties and characteristics. Of special interest are carbon fibers with high mechanical properties for structural applications and highly oriented fibers for high temperature applications [1].

Other type of fibers with lower properties are the so called *general purpose carbon fibers* (GPCF). These fibers are mainly used in civil engineering materials (as part of reinforced concrete), in electronic components (battery anodes), or as activated carbons for gas storage [2].

In this work, GPCF were prepared from a new precursor. Continuous carbon fibers were obtained using the isolated isotropic phase of a thermally treated coal-tar pitch.

Experimental

A commercial coal-tar pitch was thermally treated at 430°C for 4 h to obtain a partially anisotropic pitch. The isotropic phase of the treated pitch was isolated using hot filtration [3]. 500 g of pitch was placed in a reactor with a filter pad (5 μm) in the bottom. The sample was heated up to 300°C and nitrogen pressure was applied. The isotropic phase passed through the filter and the mesophase, together with the primary QI remained on the filter.

The isotropic phase was then extracted with different solvents in order to increase its softening point. The extract was spun at 300°C using a spinnerette with 16 holes, each one 300 μm in diameter. A nitrogen pressure of 5 kg cm^{-2} was applied. The fibers were wound at different winding speeds in order to obtain fibers of different diameters.

Green fibers were stabilized using different temperature profiles. The stabilization process was optimized by studying partially stabilized fibers in a differential scanning calorimeter (DSC). Analyses were performed under an air atmosphere, using a heating rate of 10°C min^{-1}.

Stabilized fibers were carbonized in a horizontal furnace. The temperature was raised to 1000°C at a rate of 1°C min^{-1} under a nitrogen atmosphere.

The carbon yield of green and stabilized fibers was determined in a thermobalance. Experiments were performed up to 1000°C in a nitrogen atmosphere. Similar analyses under an air atmosphere were carried out to study the reactivity of carbonized fibers.

The oxygen content of the green, stabilized and carbonized fibers was determined by elemental analysis in order to evaluate the oxygen uptake during stabilization.

Scanning electron microscopy was used to study the texture of the fibers. Finally, the mechanical properties of the carbonized fibers were determined by standard tensile strength tests.

Results and discussion

Pitch thermal treatment yielded a pitch with a 40 vol. % mesophase content. The isotropic phase of this pitch was successfully isolated by hot filtration.

The Mettler softening point of the isotropic phase (180°C) was too low to spin and to stabilize the fibers. Therefore, several solvents of different extraction ability (toluene, acetone and acetonitrile) were tested in order to extract the light components of the sample. A mix of acetonitrile/acetone (70:30) was found to be the best solvent. In this way, an isotropic pitch with a softening point of 225°C was obtained.

The spinning was performed at 300°C with 5 kg cm^{-2} of nitrogen pressure, using 400 and 1000 rpm of winding speed. In the first case fibers with an average diameter of 35 μm were obtained and in the second case the average diameter was 16 μm.

Both types of fibers were stabilized at 1°C min^{-1} of heating rate with the following holding temperatures and residence times: 125°C/ 2 h, 135°C/ 2 h, 145°C/ 1 h, 155°C/ 1 h, 175°C/ 1 h, 200°C/ 1 h and 250°C/ 1 h.

The stabilization program was fixed according to the softening point of the pitch, the starting temperature being low enough to avoid sticking between fibers. DSC analysis (Figure 1) showed that in both types of fibers stabilization was successfully completed with this program. The exothermic peak was first moved to higher temperatures, and disappeared for the completely stabilized fibers.

The stabilized fibers showed a carbon yield higher than 80 wt. % in all cases, while the carbon yield

of the green fibers was only 45 wt. %. DTG curves of the carbonized fibers under air showed a different behavior for the fibers with a different diameter. The 16 μm fibers showed the maximum weight loss rate at 625°C. The 35 μm fibers DTG curve had two peaks, one at 680°C and the other at 750°C (Figure 2). This behavior could have been due to the incomplete or non-homogeneous stabilization of the thicker fibers, as they might have been too thick for the oxygen to diffuse, and the core could be not stabilized. Both types of stabilized fibers reacted much earlier than the green fibers. This could be due to the presence of oxygen functional groups, which start reacting earlier.

Elemental analysis results are shown in Table 1. The oxygen content increases from 0.98 % to 6.81 % during stabilization, an indication of a good stabilization process. Part of the oxygen is lost during carbonization.

The mechanical properties of the fibers are also dependent on the fiber diameter. As might be expected, 16 μm carbon fibers had a higher tensile strength (414 MPa *vs* 143 MPa), and Young Modulus (36.3 GPa *vs* 28.9 GPa). The probability of defects in the fibers decreases with decreasing fiber diameter, resulting in better mechanical properties. However, these properties are not very high in any case.

Conclusions

The isolated isotropic phase of a thermally treated coal-tar pitch with a suitable softening point is a good precursor of continuous general purpose carbon fibers.

The spinning of this precursor is relatively easy, although the fibers are difficult to stabilize and the stabilization process is quite long (13 h).

Complete stabilization of fibers was only achieved for diameters in the range of 16 μm.

Resultant carbon fibers showed low mechanical properties but a surface without any defect, which makes them suitable for applications in which structural properties are not important.

References

1.- Chung D.L. Carbon fiber composites. Newton (US): Butterworth-Heinemann. 1994: 3-11.

2.- Alcañiz-Monge J., Cazorla-Amorós D., Linares-Solano A., Oya A., Sakamoto A., Hoshi K. Carbon 1997; 35: 1079-1087.

3.- Blanco C., Santamaría R., Bermejo J., Menéndez R. Carbon 1997; 35: 1191-1193.

Acknowledgements

Authors would like to thank CICYT (Project MAT97-0677) for financial support. V. Prada and C. Blanco are grateful to MEC for their research grants.

Table 1.- Main properties of the 35 μm fibers.

	Green	*Stabilized*	*Carbonized*
O	0.98	6.81	2.69
S	...	...	143
M	...	...	28.9

O, oxygen (wt. %)
S, Tensile strength (MPa)
M, Young Modulus (GPa)

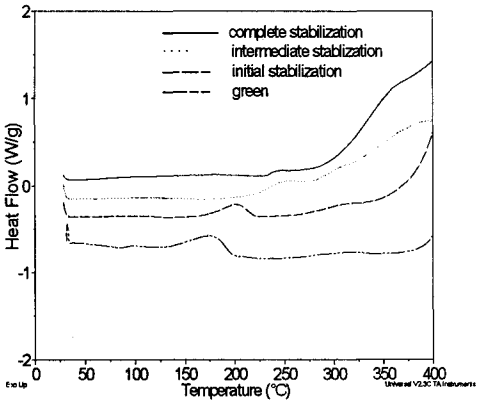

Figure 1.- DSC analyses of fibers.

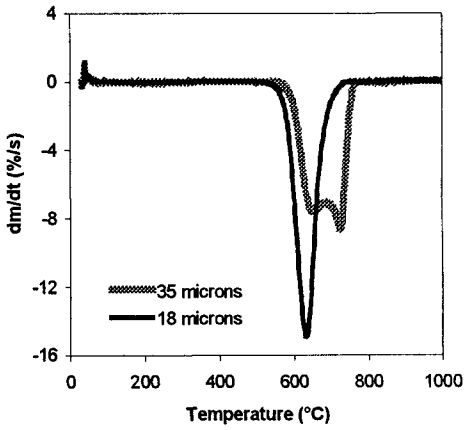

Figure 2.- DTG analysis of carbonized fibers.