# MESOPHASE DEVELOPMENT IN AIR-BLOWN PITCHES. COMPARISON WITH THERMALLY TREATED PITCHES AND THEIR ISOLATED PHASES

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## Introduction

Coal-tar pitches are used as matrix precursors of a wide range of carbon materials. Pitches are used as binders of different fillers (i.e. in electrodes and carbon brushes) and also to densify carbon fiber preforms (i.e. in carbon/carbon composites). The transformation of a pitch into a carbon matrix requires thermal treatment (carbonization). The properties of the final material depend both on the chemical composition of the parent pitch and the experimental conditions in which carbonization is carried out.

During pitch carbonization mesophase is developed. This stage is essential for the final properties of the pitch-based materials.

In this work, a comparative study of mesophase development in pitches from different origins is presented. The set of pitches studied includes a commercial impregnating coal-tar pitch, isotropic pitches (air-blown pitches and the isotropic fractions of thermally treated coal-tar pitches isolated by pitch filtration) and partially anisotropic pitches (obtained by thermal treatment). A comparison of their different behavior in the first stages of carbonization is established and the results explained from the different chemical composition of the pitches.

## Experimental

A commercial impregnating coal-tar pitch (BI5) supplied by Química del Nalón was air-blown and thermally treated to obtain pitches of different nature. Pitch air-blowing was carried out at 275°C under air for times varying between 10 and 30 h [1]. The resultant pitches were labeled AB10, AB18, AB25 and AB30, respectively. Pitch thermal treatment was carried out at 430°C under nitrogen for 2, 3 and 4 h [2]. The pitches obtained were labeled C1, C2 and C3, respectively. These thermally treated pitches were then subjected to filtration to isolate their isotropic phases, which were labeled I1, I2 and I3, respectively [3]. All pitches and corresponding isotropic fractions were characterized. Their main properties are summarized in Table 1.

Mesophase development in the different samples was studied from the pyrolysis products obtained after treatment at 430°C under nitrogen for times varying between 1 and 4 h. 8 g of each sample was placed in a Pyrex tube, and then introduced in a stainless steel reactor heated with an electrical furnace. 6 test tubes were used per batch. After completing the treatment, the reactor was quickly removed from the furnace, in order to quench the reaction. The yield of the pyrolysis process was calculated from the weight of the carbonaceous residues obtained. Each tube was then embedded in an epoxi resin and cut vertically into two pieces, one of which was polished for optical microscopy examination. A point counting procedure was used to evaluate the mesophase content of the samples.

## Results and discussion

The set of pitches studied showed a different degree of polymerization, as a result of the different treatments applied (air-blowing and thermal treatment). The polymerization degree of these pitches is reflected by the data summarized in Table 1.

Figure 1 shows the pyrolysis yield obtained for the different pitches. The lowest yield was that corresponding to the parent pitch, as might be expected. The pyrolysis yields obtained for the air-blown pitches are significantly lower than those of the thermally treated pitches. Especially striking are the close similarities between the yields of each thermally treated pitch (Ci) and its corresponding isotropic fraction (Ii).

The behavior of each pitch during pyrolysis was studied from the mesophase development during the treatments. The results obtained showed interesting differences between them. The air-blown pitches developed mesophase at a similar rate, independently of their degree of treatment. This was also the case with the isotropic fractions, where only I2 deviates from the general trend. The parent pitch developed mesophase at a similar rate to the isotropic pitches. As with the thermally treated pitches, C1 and C2 behaved similarly to the isotropic fractions. In the case of C3, with a higher mesophase content, the rate of mesophase development was slower than for the other two pitches.

Also noticeable was the fact that the air-blown pitches developed mesophase at a higher rate than the other pitches. Differences between the behavior of the different sets of pitches were found not only in the rate of mesophase formation but also in its morphology. While mesophase spheres decanted in the bottom of the reactor for the thermally treated pitches and their isotropic phases, air-blown pitches showed a homogeneous distribution of spheres throughout the sample. This indicates that the nature of the pitch strongly affects the density and viscosity of the co-existing phases.

## Acknowledgements

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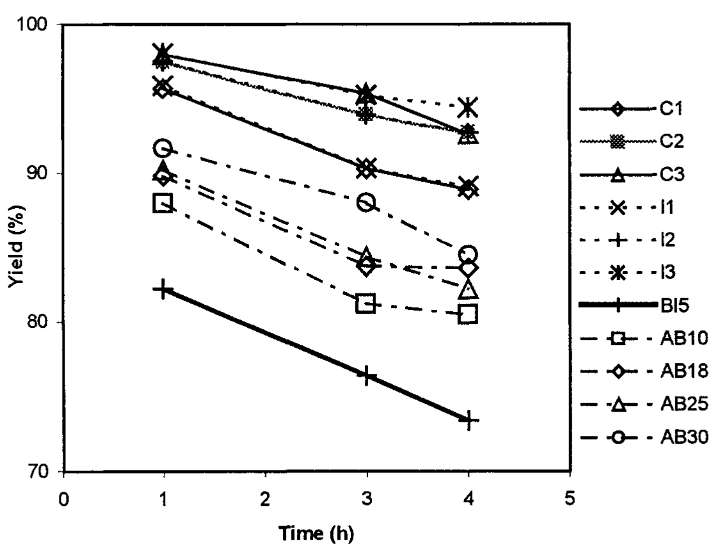


Figure 1.- Pyrolysis yield for the different pitches.

## References

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2. Blanco C., Santamaría R., Bermejo J., Menéndez R., Fleurot O., Edie D."Contribution of the isotropic phase to the rheological behavior of partially anisotropic coal-tar pitches" Carbon, in press.
3. Blanco C., Santamaría R., Bermejo J., Menéndez R. Carbon 1997; 35: 1191-1193.

Table 1.- Main properties of pitches.

| Pitch | SP | CY | TI | MC |
|---|---|---|---|---|
| Parent | 97 | 34.6 | 20.0 | 0 |
| AB10 | 139 | 48.0 | 36.6 | 0 |
| AB18 | 168 | 57.6 | 44.6 | 0 |
| AB25 | 197 | 61.8 | 51.8 | 0 |
| AB30 | 210 | 62.7 | 52.0 | 0 |
| C1 | 149 | 54.0 | 43.9 | 10 |
| C2 | 174 | 61.5 | 53.9 | 25 |
| C3 | 190 | 65.6 | 57.5 | 37 |
| I1 | 151 | 51.6 | 40.3 | 0 |
| I2 | 169 | 56.2 | 45.8 | 0 |
| I3 | 178 | 58.9 | 48.1 | 0 |

SP, Mettler softening point (°C)
CY, carbon yield (wt. %)
TI, toluene insoluble content (wt. %)
MC, mesophase content (vol. %)

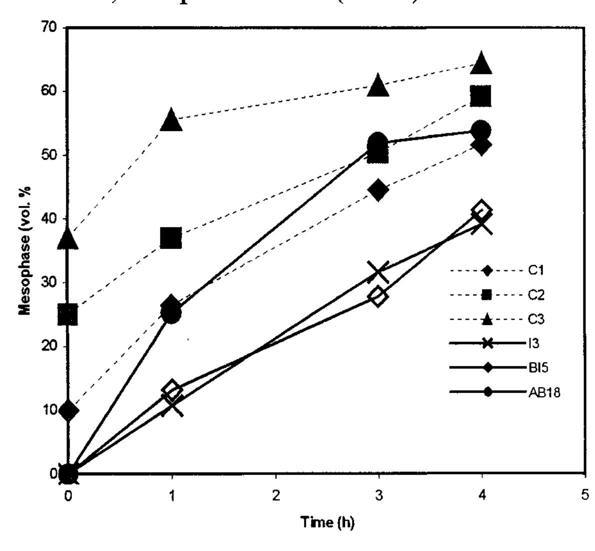


Figure 2.- Mesophase content of pitch pyrolysis products.