

Effects of the Mesostructural Order on the Electrochemical Performance of Hierarchical Micro-Mesoporous Carbons

M. Enterría,^{*a} A. Castro-Muñiz,^b F. Suárez-García,^a A. Martínez-Alonso,^a J.M.D. Tascón^a and T. Kyotani^b.

Supplementary information

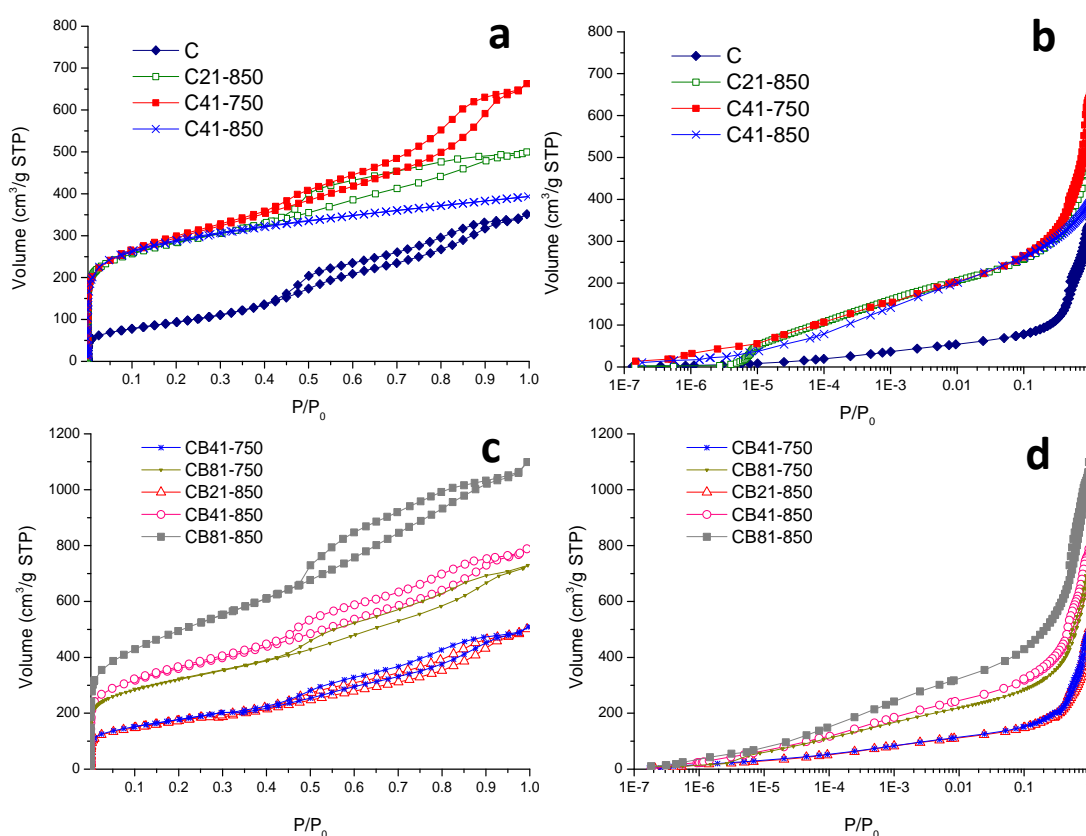


Fig. 1S. N₂ adsorption-desorption isotherms at -196 °C on a) carbons from the C series and c) carbons from the CB series and the corresponding semi-logarithmic scale isotherms b) and d).

N₂ adsorption-desorption isotherms corresponding to the carbons from the C and CB series are shown in Fig. 1S. CMK-3 type carbon (C sample) gives a type IV isotherm, typical of mesoporous materials. All the activated carbons, excepting C41-850, exhibit type IV isotherms (in the medium and high P/P₀ ranges) with some contribution of type I (in the low

^a Instituto Nacional del Carbón, INCAR-CSIC, Apartado 73, 33080 Oviedo, Spain. *E-mail: marina@incar.csic.es

^b Institute of Multidisciplinary Research for Advanced Materials, Tohoku University, Aoba-ku, Sendai 980-8577, Japan.

P/P_0 range). The N_2 uptake at $P/P_0 < 0.1$ increases with increasing KOH proportion and activation temperature. For C series a 3-fold increase (C41-750) of the adsorbed volume at $P/P_0 < 0.1$ with respect to C sample is produced. In the case of CB series even a 5-fold increase is achieved (CB81-850). On the other hand, for CB series, both isotherm shape and slope at medium and high P/P_0 remain almost unchanged (compare with pristine carbon, C). This is not the case for C series. Besides, sample C41-850 present a type I isotherm indicating the destruction of the ordered mesopore system. To sum up, the mesopore size distribution is almost maintained after the activation process for the CB series but not for the C series.^{1,2}

Fig. 1S b and d display the same high-resolution N_2 isotherms (measured in the P/P_0 range of $10^{-7} - 0.99$) in semilogarithmic scale. In this type of plot, the increase of the microporosity with increasing both KOH proportion and activation temperature can be easily appreciated.

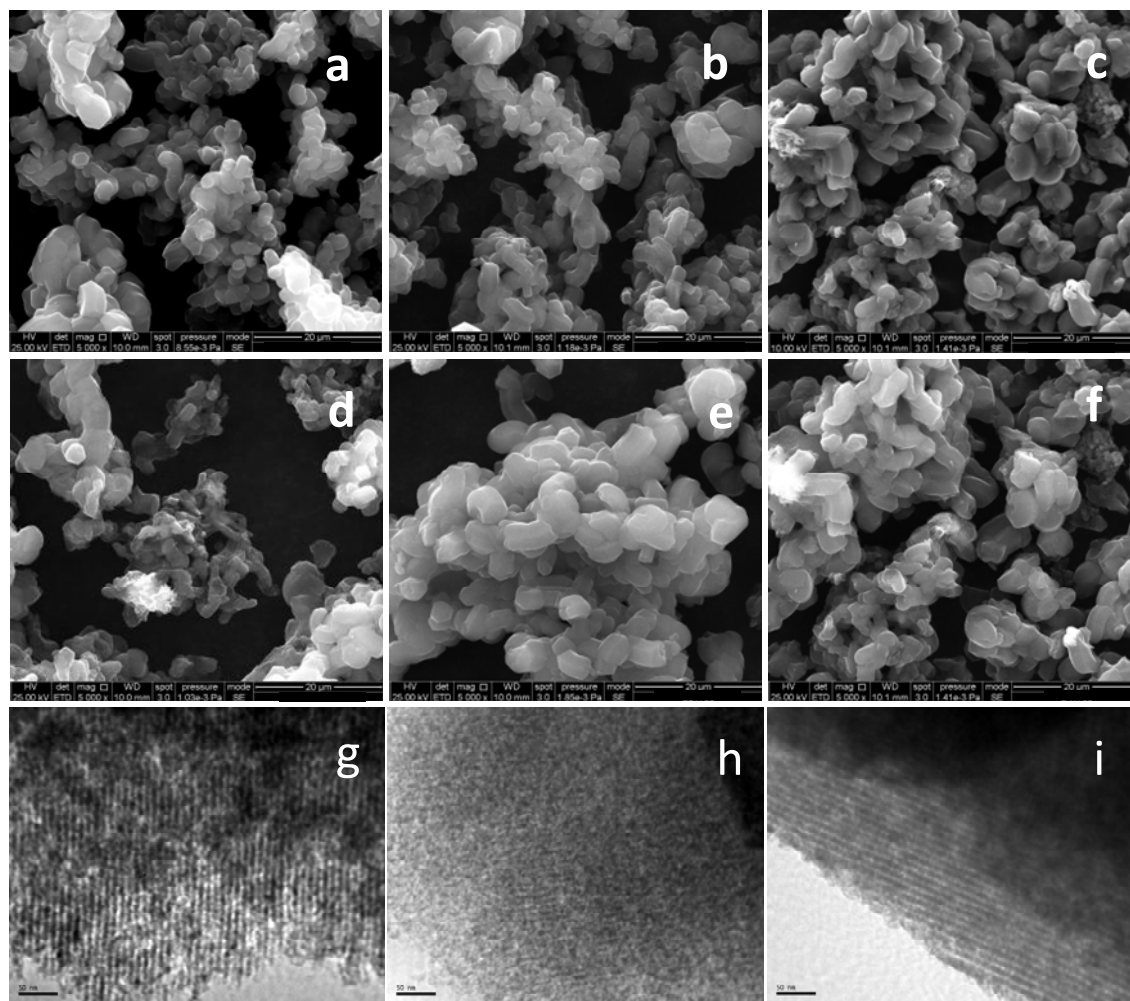


Fig. 2S. SEM images of samples: a) C, b) C21-850, c) C41-750, d) C41-850, e) CB41-850 and f) CB81-750 and TEM images of samples: g) C41-750, h) C41-850 and i) CB41-850.

Scanning electron microscopy (SEM) and transmission electron microscopy (TEM) images of some representative samples from both series (C and CB) are displayed in Fig. 2S. CMK-3 type carbon, C sample (Fig. 2S a) consist of smooth and hexagonal particles around 4-5 μm in size. This morphology is typical of this kind of material and reproduces the particle morphology of the SBA-15 template. The activated carbons (Fig. 2S b-f), in general, maintain this morphology but some particles without a well-defined morphology can be observed, especially for the samples from the C series. This is due to the loss of the mesostructural order during the activation process.

The same phenomena can be observed in the TEM images (Fig. 2S g-i). Thus, the micrograph of CB41-850 sample (fig. 2S i) displays well-defined carbon bars with a homogenous spacing between them, which is typical of a CMK-3 carbon. On the other hand, C41-750 sample (Fig. 2S g) presents carbons bars with irregular edges and a more deteriorated structure. Finally, C41-850 (Fig. 2S h) shows a compact structure where some faint ordered domains can still be appreciated, but the well-defined space between the carbon bars is lost. This evolution can be explained by the progressive collapse of the CMK-3 structure as the activation conditions become progressively harder.

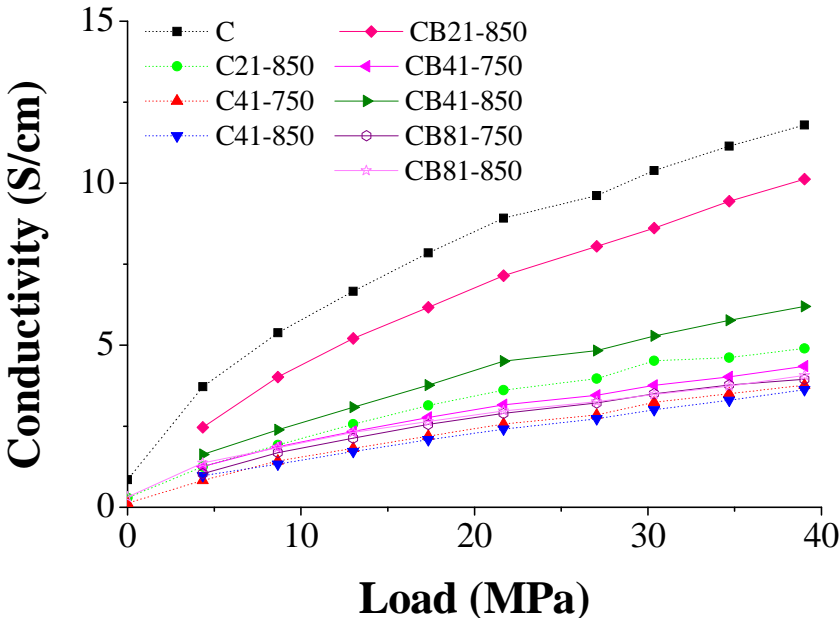


Fig. 3S. Electrical conductivity as a function of loading pressure for the C and CB sample series.

The electrical conductivity of the samples was measured using a mold to press the powdered samples in the absence of either a binder or a conducting additive as it was used for the

preparation of the electrodes for the electrochemical measurements. Fig. 3S plots the electrical conductivity of the samples as a function of the loading pressure. As usual, the conductivity increases with increasing load, yielding values for the conductivity at the highest load used in this study (40 MPa) in the range 3.5-11.8 S/cm. These values are similar to the ones obtained in other studies for carbon materials.^{3,4}

References

1. M. Enterría, F. Suárez-García, A. Martínez-Alonso and J. M. D. Tascón, *Microporous Mesoporous Mat.*, 2012, **151**, 390-396.
2. M. Enterría, F. Suárez-García, A. Martínez-Alonso and J. M. D. Tascón, *Carbon*, 2012, **50**, 3826-3835.
3. A. Celzard, J. F. Marêché, F. Payot and G. Furdin, *Carbon*, 2002, **40**, 2801-2815.
4. B. Marinho, M. Ghislandi, E. Tkalya, C. E. Koning and G. de With, *Powder Technology*, 2012, **221**, 351-358.