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TORTUOSITY OF THE POROUS STRUCTURE OF CARBON GELS

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Introduction

Carbon gels are porous materials obtained by carbonization of the organic polymers usually obtained by sol-gel polymerization reaction of hydroxylated benzene and aldehyde. The final three-dimensional polymeric network can be controlled by the polymerization process. Thus, usually varying the type and amount of solvent and/or reactants and the pH of the precursor solution, carbon gels with different porosity may be obtained. As a consequence, the potential applications of these materials are enormous as their porosity may be designed as requested. However, to the best of our knowledge these porous characteristics are usually just defined as a mean pore size and pore volume. In this work, it is shown how a series of samples with analogous average pore size may present notably differences in their polymeric structure and thus in their further behavior in different applications.

Materials and Methods

Resorcinol-formaldehyde gels were obtained from a sol-gel process performed under microwave heating[1]. The solvent used was water and the synthesis (i.e. polymerization, curing and drying steps) were performed in a one-pot process lasting just 5 hours. The control of the porous structure of these RF gels is possible by varying synthesis variables such as R/F ratio, dilution ratio D, amount of methanol in the F used, pH of the precursor solution, etc.[2-4] However, different combination of these variables may be performed to obtain similar mean pore size in the final RF gel. In this work, a series of samples with three different pore size (i.e. 30, 100 and 300 nm) were obtained by different combination of the synthesis variables. Table 1 shows the different synthesis conditions used for each sample. The RF gels were carbonized at 1000ºC under N_2 flow, and degasified under vacuum and 120ºC overnight prior to characterization. Porous properties were evaluated by mercury porosimetry (volume of meso- and macropores, mean pore size, apparent density and % of porosity) and N_2 adsorption-desorption isotherms at -196ºC (BET surface area). Parameters such as tortuosity and permeability were also obtained from the Hg intrusion profile. The carbonaceous structure was also characterized by SEM.

Carbon gels were denoted as C followed by the mean pore size and "A" or "B" depending on the synthesis conditions used and shown in Table 1.

Results and Discussion

According to previous works, decreasing the pH of the precursor solution produces RF gels with wider pores, and decreasing the dilution of the initial mixture would lead to narrower pores[2,4]. Therefore a proper combination of these two variables would lead to materials with similar pore size (i.e. samples C-300-A and C-300-B). Similar strategy may be performed by varying the pH and the amount of methanol in the precursor solution, as the higher amount of the latter may lead

to RF gels with narrower pores[3] (see samples with mean pore size of 30 and 100 nm in Table 1). Although samples with analogous pore size, microporosity and thus surface area and total pore volume may be obtained, the reality is that their porous structure is very different, as can be observed in Figure 1. Variations in tortuosity may be also performed, and thus controlled, with the chemical variables during the synthesis. This would imply different permeability in the samples, although their typical porous characteristics are similar. It seems that higher amount of methanol or a lower amount of water in the precursor solution produces fused polymeric structure with straight channels that confers to the material a less tortuous porosity. These properties would be decisive for the effectiveness of these materials in filtration or separation processes.

Table 1. Differences in the synthesis conditions used and porous properties of the carbon xerogels studied

Sample	pH/D/MeOH	S_{BET} (m^2/g)	Vp (cm^3/g)	Permeability (mdarcy)
C-30-A	5.1/4.6/10	717	1.1	45
C-30-B	4.5/4.6/13	646	1.0	59
C-100-A	4.0/5.6/10	552	1.4	17
C-100-B	3.5/5.6/13	601	1.6	44
C-300-A	5.0/7.7/10	566	2.6	198
C-300-B	3.3/5.6/10	589	2.0	432

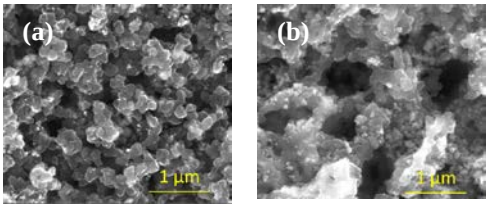

Figure 1. SEM images of some of the samples studied (a) C-300-A (b) C-300-B

Conclusions

Different combinations of synthesis variables of RF gels (mainly amount of water, methanol and pH of the precursor solution) may lead to analogous mean pore size and pore volume in the final carbon gels. However, although the materials show these similar characteristics, their behaviour may be very different for certain applications. It seems that the connectivity of the polymeric clusters and thus the pore tortuosity is also dependent on the chemical variables used during the polymerization. An increase in the methanol content or a decrease in the water amount used during polymerization process seems to produce straight porous channels and thus less tortuous pore structures. This allows us to attain an even finer tune of the porosity of the RF gels either to reduce the pressure drop or to increase the residence time of the solutes in separation processes.

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