

SOL-GEL DERIVED ACTIVATED CARBON AS A POTENTIAL GAS PHASE ADSORBENT

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Introduction

Activated carbon has been widely used as an adsorbent in the separation and purification of gas mixtures in the chemical and petrochemical industries [1]. Recently Pekala and co-workers [2, 3] developed a synthetic carbon aerogel, which was derived from a carbonized resorcinol-formaldehyde (RF) resin via a sol-gel route. They reported very high surface areas (SAs) per unit volume (to 700 m²/cm³), and that the porosity and SA can be controlled over a broad range, while the pore size can be tailored at the nanometer scale. These unique and controllable properties were attributed to the sol-gel process, which is quickly becoming one of the most promising material synthesis techniques.

The sol-gel process readily allows control of the texture, composition, homogeneity, and structural properties of the resulting materials [4]. Numerous studies have been devoted recently to understanding how the synthesis conditions affect the porous structure of sol-gel derived materials [5-9]. The objectives of this work include developing a new sol-gel synthesis route to make carbon xerogels, and studying the effect of different carbonization temperatures and activation conditions on the pore structure and properties of the resulting materials.

Experimental

Reagent-grade resorcinol (98%, Aldrich), formaldehyde (37% in water, Aldrich), sodium carbonate (GR, EM Science, Germany), and nitric acid (HNO₃, 70.4%, Mallinckrodt) were used as received. A solution containing 5 w/v% solids was prepared, in which the R/F mole ratio was fixed at 1:2. Sodium carbonate was used as the catalyst, and the R/C (resorcinol/sodium carbonate) mole ratio was fixed at 50:50. The initial pH 6 of the solution was adjusted with dilute HNO₃. The solution was sealed in a flask and magnetically stirred for 30 minutes, followed by one week of curing in an oven at 85 ± 3 °C (without stirring). Typically, gelation occurred in several hours. After curing, the gel was removed from the oven and cooled to room temperature. It was then washed and vacuum filtered three times with acetone over a period of three days. Fresh solvent was replaced daily after vacuum filtration. The washed gel was dried under N₂ in a

tube-furnace. Using a heating rate of 0.5 °C/min, it was heated to 65 °C and held there for 5 hours; it was then heated to 110 °C and held there for another 5 hours. Finally, the carbon xerogel was formed by pyrolysis of the dried gel at the desired temperature for 3 hours in a N₂ atmosphere with both heating and cooling rates set at 5 °C/min. Activation of the carbon xerogel was carried out with 5% CO₂ during pyrolysis in nitrogen at 1050 °C.

Results and Discussion

It was shown previously [9] that the initial pH of the RF solution had a significant effect on both the SA and pore volume (PV) of carbon xerogels synthesized using the modified procedure. These results are displayed in Figure 1. No SA or PV existed for the carbon xerogels synthesized using an initial pH between 7.0 and 7.5. As the pH decreased, however, both the SA and PV increased substantially, indicating that more pores and SA were created in the RF polymer that remained intact in the carbon xerogel. The pH window for making high SA carbon xerogels ranged between 5.5 and 6.5, where a pH < 5.5 always yielded a precipitate instead of a gel. In this study, an initial pH of 6.0 was used to make the RF resin.

Figure 2 shows the effect of the carbonization temperature on the SA and PV of the carbon xerogels. The general trend was that the SA and PV decreased with increasing temperature. As the temperature increased from 600 to 900 °C, the SA slowly decreased, while the PV decreased sharply. This result indicated the loss of larger pores, which contribute more to the PV than the SA. In contrast, as the temperature increased from 900 to 1200 °C, the SA decreased sharply, but the PV only slowly decreased. This result indicated the loss of smaller pores, which contribute more to the SA than the PV.

Figures 2 and 3 show the effect of CO₂ activation on the carbon xerogels. Interestingly, linear relationships existed between the activation time and both the SA and PV. Moreover, for three hours of activation, the SA more than tripled and the PV more than doubled, compared to without activation. These very large SAs and PVs may give rise to some unusually but highly desirable gas phase adsorbent properties.

The effect of activation time on the weight loss of the carbon xerogel also exhibited a linear relationship, as

shown in Figure 3. This linear relationship was indicative of a uniform, first order reaction process taking place. The total weight loss was about 47% for the carbon without activation (i.e., just due to carbonization), while that activated for 3 hours lost 78 % of its total weight. Clearly, the higher SAs and PVs were obtained at the expense of losing a significant amount of carbon material during the activation process.

Acknowledgments

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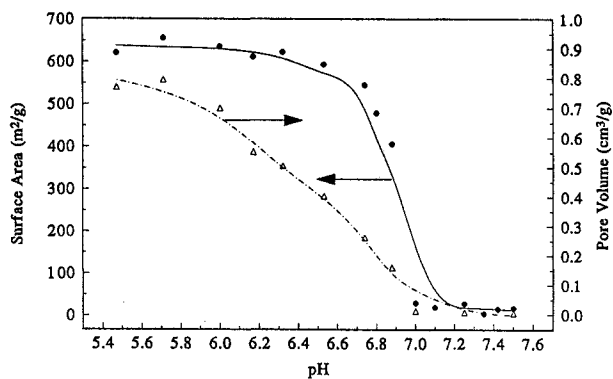


Figure 1. Effect of the initial solution pH on the SA and PV of carbon xerogels carbonized in nitrogen at 1050 °C.

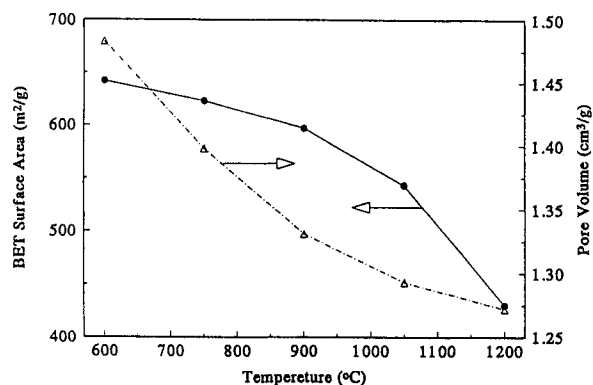


Figure 2. Effect of the carbonization temperature on the SA and PV of carbon xerogels synthesized with an initial pH of 6.

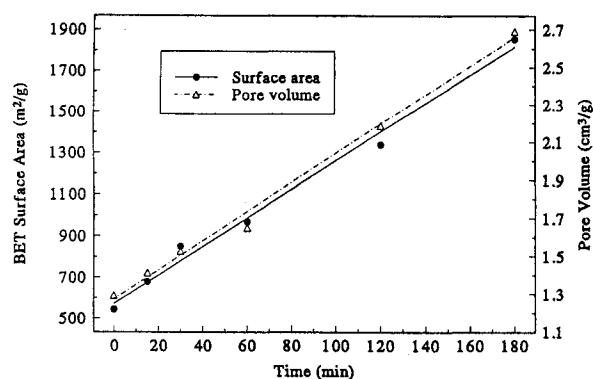


Figure 3. Effect of the activation time on the SA and PV of carbon xerogels synthesized using an initial pH of 6, and activated with 5% CO₂ in nitrogen at 1050 °C.

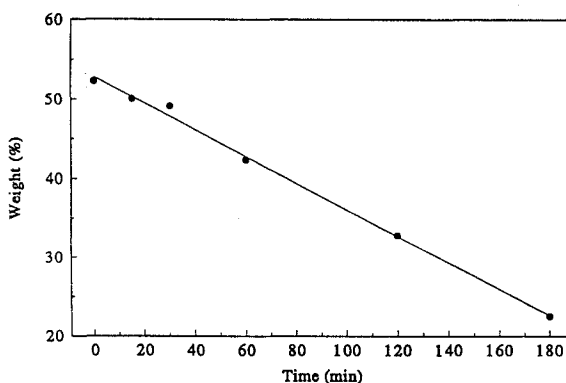


Figure 4. Effect of the activation time on the weight loss of carbon xerogels synthesized using an initial pH of 6, and activated with 5% CO₂ in nitrogen at 1050 °C.