

MODIFICATION OF THE PORE STRUCTURE OF CARBON AEROGELS AND XEROGELS FOR ENERGY STORAGE

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Introduction

Pekala [1] introduced organic aerogels derived from the polycondensation of resorcinol with formaldehyde through a reaction mechanism that is similar to the sol-gel process. The resulting organic gels were dried supercritically in CO₂ to produce organic aerogels. Since this initial work, numerous articles have been published on the properties and uses of these novel organic aerogel materials, with one major application being energy storage. The organic gels have also been dried conventionally to produce organic xerogels, and both types of gels have been carbonized in an inert atmosphere to produce carbon aerogels and xerogels. Most of these references have been cited in [2]; however, none of them have directly compared the physical and chemical properties of carbon aerogels with those of carbon xerogels. Therefore, the objective of this study is to illuminate the differences between carbon aerogels and xerogels using a statistical design approach. Four factors were investigated in a 2⁴ full factorial design: initial solution pH, weight percent solids, pyrolysis temperature and gel type. The response variables were surface area, pore volume and electrochemical capacitance.

Experimental

Resorcinol (ACS, 99+%, Alfa Aesar), formaldehyde (37% in water, Aldrich), sodium carbonate (anhydrous, ACS, Fisher), and acetone (optima, 99.6%, Fisher) were used as received. The synthesis procedure for these gels was developed based on the procedure for making carbon aerogels [1] and xerogels [2]. The solutions contained either 5 or 20 w/v% solids, in which the R/F (resorcinol/formaldehyde) mole ratio was fixed at 1:2. Sodium carbonate was used as the catalyst, where the R/C (resorcinol/sodium carbonate) mole ratio was fixed at 50:1. Four, 1 L batches of gel were made in a glove box under a nitrogen atmosphere: one with a low pH and low wt% solids, one with a high pH and low wt% solids, one with a low pH and high wt% solids, and one with a high pH and high wt% solids. These solutions were poured into 100 ml glass containers and tightly sealed with caps. They were removed from the glove box and placed in an oven at 87 °C for one week to gel and cure. Each cured batch was

divided into two parts. The first part was designated as xerogel and dried accordingly [2]. The second part was designated as aerogel and dried accordingly [1]. The dried xerogel and aerogel resins were again separated into two parts: one pyrolyzed at 800 °C and the other at 1050 °C in a tube furnace under 1 L/min nitrogen flow. In this way, the initial four batches of gel created 16 different carbonized materials, i.e., 8 aerogels and 8 xerogels.

A Micromeritics Pulse Chemisorb 2700 Analyzer was used to obtain the surface areas and pore volumes of both the carbon aerogel and xerogels. An EG&G Potentiostat Model 273A was used to run constant current (1 mA) charge and discharge tests on all 16 samples. A small sample pellet was made by combining the carbonized gel powder with a Teflon binder in a 20:1 mass ratio. The pellet was regenerated at 150 °C for 1 hr and then soaked in 6 M KOH overnight before being tested.

Results and Discussion

Table 1 displays the results from the 2⁴ full factorial design study. The surface area ranged from 460 to 929 m²/g, the pore volume ranged from 0.20 to 1.42 cm³/g, and the capacitance ranged from 59 to 179 F/g (single electrode). Clearly, significantly different pore structures resulted for all of these materials that affected the electrochemical capacitance. Moreover, the results in Table 1 reveal significantly different pore structures for the carbon aerogels compared to the carbon xerogels. Figure 1 shows the correlation of the electrochemical capacitance with the surface areas and the pore volumes of all these samples. The electrochemical capacitance increased with an increase in both these variables in a correlative manner.

References

1. Pekala RW. *J. Mater. Sci.* 1989, 24: 3221.
2. Lin C and Ritter JA. *Carbon.* 1997, 35:1271.

Acknowledgements

This material is based upon work supported in part by the U. S. ARO under Grant No. DAAH04-96-0421 and in part by the U. S. DOE under Cooperative Agreement No. DE-FC02-91ER75666.

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Table 1. 2⁴ Factorial Design Results

Initial Solution pH	Weight % Solids	Pyrolysis Temperature °C	Gel Type	Surface Area m ² /g	Pore Volume cm ³ /g	Capacitance F/g
5.5	5%	800	Aerogel	560.98	0.3196	127.7
5.5	5%	800	Xerogel	568.79	0.3648	160.9
5.5	5%	1050	Aerogel	508.40	0.3000	68.32
5.5	5%	1050	Xerogel	521.35	0.2794	105.13
5.5	20%	800	Aerogel	516.52	0.2444	85.84
5.5	20%	800	Xerogel	492.60	0.2592	63.07
5.5	20%	1050	Aerogel	473.67	0.2331	59.03
5.5	20%	1050	Xerogel	459.92	0.2463	74.00
7.0	5%	800	Aerogel	899.51	0.9220	179.12
7.0	5%	800	Xerogel	590.66	0.2025	112.60
7.0	5%	1050	Aerogel	752.90	1.323	142.30
7.0	5%	1050	Xerogel	539.91	0.3959	106.11
7.0	20%	800	Aerogel	929.37	1.3128	146.02
7.0	20%	800	Xerogel	585.84	0.4386	124.26
7.0	20%	1050	Aerogel	803.54	1.4190	143.60
7.0	20%	1050	Xerogel	514.87	0.4416	103.84
			Mean	607.43	0.5439	112.62

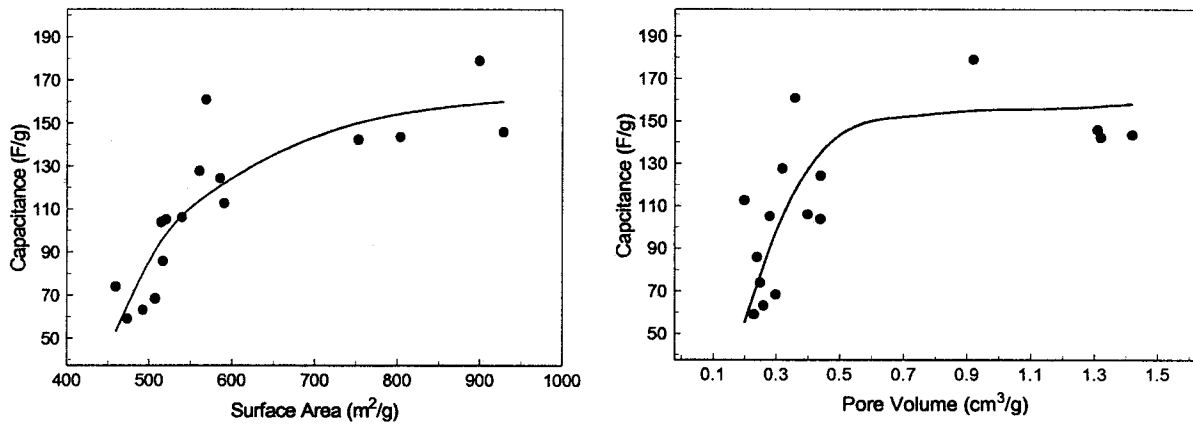


Figure 1. Correlation of the electrochemical capacitance with the surface areas and the pore volumes of the carbon aerogels and xerogels.