

A CARBON FIBER BASED MONOLITHIC ADSORBENT FOR GAS SEPARATION

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Introduction

Monolithic adsorbents based on isotropic pitch fibers have been developed jointly by ORNL and the University of Kentucky, Center for Applied Energy Research. Our monoliths are attractive for gas separation and storage applications because of their unique combination of physical properties and microporous structure. At ORNL we currently produce the monoliths in billets 10-cm diameter and 25-cm long. Here we report the results of a study directed toward attaining uniform activation in these billets.

Experimental

The manufacturing process for our carbon fiber composite molecular sieve (CFCMS) materials has been reported elsewhere [1]. Activation was performed in a three-zone Lindburgh furnace fitted with an 20-cm diameter Inconel retort. The CFCMS samples were dried in vacuum at 300°C, heated to 850°C under He for one hour, cooled to 200°C for a chemisorption treatment in flowing O₂, and then heated again to 850°C in He. The chemisorption and activation steps were repeated until the desired burn-off was attained. Micropore structure characterization was performed using a Quantichrome Autosorb-1 apparatus.

Results and Discussion

A total of 5 billets were subjected to two cycles each of O₂ chemisorption/activation. The burn-off attained ranged from 8.5 to 13.4%, with an average burn-off of 10.4%. A typical (Type I) N₂ isotherm (77K) for sample SMW-8A is shown in Fig.1. The DR micropore volume, BET surface area and DA micropore width for four SMW billets are reported in Table 1. The designations A and B indicate samples taken from opposite ends of the 25-cm long billets. The micropore volumes varied from 0.16-0.34 cm³/g and the BET surface areas range from 425-940 m²/g. Billet SM-15 was subjected to a more detailed examination to determine the uniformity of activation throughout the billet. Samples were taken at several radial locations across slices cut periodically along the billet length. The BET surface area, DR micropore volume and DA micropore size

distribution were determined. Figure 2 shows the variation of DR micropore volume throughout billet SM-15. The DR volume varied from 0.12-0.35 cm³/g, but there was no systematic radial or axial variation.

Figure 3 shows a breakthrough plot for an Ar/H₂S/H₂ mixture flowing through a sample of our CFCMS material. The H₂S was adsorbed, and breakthrough occurs some 18 minutes into the experiment. The H₂ and Ar were not adsorbed. Upon total breakthrough, the feed gas mixture flow was terminated and the desorption of the adsorbed H₂S was achieved by applying an electric current (4.5A @ 1V). Desorption occurred rapidly, as indicated by the increased concentration of H₂S in the exhaust gas stream. Regeneration of the CFCMS was thus achieved by direct electrical heating of the CFCMS. The ability of the material to readily adsorb H₂S suggests the removal of natural gas (NG) odorant additives might be achieved with a guard bed containing a billet of activated CFCMS. Pressure drop data for He flow through an activated billet are shown in Figure 4. The measured pressure drop is slightly greater than for a packed bed of granular carbon. CFCMS is currently being evaluated as a guard bed material for a NG fueled device.

Conclusions

Reasonably uniform activation was achieved in our CFCMS materials. Micropore volumes of 0.12-0.35 cm³/g were attained. The ability of the materials to adsorb H₂S, the acceptable pressure drop, and its electrical heating capability suggests that CFCMS will make an ideal guard bed material.

References

1. Burchell, T.D., Judkins, R.R., Rogers, M.R. and Williams A.M., *Carbon* (1997) 35, In Press.

Acknowledgments

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Table 1. Surface area, micropore volume, and micropore size data for CFCMS billets

Billet No.	BET Surface Area (m ² /g)	DR Micropore Volume (cm ³ /g)	DA Micropore Dia. (nm)
SMW1-A	567	0.27	1.54
SMW1-B	425	0.16	1.64
SMW3-A	607	0.23	1.44
SMW3-B	448	0.17	1.48
SMW4-A	940	0.34	1.56
SMW4-B	488	0.19	1.54
SMW8-A	707	0.27	1.48
SMW8-B	441	0.16	1.54

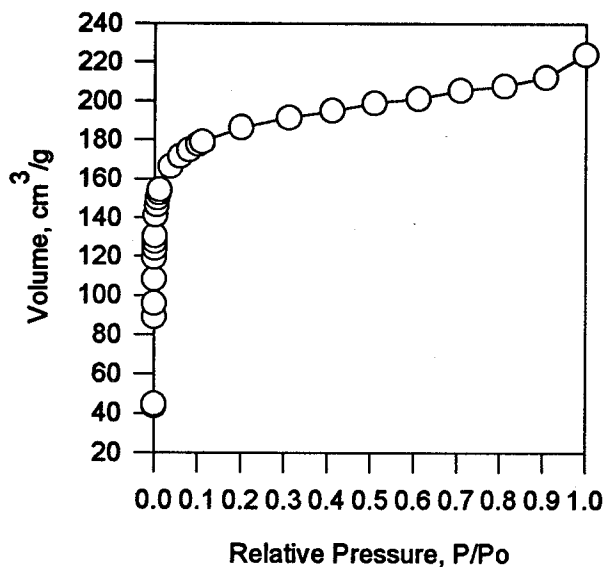


Figure 1. Typical 77K N₂ isotherm for sample SMW8A.

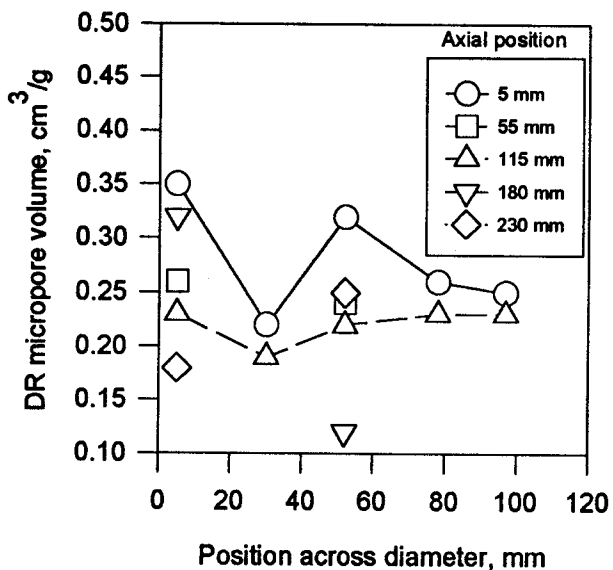


Figure 2. Variation of DR vol. in a large CFCMS billet.

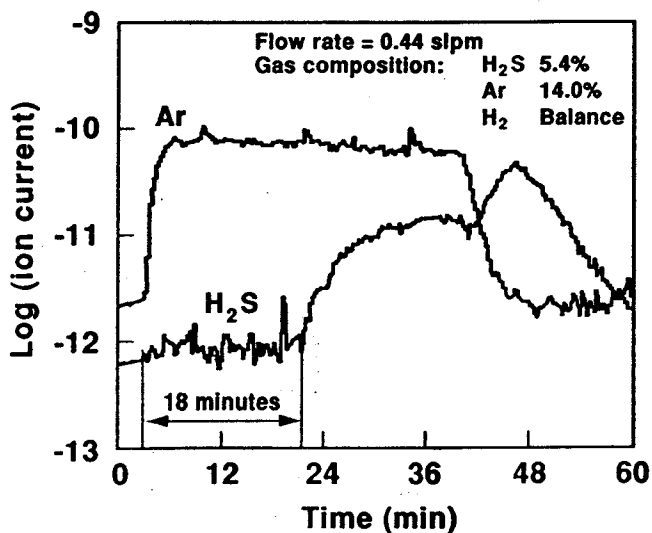


Figure 3. H₂S breakthrough plot for activated CFCMS showing the electrical desorption of the H₂S.

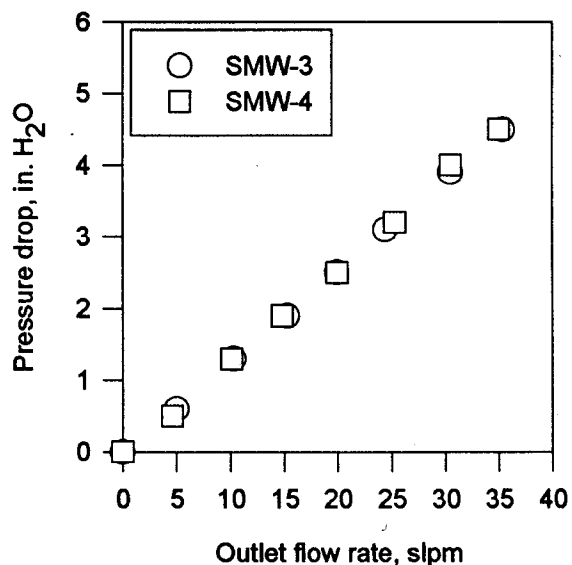


Figure 4. Pressure drop through a large monolith as a function of the He flow rate.