SOME NEW RESULTS ON ISOTHERMAL, ISOBARIC CHEMICAL VAPOR INFILTRATION OF PYROCARBON

W. Benzinger and K. J. Hüttinger
Institut für Chemische Technik
Universität Karlsruhe, Kaiserstr. 12, D-76128 Karlsruhe, Germany

Introduction

Fundamentals and problems of chemical vapor infiltration of pyrocarbon have been described by Kotlensky [1]. In accordance with his presentation it is generally assumed that extremely low infiltration rates should be preferred for an optimum pore filling. An opposite view has been presented recently [2,3]. This paper shows some experimental results supporting this view.

Experimental

Figure 1 shows a cross-section of the deposition reactor. The porous substrate, made of aluminia ceramic, is 16 mm in diameter and 20 mm in height. Total porosity amounts to 23 %, the pore diameters are in the range from 1 to 30 μm. Infiltration studies were performed with pure CH_4, CH_4/Ar-, CH_4/H_2/Ar- and CH_4/H_2-mixtures at 1100°C, various total pressures, and at a residence time of the gas in the annulus around the substrate of 0,16 s.

Results

Figure 2 shows relative mass gains of the substrate due to infiltration of the pores obtained at a total pressure of 20 kPa with increasing CH_4 initial partial pressures. Infiltration rates and degrees of pore filling, as found by mercury porosimetry and calculated from an extrapolation of the curves, increase with increasing partial pressure. A plot of reciprocal infiltration rates as a function of reciprocal CH_4 initial partial pressures yields a straight line indicating progressive saturation adsorption with increasing CH_4 partial pressure. This result explains increasing degrees of pore filling, because more and more active sites at the internal surface are involved in pyrocarbon deposition.

Variation of total pressure up to 100 kPa with pure CH_4 shows increasing initial infiltration rates, but the maximum degree of pore filling is found at 20 kPa. Hydrogen inhibits pyrocarbon deposition, but H_2 in the feed gas was proposed to improve the pore filling [2,3]. Figure 3 shows results obtained at 20 kPa total pressure with (a) a CH_4/Ar-mixture (3:1) and (b) a CH_4/Ar/H_2-mixture (6:1:1).The inhibiting effect of H_2 is obvious. The results of mercury porosimetry after 52 hours infiltration time (mixture (a)) and 77 hours (mixture (b)) are shown in Figure 4. The degrees of pore filling amount to 83 % (a) and 91 % (b). The more interesting effect of H_2 results from the micropore volume (d_{pore} < 0,1 μm) which was generated; it is clearly lower with hydrogen.

To overcome the inhibiting effect of H_2 a higher CH_4 partial pressure has to be used. A result with a CH_4/H_2-mixture (7:1) at 50 kPa total pressure is presented in Figure 5. At such a pressure infiltration is even faster than that with pure CH_4 at 20 kPa, which is presented for comparison. The degree of pore filling determined by mercury porosimetry amounts to 87%.

Conclusions

(1) Infiltration is accelerated by increasing partial pressures up to a limiting value. (2) Increasing infiltration rates have a beneficial effect on the degree of pore filling. (3) CH_4/H_2-mixtures (ratio of about 7:1) are optimum for a maximum degree of pore filling.

References

1. W.V. Kotlensky, in: *Chemistry and Physics of Carbon,* Vol. 9, (Edited by P.L. Walker, P.A. Thrower), p. 173, M. Dekker, New York, 1973.

2. W. Benzinger and K.J. Hüttinger, Carbon, 34, 1465 (1996).

3. German Patent Application No. 196 46 094.8 (1996)

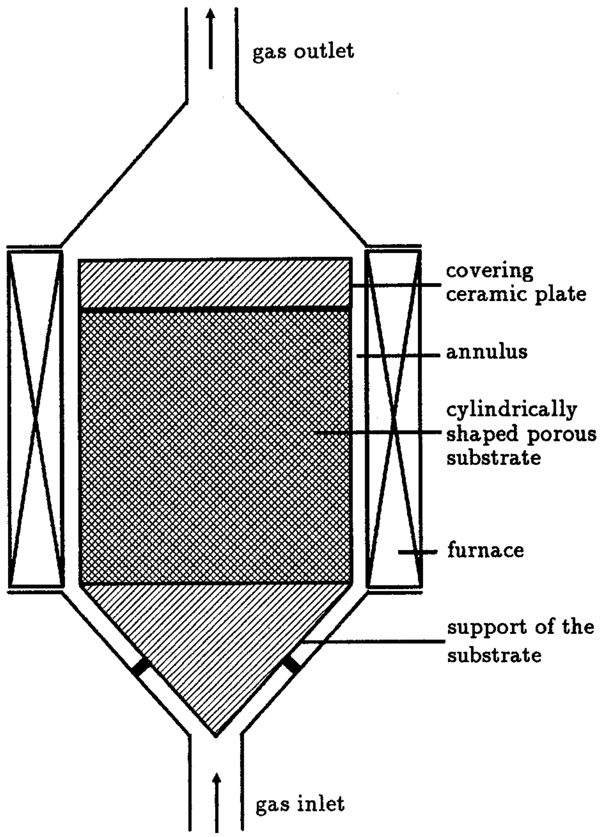

Figure 1. Scheme of the infiltration reactor.

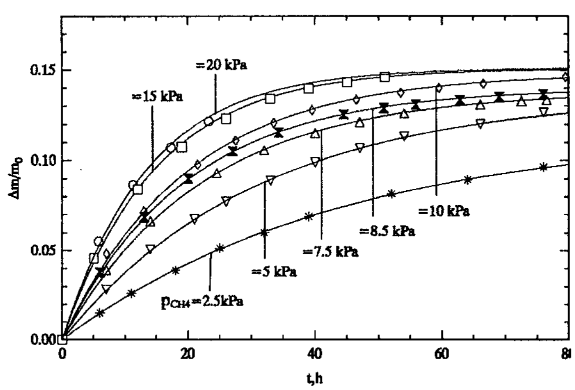

Figure 2. Relative mass gains of the porous substrate with increasing CH_4 partial pressures at a total pressure of 20 kPa.

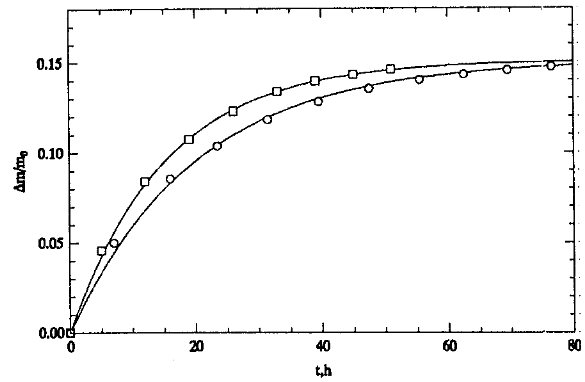

Figure 3. Relative mass gains obtained with mixture a (□) and mixture b (○) at a total pressure of 20 kPa.

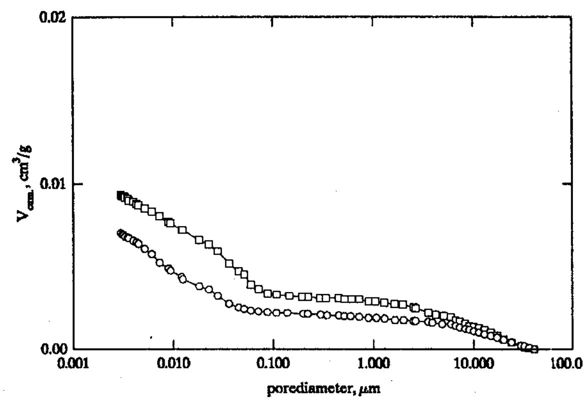

Figure 4. Results of mercury porosimetry after infiltration of the porous substrate using mixture a (□) and mixture b (○).

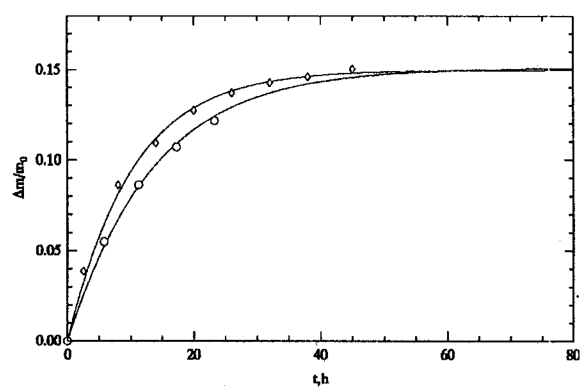

Figure 5. Relative mass gains at a total pressure of 50 kPa for a CH_4/H_2 mixture (7:1) (◊), and at a total pressure of 20 kPa for pure CH_4 (○).