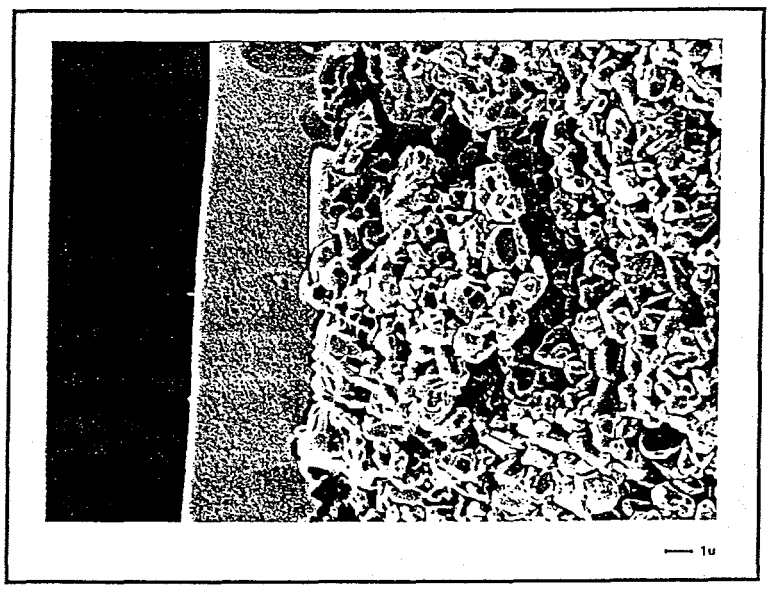
(ii) Bubble formation occurs in the carbon membrane if the PVDC film thickness is >~15 μ (**Figure 12**). The bubbles cause "local roughness", giving the tube interior surface a matte appearance.
(iii) Local variations in thickness can be induced by porosity variations and tube imperfections (e.g., pits), thus leading to gross local defects (e.g,. delamination, bubbles).
(iv) Batch-to-batch variations in tube porosity result in SSF membranes with variations in thickness if the PVDC is coated from an emulsion with a constant solids content.
(v) **Figures 13 and 14** show the variation in carbon coating thickness along the tube length and circumference as observed by SEM. There is some variation in the thickness along the tube circumference at the top, but not along the length for this particular tube. Local variations in tube porosity are likely responsible for this variation. Improvement in uniformity may allow one to reduce the membrane thickness.

To successfully prepare a membrane, a uniform and defect-free coating with the desired separation properties is prepared by judiciously choosing the conditions described in the above membrane preparation steps.

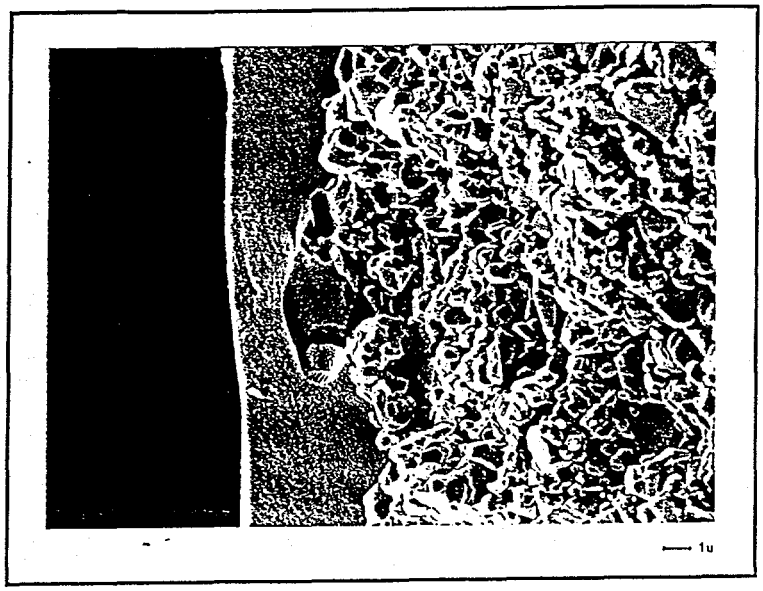
The as-is prepared SSF membrane on the alumina tube consists of nascent carbon, which can react slowly with atmospheric air or water. A membrane passivation step that allows controlled oxidation of the carbon membrane was developed on carbon sheets (10) and transferred to tubes. This step involves introducing air into the pyrolysis furnace at 350 C and exposing the membrane to air for a short period, followed by cooling in nitrogen. The SSF membrane prepared on the tubular support was treated in air at 350 C for various times and the mixed gas performance evaluated . The propylene rejection and the A/F at different hydrogen recoveries is shown in **Figures 15 and 16** for different passivation times. The data indicate that the the membrane performance deteriorates with 120 and 180 min of treatment. The results between 0 and 60 min treatment were indistinguishable. A thirty minute treatment in air was chosen as the standard passivation condition.

3.4 Reproducibility of SSF Membrane Preparation

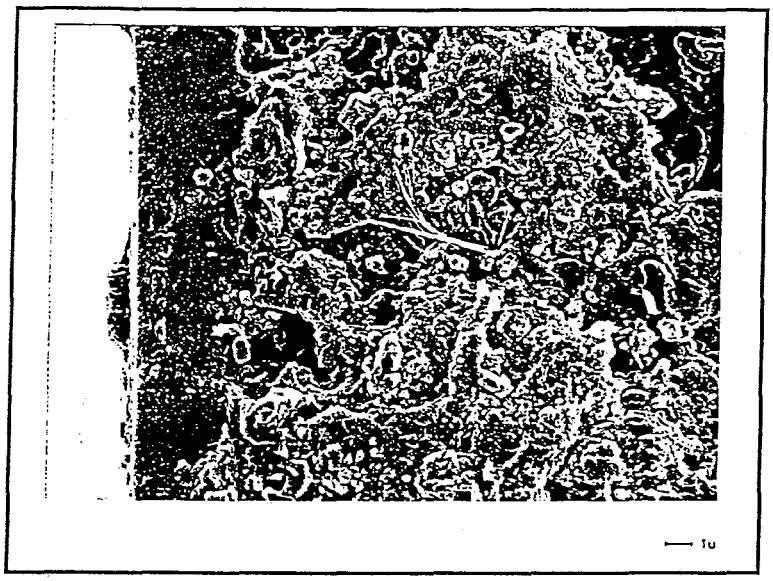
After the alumina tube preparation technique was improved with the tube supplier so that tubes with consistent properties were obtained, the reproducibilty of the SSF membrane preparation technique was evaluated using a single batch of alumina tubes. **Tables 12 and 13** show results of mass/thickness of PVDC coating, helium permeance with PVDC coated tubes, pyrolyzed membrane thickness and the permeance of helium and carbon dioxide through the carbonized membrane for 65 tubes. The data show that the SSF membrane preparation on these tubes is reproducible. The variabilities in the PVDC average coating thickness and He permeance are low; the variations in carbon coating thickness and pure gas He and CO_2 permeances through the SSF membrane are also low. Pure gas permeation measurement with He and CO_2 though the SSF membrane is a good screening tool to determine if the pores in the membrane are in the desired size range. In separations by sieving and Knudsen mechanisms, pure He permeance is greater than pure CO_2 permeance. However, in the selected surface flow regime, the permeation



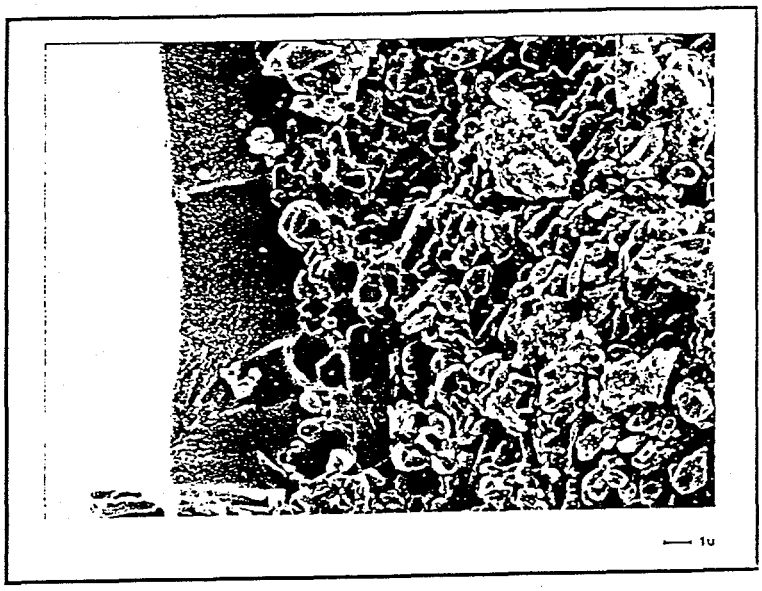
Title Tube 18 bottom MA18B1 Date 04-07-95 Time 10:01
Comment x-section at coating/support interface
Mag X5,000 kV 5 WD 9mm Spot 6 Scan P3



Title Tube 18 bottom MA18B2 Date 04-07-95 Time 10:07
Comment x-section at coating/support interface
Mag X5,000 kV 5 WD 9mm Spot 6 Scan P3

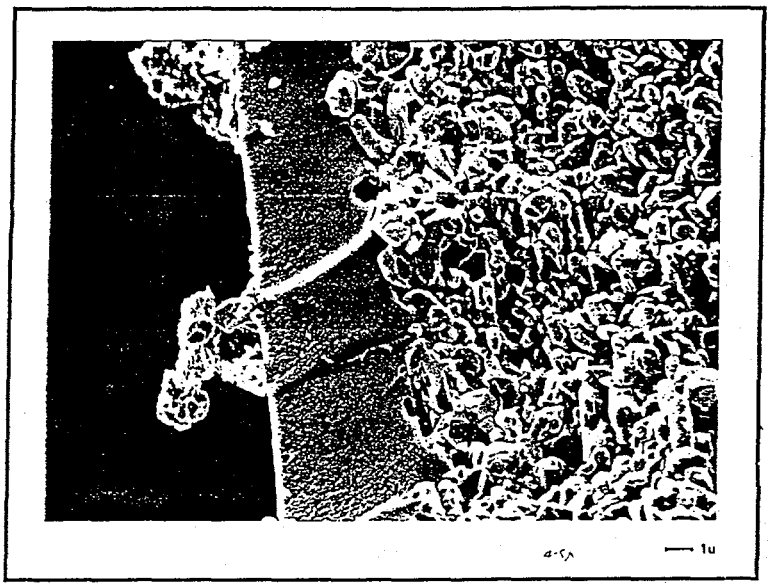


Title Tube 18 bottom 2 MA18B22 Date 04-07-95 Time 10:22
Comment x-section at coating/support interface
Mag X5,000 kV 5 WD 10mm Spot 6 Scan P3

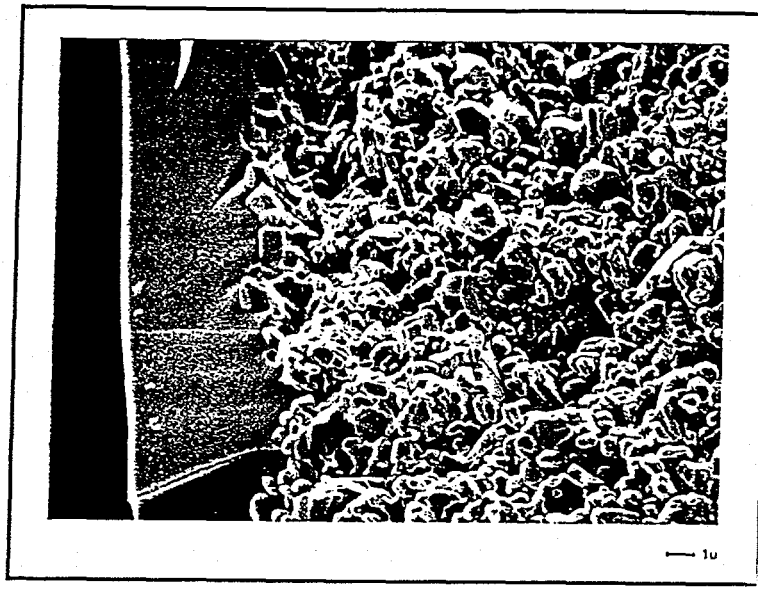


Title Tube 18 bottom 2 MA18B21 Date 04-07-95 Time 10:14
Comment x-section at coating/support interface
Mag X5,000 kV 5 WD 11mm Spot 6 Scan P3

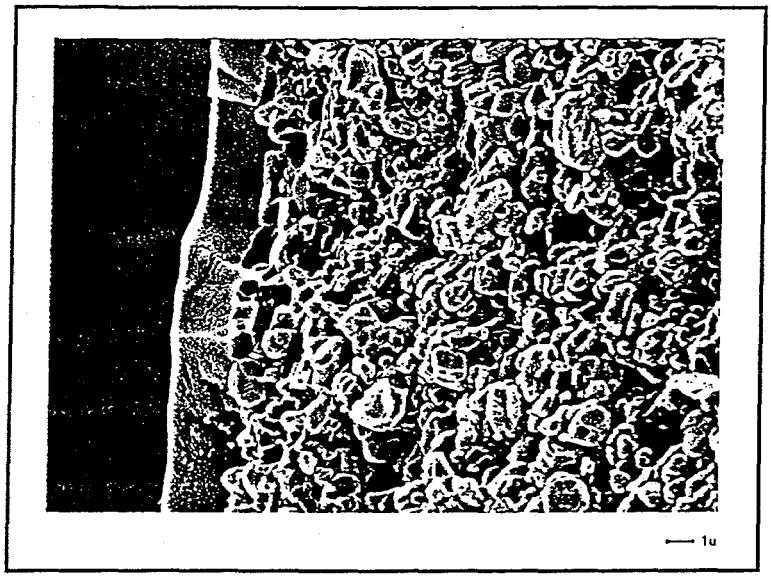
Figure 13. SEM Showing Coating Uniformity in the Tube Circumference at the Bottom of the Membrane



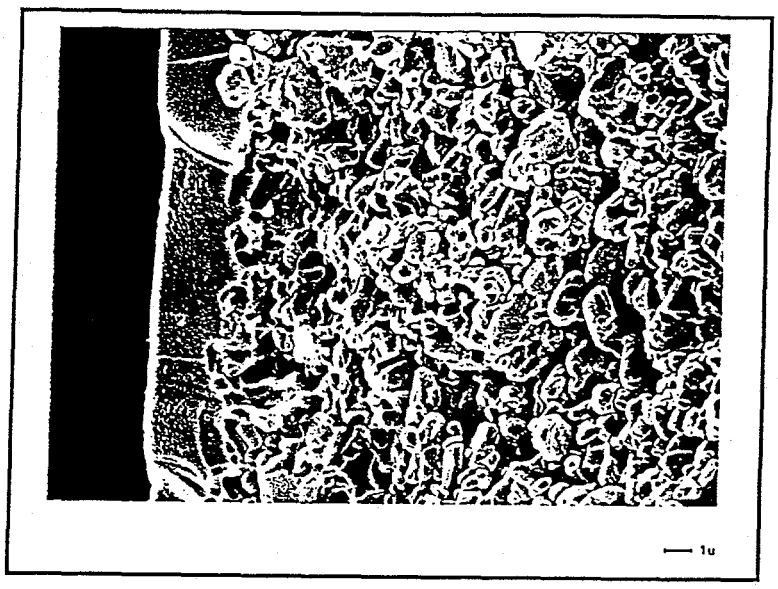
Title Tube 18 top MA18T1 Date 04-07-95 Time 09:20
Comment x-section at coating/support interface
Mag X5,000 kV 5 WD 13mm Spot 7 Scan P3



Title Tube 18 top MA18T2 Date 04-07-95 Time 09:26
Comment x-section at coating/support interface
Mag X5,000 kV 5 WD 12mm Spot 7 Scan P3



Title Tube 18 top 2 MA18T21 Date 04-07-95 Time 09:35
Comment x-section at coating/support interface
Mag X5,000 kV 5 WD 12mm Spot 7 Scan P3



Title Tube 18 top 2 MA18T22 Date 04-07-95 Time 09:45
Comment x-section at coating/support interface
Mag X5,000 kV 5 WD 12mm Spot 6 Scan P3

Figure 14. SEM Showing Coating Uniformity in the Tube Circumference at the top of the Membrane

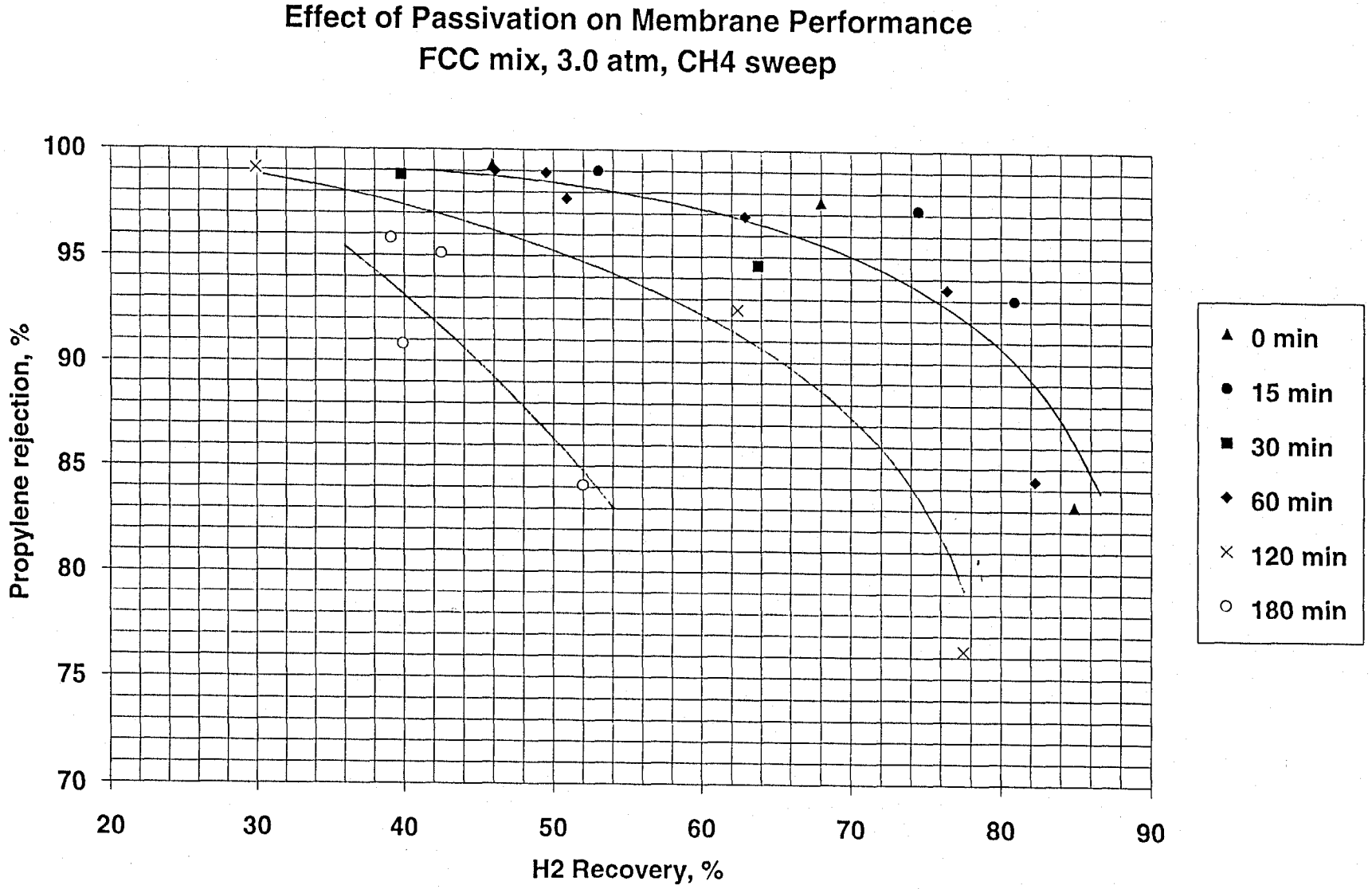


Figure 15. Effect of Passivation Time on Propylene Rejection

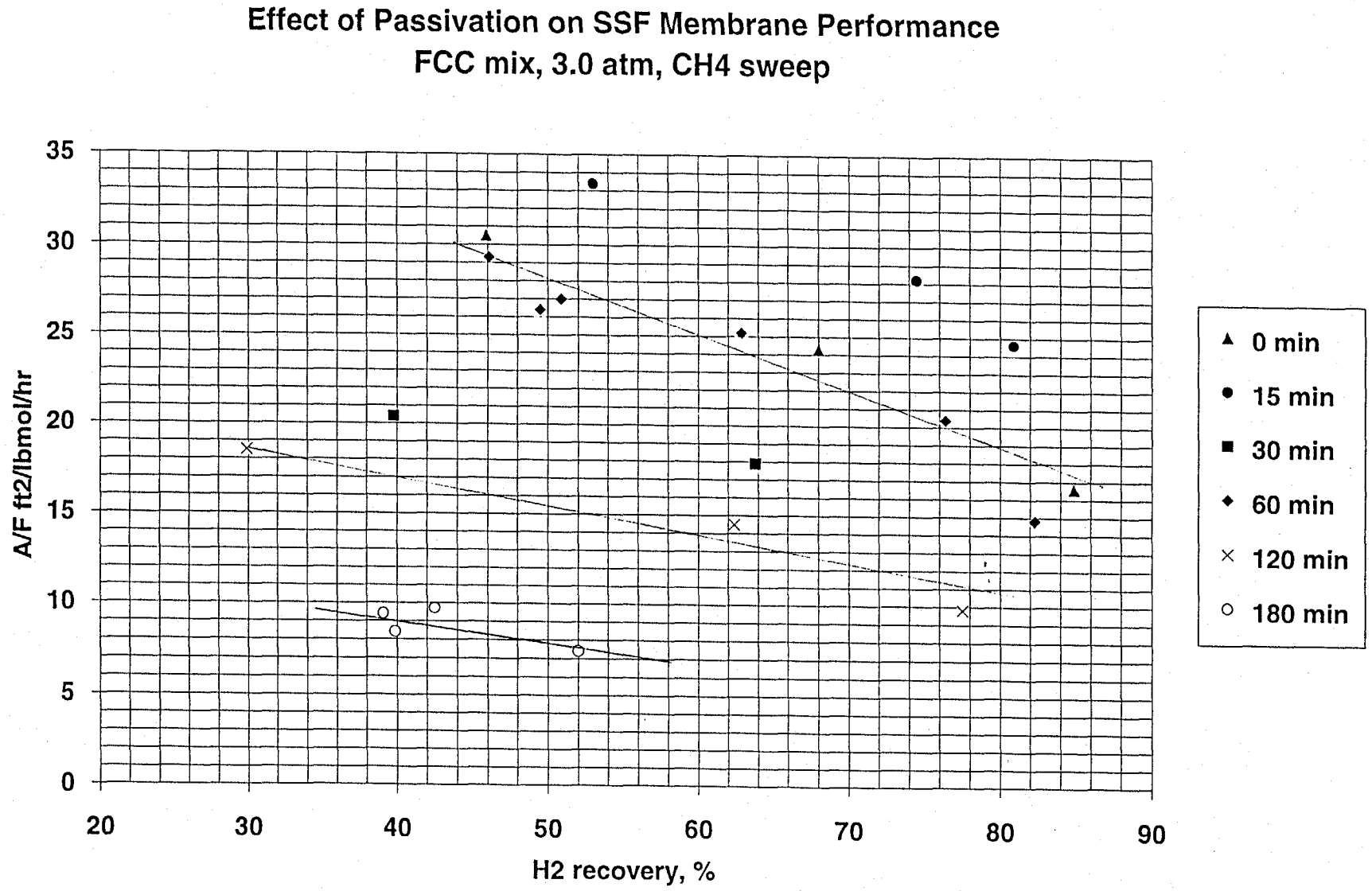


Figure 16. Effect of Passivation Time on Membrane A/F

REPRODUCIBILITY OF SSF MEMBRANE PREPARATION

TABLE 12

Tube #	Mass of polymer	He P/l x10E5 B/cm PVDC	Mass of carbon	He P/l x10E5 B/cm membrane	CO2 P/l x10E5 B/cm membrane	H2 recovery membrane	C3= rejection membrane	A/f ft2/lbmol/h membrane
14006-35-1bcl	0.1092	11.0	0.0267	144	331	54	97.6	24.3
14006-35-2bcl	0.1078	10.2	0.0267	116	327	44.8	98.4	27.2
14006-35-3bcl	0.1111	12.4	0.0282	95	337	49.9	97.9	24.4
14006-35-4bcl	0.1089	11.5	0.0269	121	356	51.3	98.4	27.1
14006-35-5bcl	0.1117	15.6	0.0264	135	359	47.6	98.2	24.1
14006-35-6bcl	0.1177	10.7	0.0348	147	402	43.9	97	19.9
14006-35-7bcl	0.1224	14.2	0.0374	190	388	48	96.9	19.9
14006-35-8bcl	0.1189	12.8	0.0371	226	330	48.5	94.8	14.3
14006-35-9bcl	0.1233	11.5	0.0362	165	407	46.4	97.7	19.3
14006-35-10bcl	0.1250	10.8	0.0383	198	349	41.4	91.8	12.9
14006-35-11bcl	0.1156	13.9	0.0336	107	329	55.4	96.6	21.7
14006-35-12bcl	0.1145	12.2	0.0323	155	319	64.6	96.5	22.4
14006-35-13bcl	0.1165	10.8	0.0343	140	393	55.5	96.1	20.2
14006-35-14bcl	0.1221	12.8	0.0345	167	340	50.9	96.4	17.6
14006-35-15bcl	0.1191	10.4	0.0353	160	385	51.7	96.6	20.4
14006-35-16bcl	0.1177	9.7	0.0340	178	379	46.8	97.4	20.6
14006-35-17bcl	0.1210	9.6	0.0362	225	392	44.4	94.3	13.6
14006-35-18bcl	0.1234	20.1	0.0358	278	412	45.5	81.6	7.7
14006-35-19bcl	0.1236	14.1	0.0356	244	398	37.6	95.3	12.7
14006-35-20bcl	0.1225	17.5	0.0341	228	396	52.5	90.2	13.1
14006-35-1	0.1305	23.2	0.0366	167	383	52.2	93.5	20.0
14006-35-2	0.1323	13.8	0.0364	173	374	40.3	95.8	20.1
14006-35-3	0.1244	12.5	0.0443	173	382	64	96.5	23.6
14006-35-4	0.1347	8.8	0.0329	97	323	61.7	98.3	27.3
14006-35-5	0.1269	19.2	0.0340	184	378	50.6	97.9	24.0
14006-35-6	0.1297	9.7	0.0358	171	399	65.6	95.1	21.1
14006-35-7	0.1216	10.4	0.0300	155	362	64.1	97.6	27.3
14006-35-8	0.1276	12.3	0.0334	185	401	38.6	99.1	24.0
14006-35-9	0.1244	12.2	0.0340	202	427	55.2	95.8	19.4
14006-35-10	0.1193	8.8	0.0325	179	367	47.4	98.8	27.7
14006-35-11	0.1229	12.3	0.0332	165	365	33.9	99	21.6
14006-35-12	0.1223	9.3	0.0304	163	384	80.7	93.5	21.5
14006-35-13	0.1250	12.5	0.0445	175	379	47.2	98.5	24.2
14006-35-14	0.1226	12.6	0.0320	161	378	63.1	96.8	24.5
14006-35-15	0.1257	17.7	0.0327	172	405	52.3	97.6	21.6
14006-35-16	0.1289	11.7	0.0323	200	423	32.8	98.6	21.3
14006-35-17	0.1243	10.4	0.0326	179	335	50.5	97.2	21.4
14006-35-18	0.1107	9.9	0.0283	174	312	80.7	92.7	21.4
14006-35-19	0.1342	16.7	0.0349	179	350	34.6	98.3	19.1
14006-35-20	0.1198	9.9	0.0319	110	331	45.1	98.7	38.0
bcl tubes								
Average	0.1176	12.6	0.0332	171	366.5			
Std dev.	5.50E-03	2.70E+00	4.00E-03	49.5	31.6			
others								
Average	0.1254	12.7	0.0341	168.1	372.9			
Std dev.	5.60E-03	3.80E+00	4.10E-03	25	31.2			

REPRODUCIBILITY OF SSF MEMBRANE PREPARATION

TABLE 13

Tube #	Mass of polymer	He P/l x10E5 B/cm PVDC	Mass of carbon	He P/l x10E5 B/cm membrane	CO2 P/l x10E5 B/cm membrane	H2 recovery membrane	C3= rejection membrane	A/f ft2/lbmol/h membrane
14006-20-1	0.1199	8.1	0.0308	65.2	294	56.3	96.9	33.5
14006-20-2	0.1278	6.9	0.0337	94.1	395	50.9	97.9	26.7
14006-20-3	0.1143	4.6	0.0299	78	298	53.9	97.6	34.2
14006-20-4	0.1166	6	0.0311	93	423	47.8	98.2	28.4
14006-20-5	0.12	8.7	0.0324	101.3	435	39.8	98.5	24.5
14006-20-6	0.1186	8.8	0.0319	105.4	475	46.3	98.2	22.4
14006-20-7	0.1325	6.5	0.0361	59.5	262	51.5	97.6	37.9
14006-20-8	0.1342	8.2	0.0344	80.8	415	50.7	97.7	27.3
14006-20-9	0.1357	7.5	0.031	101.5	429	47.1	98.2	25.1
14006-20-10	0.1247	8.8	0.0331	101.1	471	46.8	98.8	26.5
14006-20-11	0.1267	9.8	0.0351	94.6	408	44.4	98.5	25.3
14006-20-12	0.1249	8.9	0.0321	118	390	30.5	96.6	16.9
14006-20-13	0.1179	9.5	0.0305	108	400	39	97.6	19.3
14006-20-14	0.1172	8.7	0.0291	102	411	47.6	98.3	23.7
14006-20-15	0.1138	9.9	0.0293	106.3	407	44.9	97	20.9
14006-20-16	0.1305	8.1	0.0279	118.3	446	37.2	96.6	17.3
14006-20-17	0.121	13.7	0.0289	96.9	398	42.9	98.3	24.7
14006-20-18	0.1293	10.2	0.0306	101.8	438	34.2	98.8	25.3
14006-20-19	0.1377	11.9	0.0341	70.4	340	48.2	98.4	35
14006-20-20	0.1353	11	0.0347	81.4	367	46.4	98.1	27.5
14006-20-21	0.112	8.9	0.0281	129.1	432	38.6	98.3	22.9
14006-20-22	0.1174	8.1	0.0346	90.9	362	40.1	97.2	24.8
14006-20-23	0.1846	9.3	0.0387	84.9	425	44.7	98.6	27.7
14006-20-24	0.1441	14.7	0.0548	100.8	425	39.8	98.6	23
14006-20-25	0.1275	13.4	0.0318	95.2	449	55.6	97.5	23.3
Average	0.1275	9.2	0.0354	95.1	400			
Std dev.	1.46E-02	2.35E+00	1.41E-02	16.4	53.6			