of CO_2 is greater than that of helium. This screening test is not successful in distinguishing between marginal, good or very good tubes. Thus, the value of this test is strictly in screening to determine if mixed gas permeation tests should be carried out.

3.5 Mixed Gas Performance Characteristics

Mixed gas performance properties of the SSF membranes were measured using a hydrogen/hydrocarbon mixture. This gas mixture, referred to as the refinery Fluid Catalytic Cracker (FCC) off-gas, contains 20% H_2, 20% CH_4, 8%C_2H_4, 8%C_2H_6, 15% C_3H_8,and 29% C_3H_6. The performance definitions with pure and mixed gases were shown in **Table 1**. For the SSF membrane, the objective is to simultaneously maximize the H_2 recovery in the high pressure effluent and the rejection of the other components, viz. hydrocarbons, to the permeate side so that the H_2-enriched stream is recovered as the high pressure effluent. In addition to the rejections of hydrocarbons at different hydrogen recoveries, the membrane area required for the separation is an important membrane performance characteristic. This is noted as A/F where A is the membrane geometric area and F the feed rate to the membrane at the specific hydrogen recovery.

Figures 17-22 show some typical mixed gas membrane performance characteristics with the SSF tubular membranes using the FCC gas at a feed pressure of 3 atm and sweeping with methane at ~1atm. The data show the following :
(i) As the hydrogen recovery in the high pressure effluent increases, the hydrocarbon rejection to the permeate stream decreases.
(iii) The highest rejection from the mix is the species most selectively adsorbed (propylene) and the rejection decreases for the less selectively adsorbed species (C_3>C_2>C_1).
(iii) For example, at 50% hydrogen recovery, the propylene and propane rejections are >98% and >97%, respectively.
(iv) The membrane area increases at lower hydrogen recoveries (i.e., higher hydrocarbon rejections). This indicates that the membrane area required for separation is higher if more hydrocarbon molecules permeate to the low pressure side of the membrane.
(v) There can be a significant variability in the membrane A/F from tube to tube, perhaps due to small differences in the support or membrane structure **(Figure 22)**.

The membranes were also tested at feed pressures of 5 atm. The data show that the membrane separation properties are not changed with this gas mixture except that the A/F expectedly decreases as the feed pressure is increased **(Figure 22)**.

With the above benchmark data from some tubes, an effort was initiated to prepare a larger number of tubes which would demonstrate reproducibilty and lot-to-lot variability in membrane performance. In addition, these tubes would be used in a multi-tube module so that a larger membrane area could be tested in addition to developing the module design and scale-up criteria.

PHASE1.XLS Chart 1

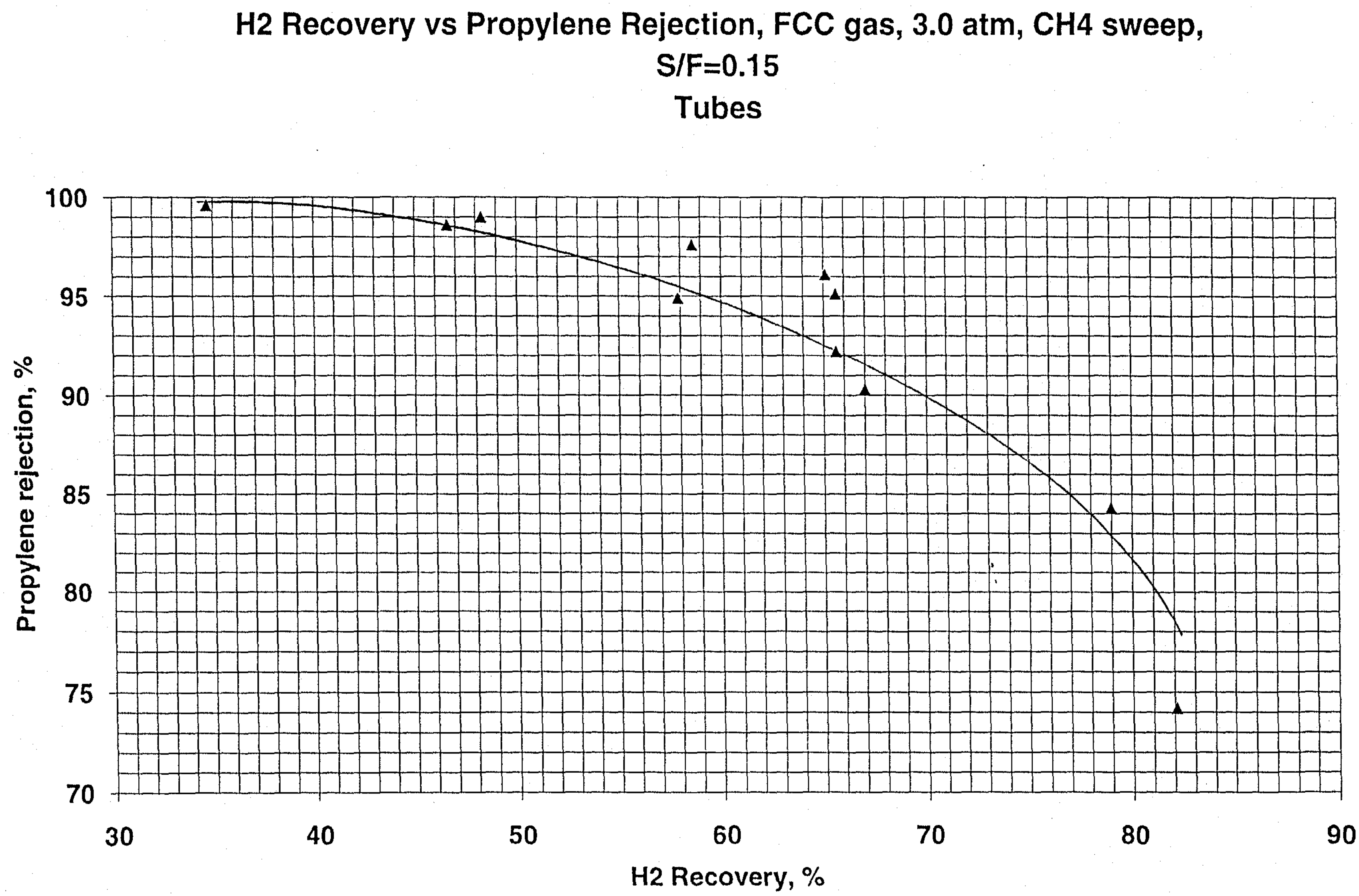


Figure 17. Typical H_2 Recovery vs Propylene Rejection Profiles for Tubes Coated with SSF Membrane and Tested with FCC Mix

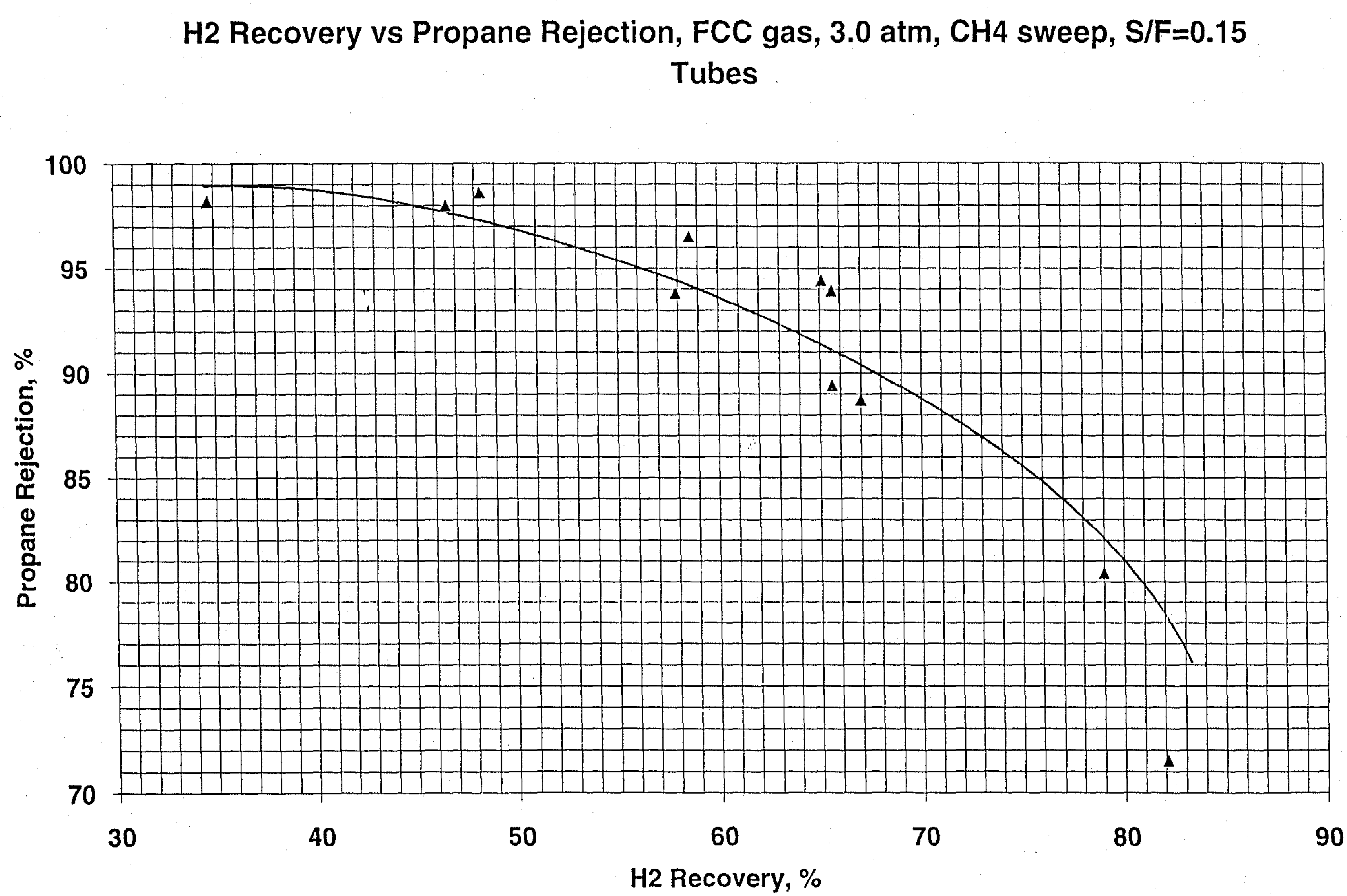


Figure 18. Typical H_2 Recovery vs Propane Rejection Profiles for Tubes Coated with SSF Membrane and Tested with FCC Mix

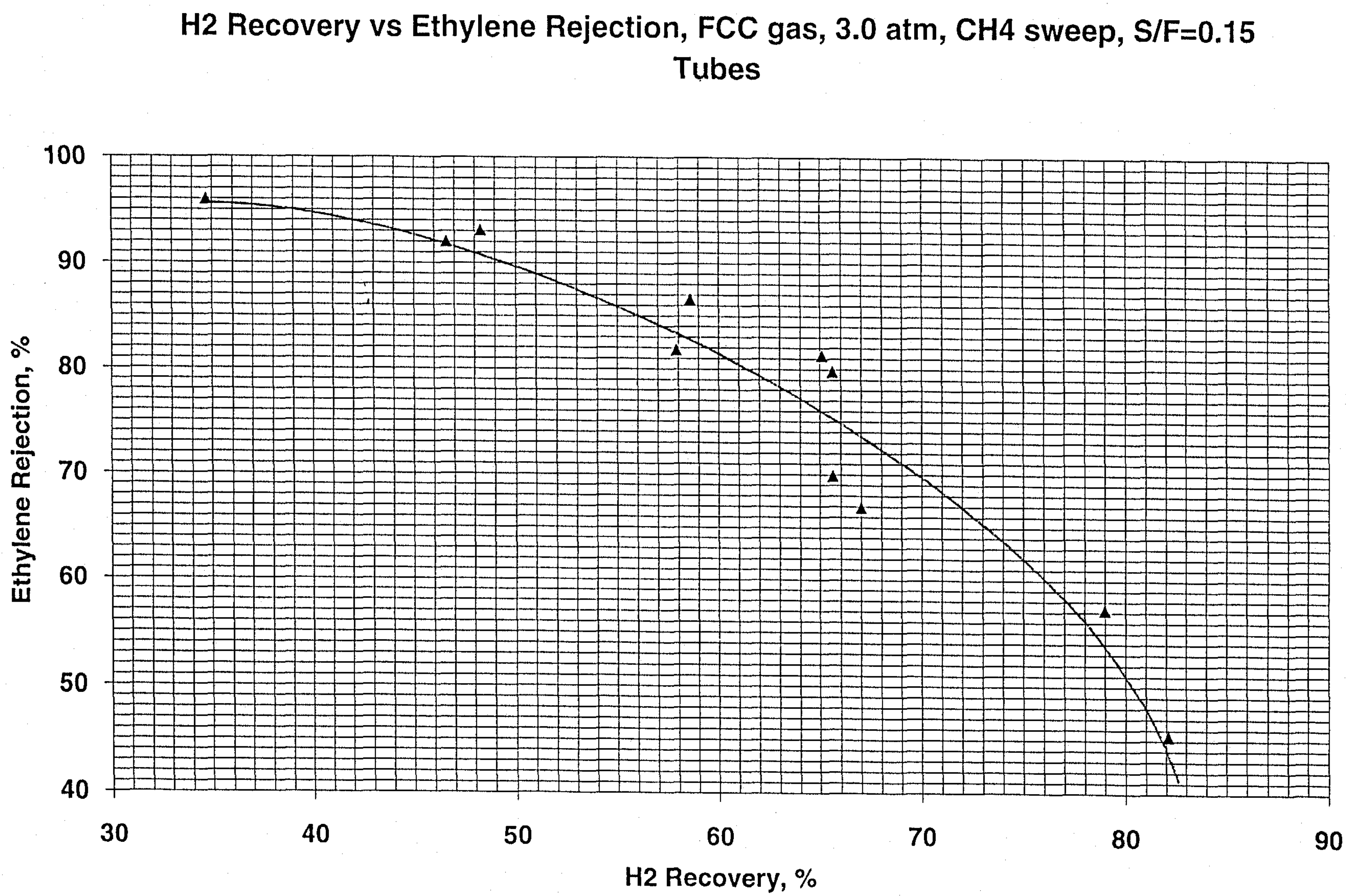


Figure 19. Typical H_2 Recovery vs Ethylene Rejection Profiles for Tubes Coated with SSF Membrane and Tested with FCC Mix.

PHASE1.XLS Chart 4

H2 Recovery vs Ethane Rejection, FCC gas, 3.0 atm, CH4 sweep, S/F=0.15
Tubes

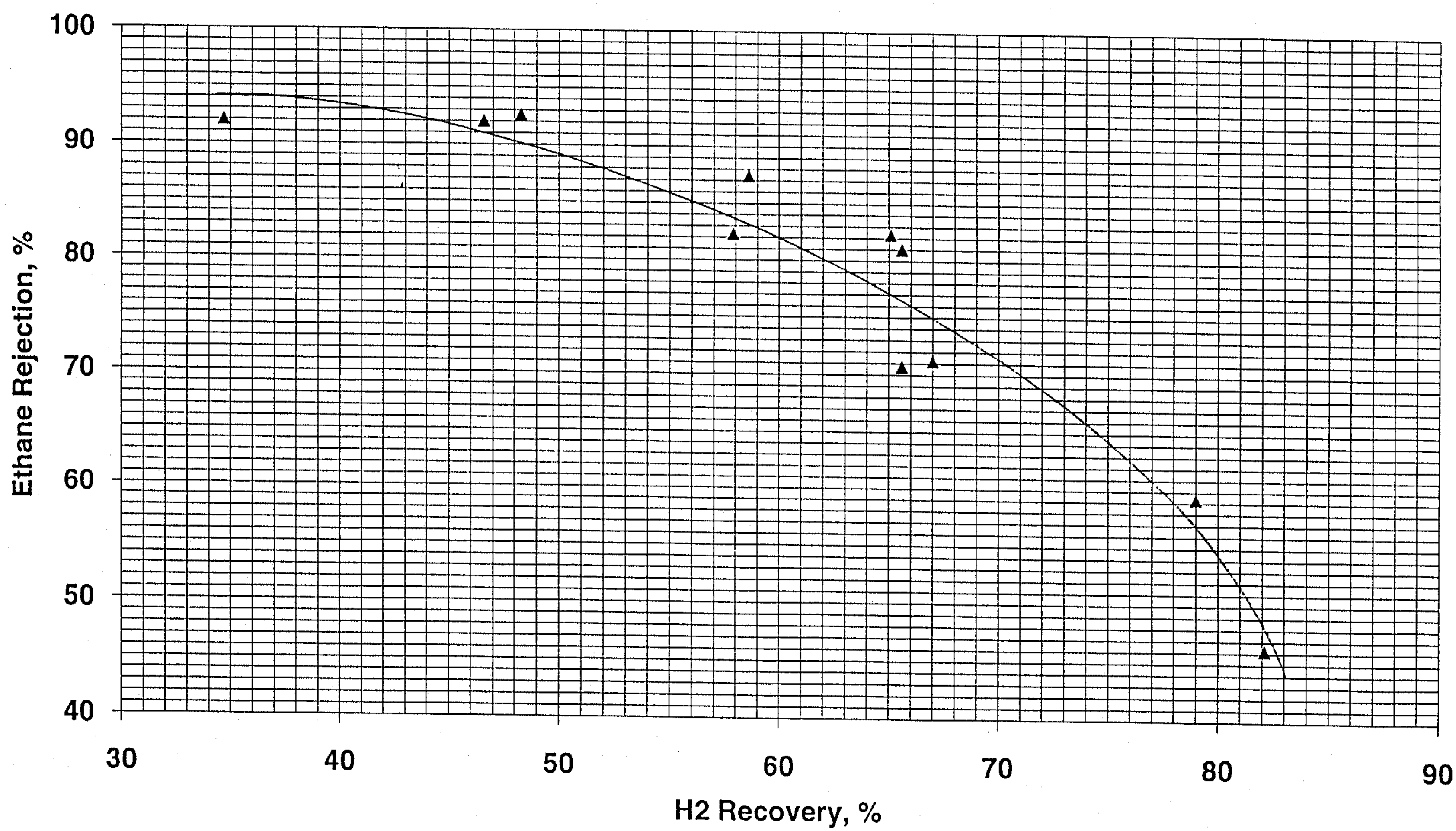


Figure 20. Typical H_2 Recovery vs Ethane Rejection Profiles for Tubes Coated with SSF Membrane and Tested with FCC Mix.

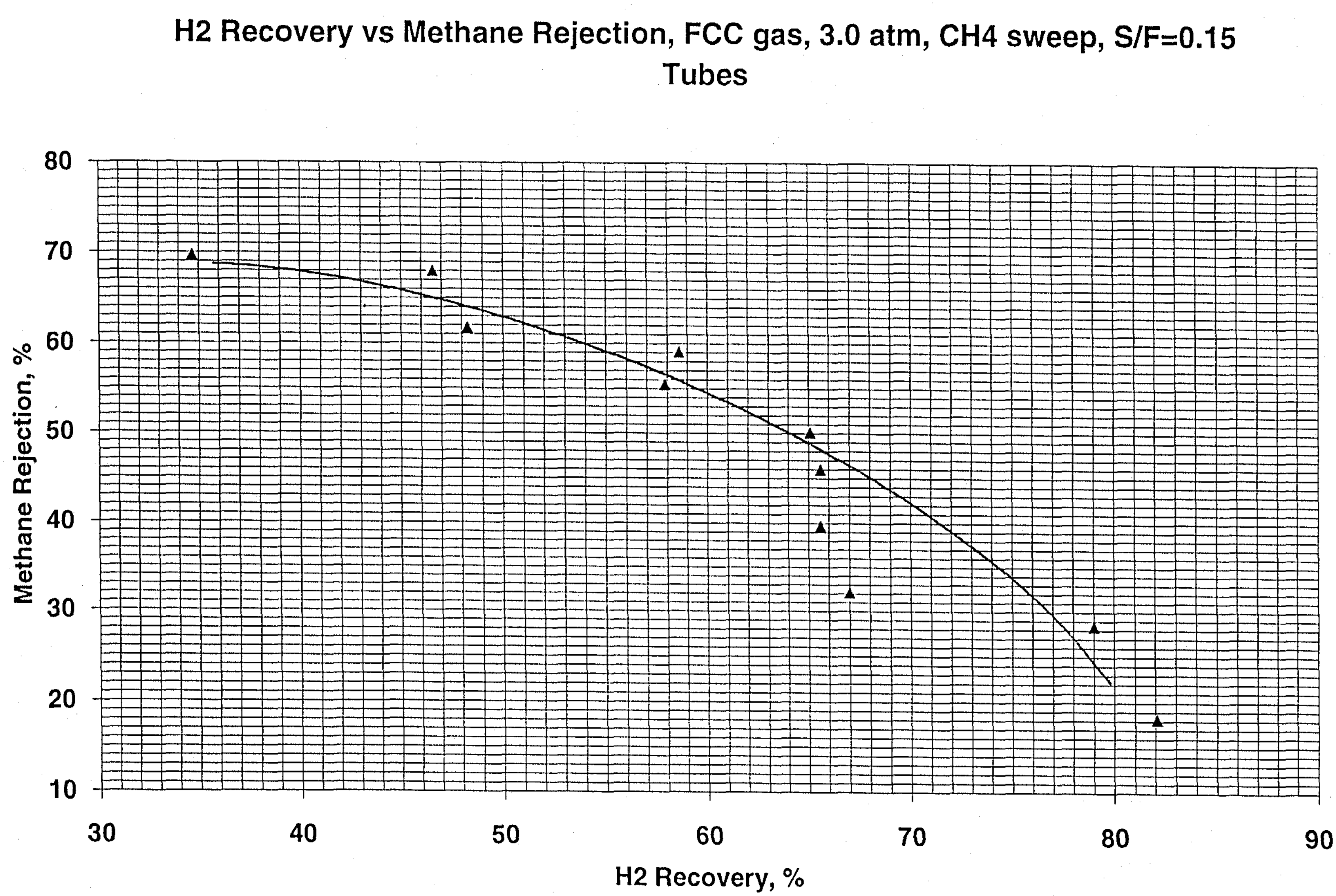


Figure 21. Typical H_2 Recovery vs Methane Rejection Profiles for Tubes Coated with SSF Membrane and Tested with FCC Mix

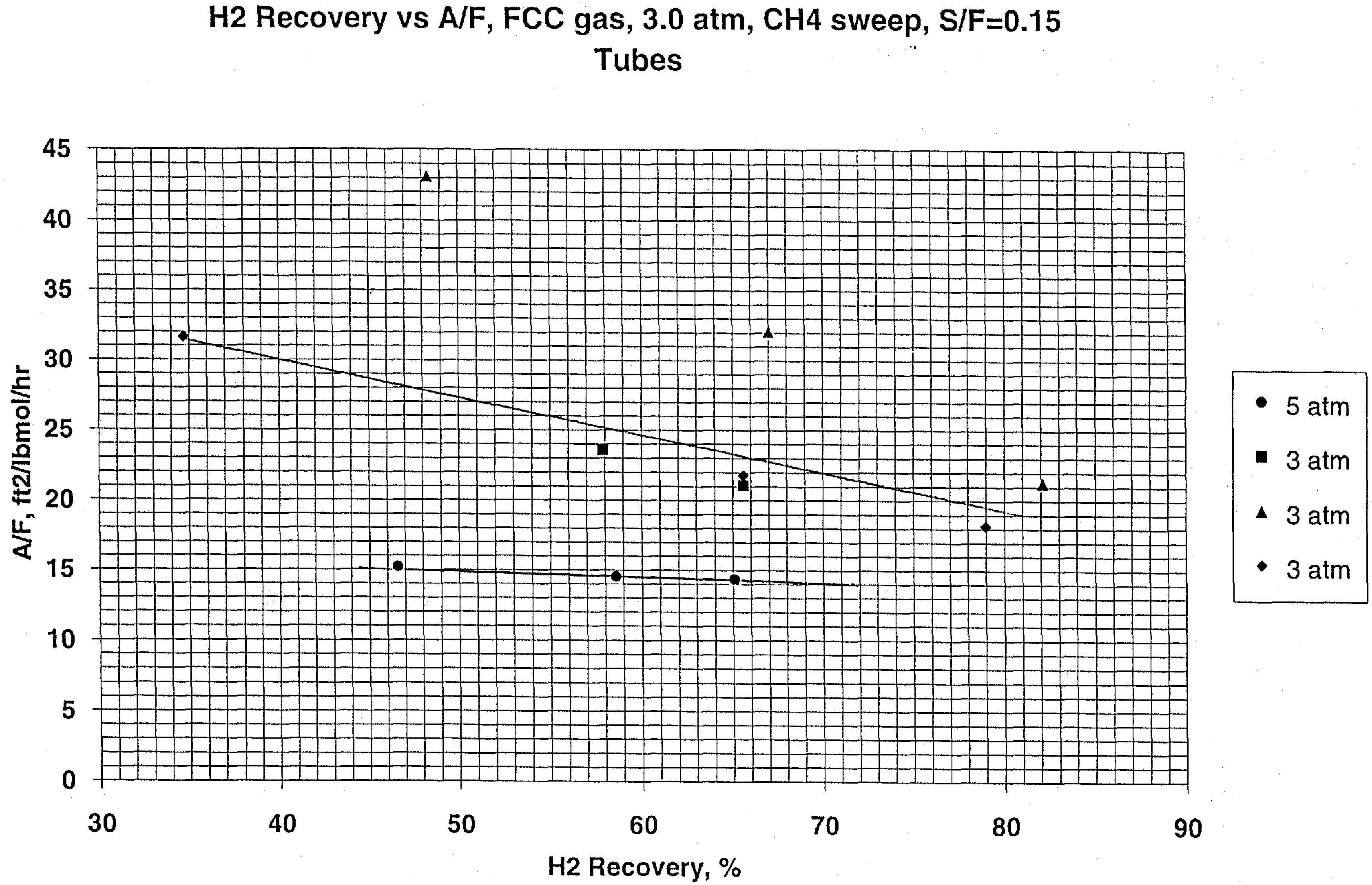


Figure 22. Typical H_2 Recovery vs Membrane A/F Profiles for Tubes Coated with SSF Membrane and Tested with FCC mix; note lower A/F at higher pressures

3.6 Design/Construction of Multi-Tube Module

One of the deliverables in this program is the scale-up of the SSF membrane from single tubes to multiple tubes in a housing. A module was designed to house 19 tubes which represent 1 ft^2 of membrane area. A triangular pitch hexagonal shaped layout with a center-to-center distance of 1.3 x Tube O.D was designed and built. The details of the module are shown in **Figure 23**.

The module has the capability of being fed from the top or the bottom and with a countercurrent or co-current sweep. The module was cleaned with a hydrocarbon solvent followed by a low boiling point halogenated solvent to remove contamination after welding.

3.7 Design/Construction of Membrane Module Test Unit

A membrane test unit was designed to test the performance of the 1 ft^2 membrane area module. The key features of this system are :

(i) Gas supply through individual cylinders so that suitable gas mixtures can be blended. Single gas tests can also be performed.
(ii) On-line gas chromatograph (GC) for analysis of feed, effluent and permeate streams.
(iii) Variable pressure (max 125 psig) and flow rate (max 50 liters/min) capability so as to be able to deliver different gas flow rates at a fixed pressure.
(iv) Capability of feeding gas to membranes from 0.2 to 2 ft^2 in membrane area.
(v) Safety alarms, shrouds and shutdown for flammable gas handling.

The process and instrumentation diagram (PID) for the system is shown in **Figure 24**. The system was constructed, debugged and put into operation. A photogarph of the system is shown in **Figure 25**.

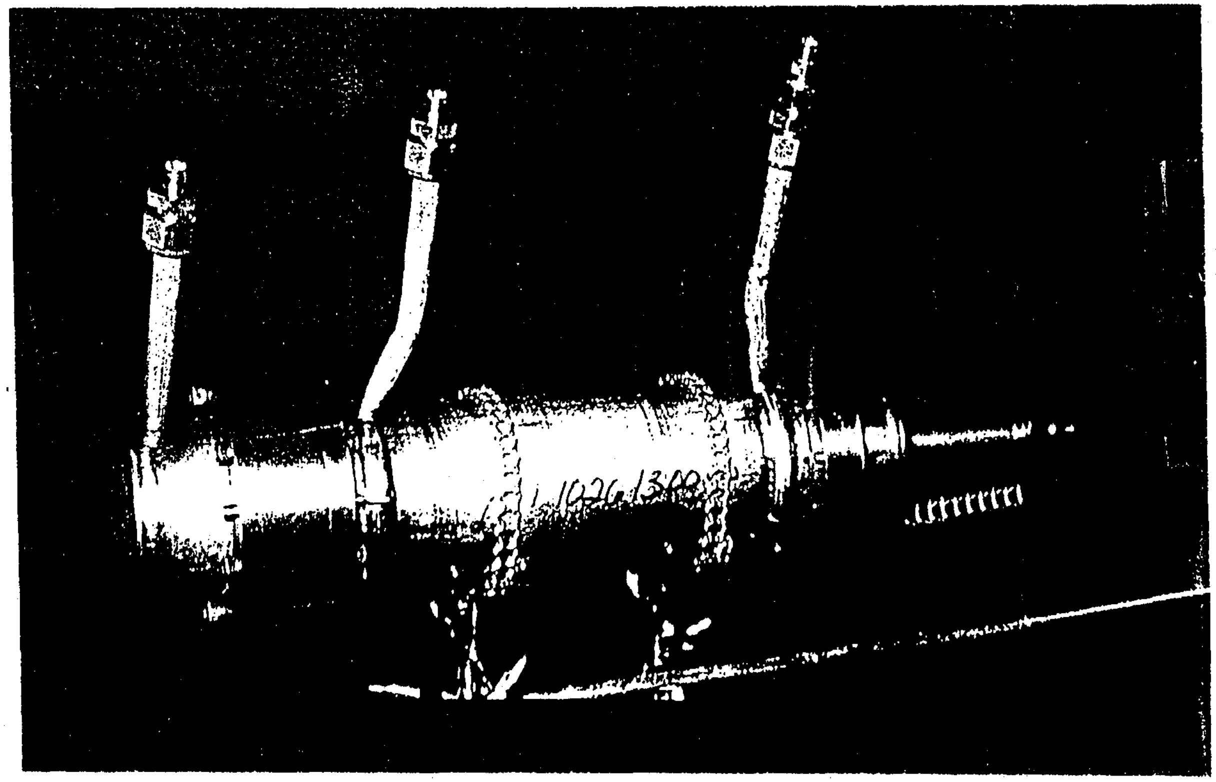
3.8 Fabrication of Tubes for Multi-Tube Module

Tubes were coated with the SSF membrane as described in section 3.3. Each tube was tested with pure and mixed gases. Some of the membrane characteristics from these runs were summarized in **Tables 12 and 13**. Some results from mixed gas testing are shown in **Figures 26-27**. The data for these two sets show that :

(i) About 85% of the tubes prepared meet the recovery-rejection criteria as shown by the dashed line in **Figure 26.**

(ii) There is some variability in the tube A/Fs; some of the tubes have a high A/F and hence may not meet the overall membrane performance criteria of acceptable separation and permeability properties. The reasons for these variations are not clear, but could be related to small changes in the membrane or support pore size from tube to tube.

(a)



(b)

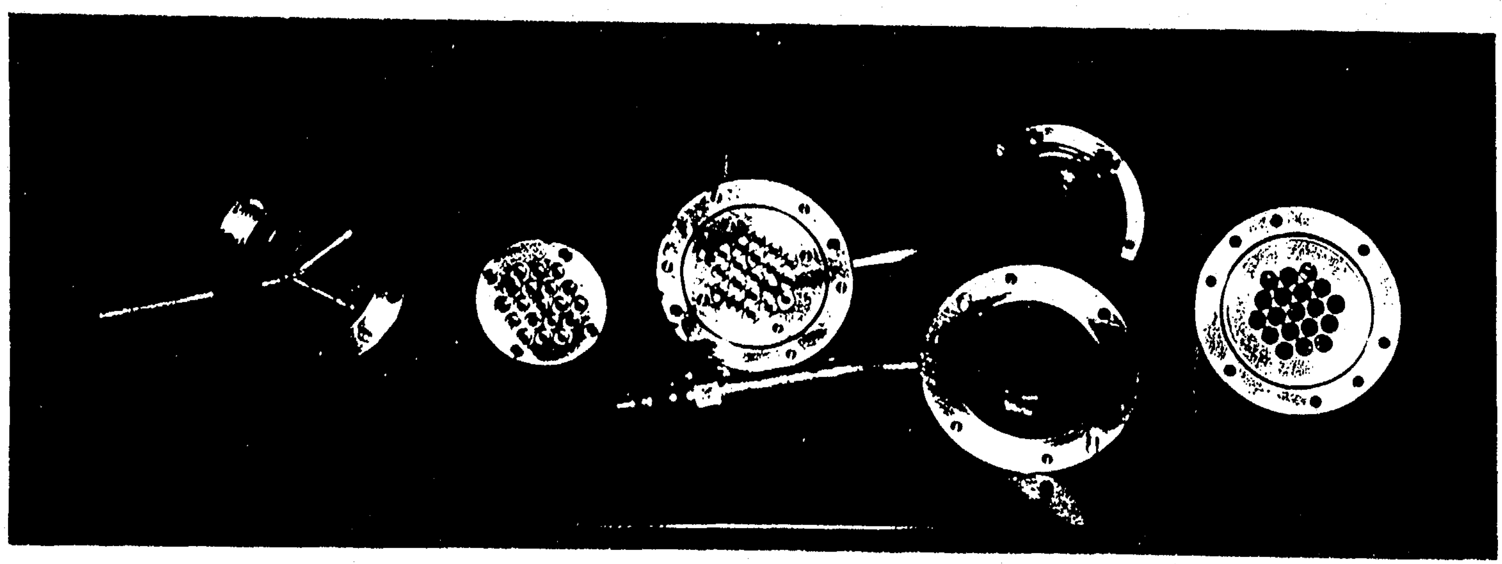


Figure 23. Multi-Tube Module Shell (a) and Interior Parts (b)

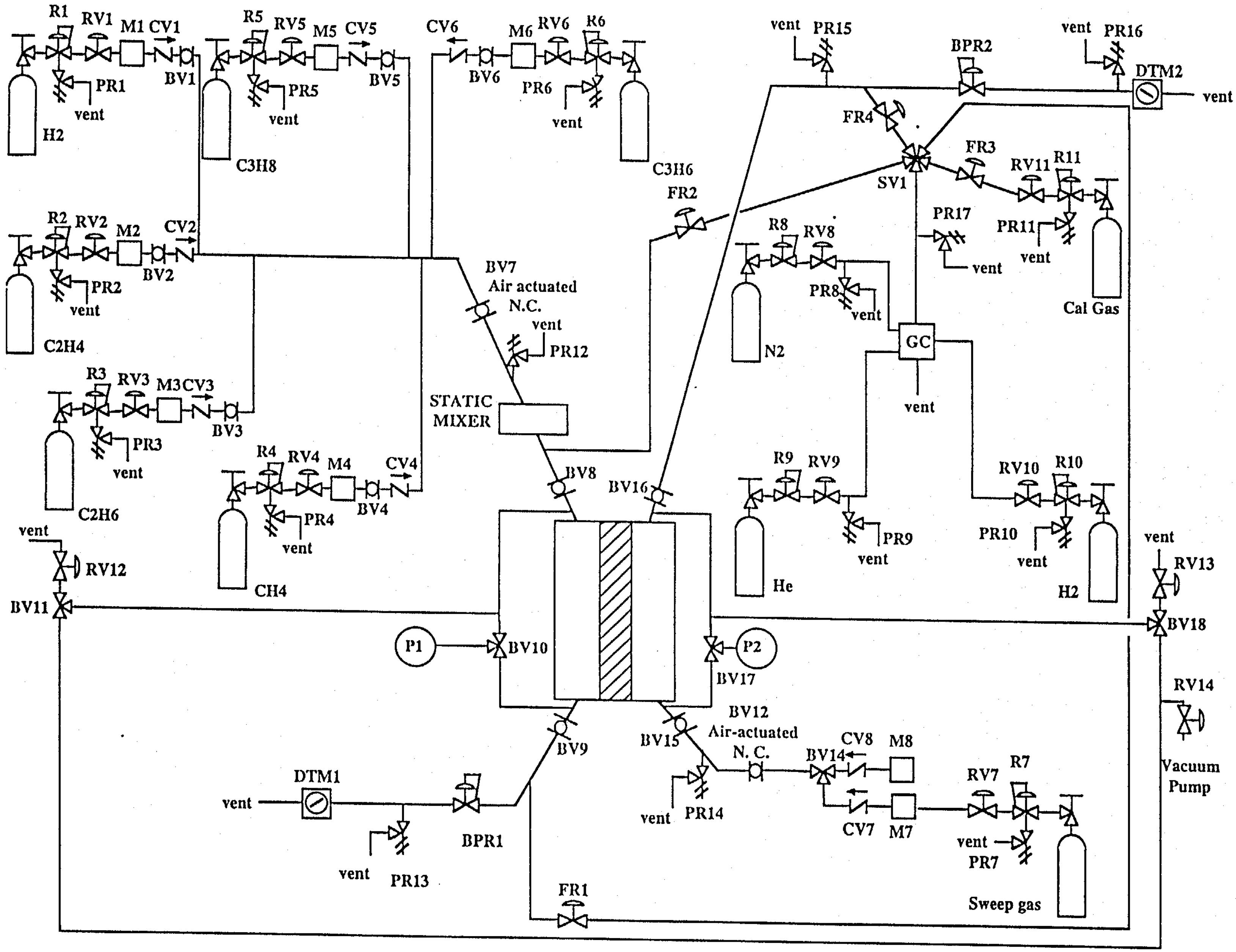
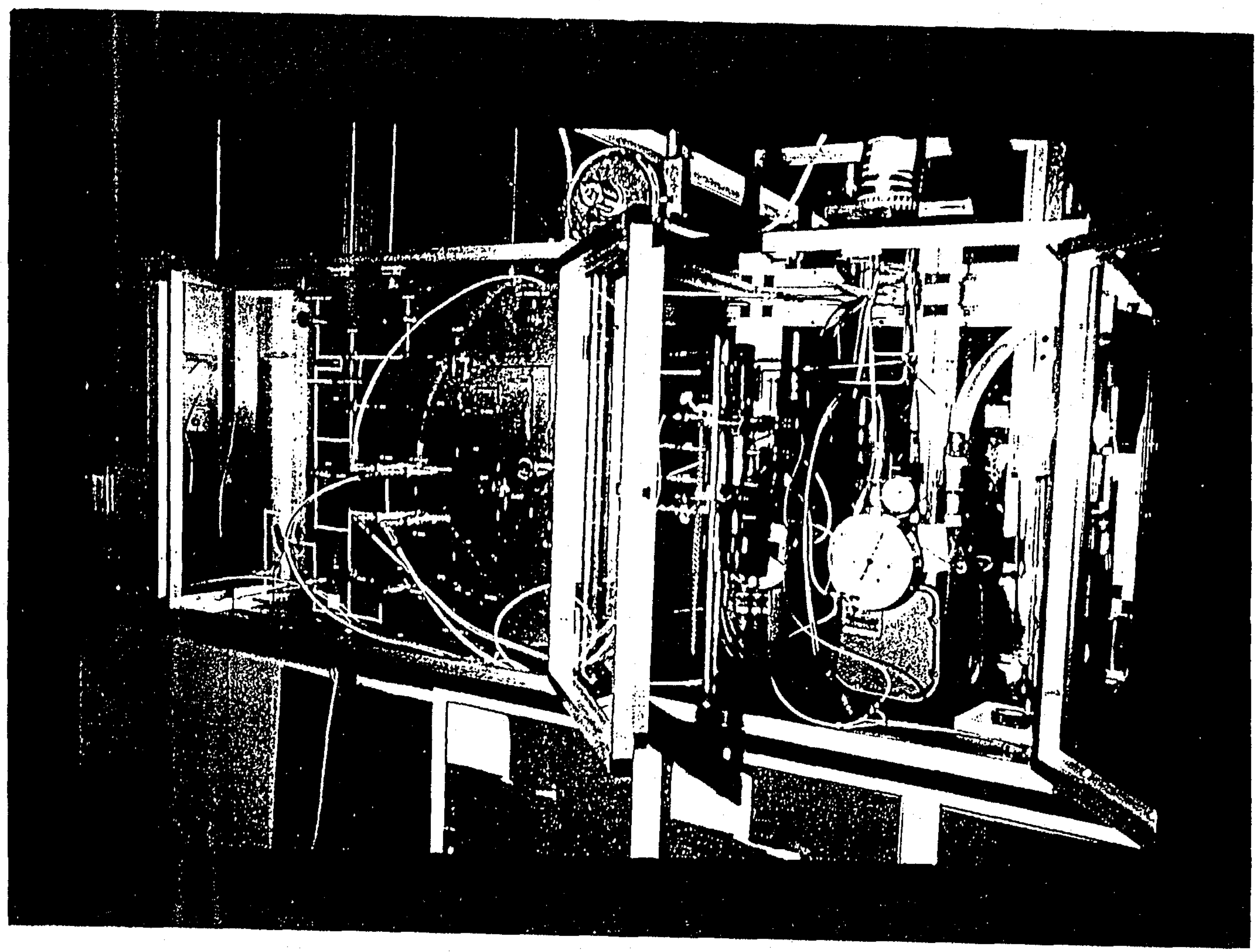


Figure 24. PID for Membrane Test Apparatus

Figure 25. Photograph of Membrane Test Apparatus



PHASE1.XLS Chart 9

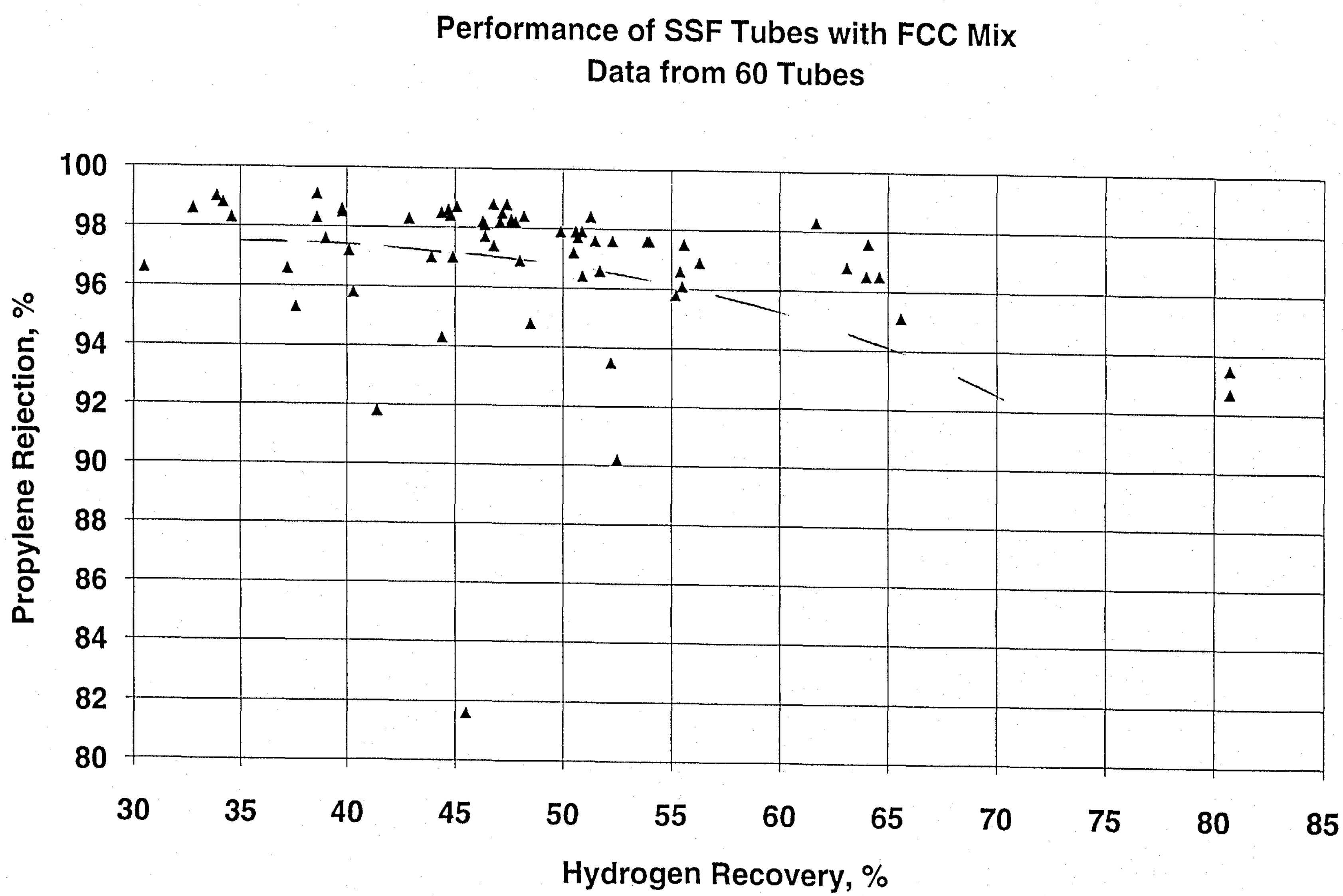


Figure 26. Performance Data from 60 Tubes : H_2 Recovery vs Propylene Rejection

PHASE1.XLS Chart 10

Performance of SSF Membranes with FCC Mix Data from 60 Tubes

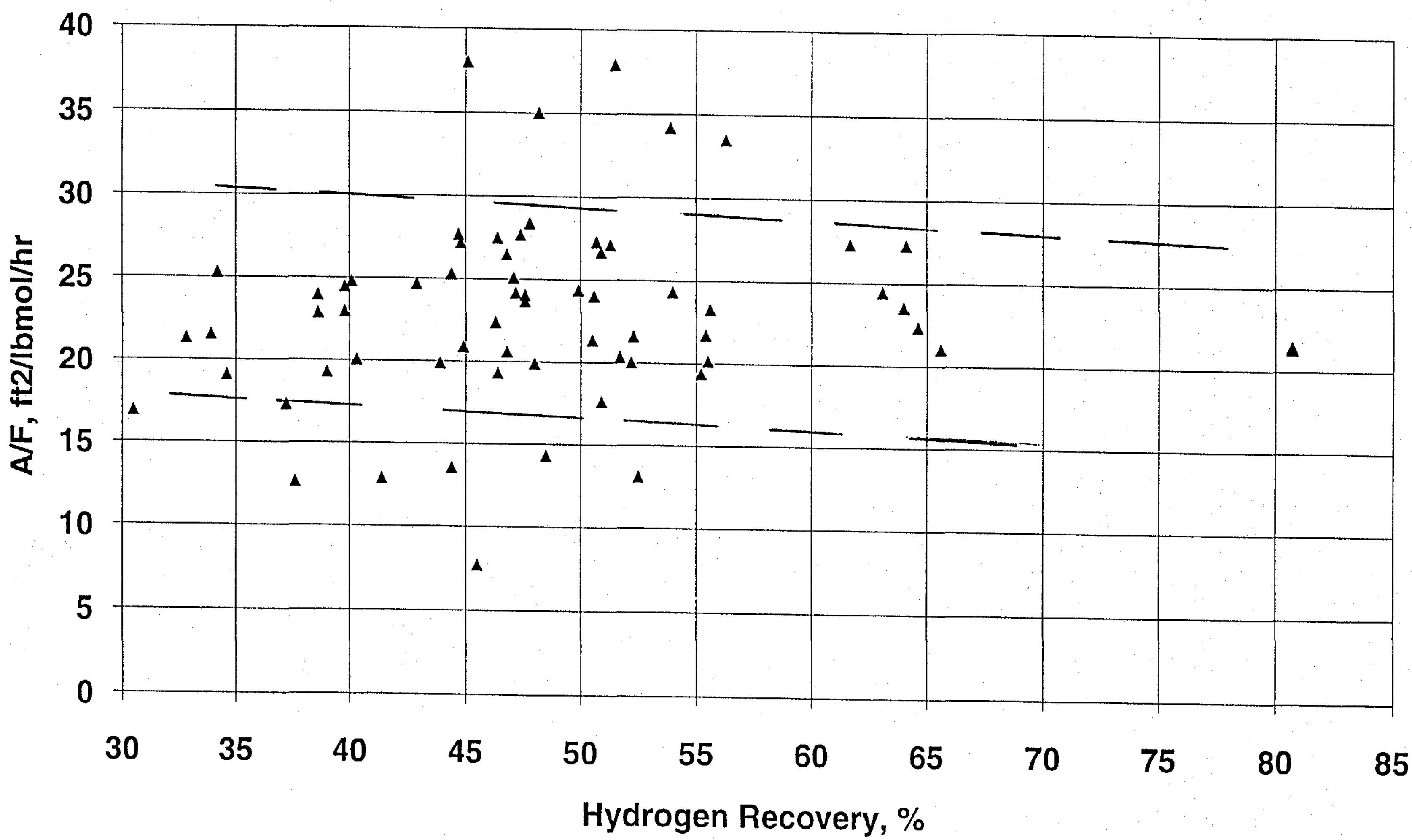


Figure 27. Performance Data from 60 Tubes : H_2 Recovery vs A/F