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DEVELOPMENT OF CHARCOAL SORBENTS FOR HELIUM CRYOPUMPING

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by
D. W. Sedgley & A. G. Tobin
Grumman Aerospace Corporation
Bethpage, NY 11714

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1 - SUMMARY

Improved methods for cryopumping helium were developed for application to fusion reactors where high helium generation rates are expected. This study period evaluated charcoal particle size, bonding agent type and thickness, and substrate thickness. The optimum combination of charcoal, bond, and substrate was used to form a scaled-up panel for evaluation in the Tritium Systems Test Assembly (TSTA) at Los Alamos.

The optimum combination is a 12x30 mesh coconut charcoal attached to a 0.48 cm thick copper substrate by a 0.015 cm thick silver phosphorus copper braze. The scaled-up panel was attached to a TSTA compound pump dewar and tested for helium pumping performance in an LN-shielded chamber. Based on the tests, the projected helium pumping performance for the optimum charcoal panel, assembled as a double chevron compound pump, is 4.9 liter $sec^{-1} cm^{-2}$.

A copper cement bond for attaching charcoal to a substrate was identified and tested. Helium pumping performance of this combination was comparable to that of the charcoal braze system. Evaluation of the cement is less mature than that of the braze, but the cement offers manufacturing flexibility that makes further study worthwhile.

Environmental tests showed the charcoal's susceptibility to vacuum chamber contamination. Performance degradation followed exposure of ambient temperature charcoal to a vacuum for prolonged periods. Maintaining a liquid nitrogen-cooled shield between the charcoal and the source of contamination prevented this degradation. A combination of bake-out and LN shielding effected recovery of degraded performance.

2 - INTRODUCTION

Charcoal cryopump systems for fusion reactor applications were developed in this second year effort to produce high performance fusion compatible (inorganic) units for vacuum pumping helium. The first year effort (Ref 1), concentrated on evaluation of charcoal type, size and distribution, methods of charcoal attachment, and utility of inorganic bonds for charcoal retention. The results showed that a fine (12x30 mesh), coconut PCB charcoal attached to a copper substrate with either a silver copper braze or a copper cement performed equal to or better than the same charcoal attached by organic epoxy. Epoxy bonding of charcoal is commonly used for commercial cryopump applications, but is not considered compatible for fusion reactor use.

This program continued the evaluation of compositions for charcoal retention, particularly of bonds made of brazes and low temperature inorganic cement. Brazes which might enhance wetting of the charcoal, including the successful Sil-Fos* (Silver Copper Phosphorus) braze bond from the first year program, and copper cement were prepared and tested for their ease of fabrication, durability, and performance in substrate bond charcoal assemblies.

The first year test results showed that helium pumping performance improved with decreased charcoal particle size. Because charcoal particle sizes smaller than those tested are commercially available, this program included evaluation of a PCB coconut charcoal finer than the best performing 12x30 mesh charcoal.

The current program concluded with delivery of a scaled-up charcoal pumping panel for installation in a compound pump from the vacuum system of the Los Alamos Tritium Systems Test Assembly (Ref 2). The panel, whose design was based on the results of the charcoal bond evaluation, was installed onto the compound pump's helium dewar for pre-test before shipment to Los Alamos.

The charcoal bond samples and the scaled-up panel were prepared and analyzed at Grumman. The performance test portion of the program was conducted at the Vacuum Technology Laboratory of Lawrence Livermore National Laboratory (LLNL). The expert support of W.R. Call of LLNL, T.H. Batzer, consultant to LLNL, and W.J. Poit of Grumman is gratefully acknowledged.

**Product of Handy & Harmon, NY*

3 - OBJECTIVE

The overall program objective was to evaluate the helium cryosorption capability of charcoal without using organic bonds for attaching charcoal to a metal substrate. Last year's program (Ref 1), showed that high helium pumping performance is attainable with charcoal bonded to a liquid helium-cooled substrate using a braze or a copper cement. Although these bonds were shown to be superior to other methods of attachment, they were not verified for reproducibility, durability or capability when scaled-up.

This program concentrated primarily on the characterization of brazes and cements for charcoal bonding. Samples which warranted testing were evaluated at LLNL for thermal cycle durability and helium pumping performance. The bonding agent which performed the best in a substrate bond charcoal sample was incorporated into a 16 in. (40.5 cm) diameter panel for use in a TSTA compound cryopump.

Specific program objectives were:

- Preparation of braze and cement bonded charcoal samples under controlled conditions
- Thermal cycle and performance testing of samples
- Preparation of a scaled-up charcoal bond panel for installation and testing in a TSTA compound cyropump
- Preliminary assessment of charcoal characteristics which could account for the varying performance among charcoals from different sources
- Evaluation of finer coconut charcoal grades for pumping helium.

4 - EVALUATION OF BONDING AGENTS

Prior research (Ref 1), concluded that the thermal conductivity of the bonding agent between the charcoal and the substrate exerted a strong influence on the helium pumping performance of the charcoal. In particular, a given charcoal used with bonding agents exhibiting high thermal conductivity appeared to yield the highest pumping performance. On this basis, several braze alloys and a copper cement were evaluated for compatibility with both the charcoal and the copper substrate.

These compositions are listed in Table 1 along with two samples for particle size evaluation as described in Subsection 4.3. The selection of Sil-Fos was based on the "self fluxing" action of the phosphorus additive while the selection of the other braze compositions was based on the Mn additive, a strong carbide former that might enhance "wetting" of the charcoal by the braze alloy.

Table 4-1 Braze Alloy Compositions Evaluated for Compatibility with Charcoal

COMMERCIAL DESIGNATION	COMPOSITION, WT %			SOLIDUS (°C)	LIQUIDUS (°C)
	Ag	Cu	OTHER		
SIL-FOS	15	80	5 P	640	705
BRAZE 495	49	16	23 Zn, 7.5 Mn, 4.5 Ni	625	705
BRAZE 852	85		15 Mn	960	970
HI-TEMP 095		52.5	9.5 Ni, 38 Mn	880	925
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To evaluate the various braze alloy compositions, a specimen of each braze alloy was placed on a 2x2x3/16 in. (5.1 cm x 5.1 cm x 0.48 cm) copper substrate and heated in air with a propane torch. In our previous work (Ref 1), we furnace-brazed to avoid oxidation of the braze alloy. However, the high braze temperature to which the charcoal was exposed in that process adversely affected the charcoal's pumping ability and furnace-brazing was discontinued. In the case of the brazes 495, 852 and 095, a fluxing agent was painted on the surfaces before application of the propane torch.

With the exception of Sil-Fos braze, it was observed that both braze alloy and copper substrate oxidized severely before melting. This condition led to poor braze flow and poor coverage of the substrate which proved to be an inadequate base for bonding the charcoal. It was therefore decided to eliminate these brazes from further consideration.

The Sil-Fos braze alloy melted readily on the copper substrate and yielded complete coverage. It was thereby possible to sprinkle the charcoal granules on the liquid Sil-Fos braze during the heating operation and to effect their retention during the cool-down phase. This was accomplished by removing the torch from the workpiece and allowing air cooling by natural convection and conduction. The procedure usually required about 20 to 30 minutes.

Because approximately 95-98% of the effective surface area was covered with charcoal granules, the Sil-Fos braze was identified as the baseline braze bonding agent for all subsequent evaluations.

Evaluation of inorganic bonding agents centered on a copper-based cement which cures at approximately 120 C (395 K) in air and can be applied by brushing it onto the substrate. It was identified as the baseline material for low temperature curing bonding agents. This bonding agent was previously identified (Ref 1).

The samples prepared for testing are listed in Table 2 and described in the following subsections.

Table 4-2 Description of Charcoal Samples

SAMPLE #	SUBSTRATE	CHARCOAL TYPE PCB SIEVE #	BOND TYPE	NUMBER OF TEST RUNS
100	AL ALLOY	< 325	EPOXY	0
101	AL ALLOY	30 x 140	EPOXY	3
102	1/8" COPPER	12 x 30	SIL-FOS 3 MIL	3
103	1/8" COPPER	12 x 30	SIL-FOS 6 MIL	3
104	1/8" COPPER	12 x 30	SIL-FOS 6 MIL	0
105	1/8" COPPER	12 x 30	SIL-FOS 9 MIL	2
106	1/8" COPPER	12 x 30	SIL-FOS 9 MIL	0
107	1/8" COPPER	12 x 30	COPPER CEMENT DILUTED	0
108	1/8" COPPER	12 x 30	COPPER CEMENT UNDILUTED	0
109	1/8" COPPER	12 x 30	COPPER CEMENT UNDILUTED	3
110	1/16" COPPER	12 x 30	SIL-FOS 6 MIL IMPROVED MFG BY USE OF DAM	5
111	1/16" COPPER	12 x 30	COPPER CEMENT SLIGHTLY DILUTED	4

4.1 OPTIMIZATION OF SIL-FOS BRAZE BOND

To identify the effects of bondline thickness on charcoal helium pumping performance, 4 in. (10.2 cm) diameter copper substrates were prepared with three different thicknesses of braze bond layer (see Table 2). Since the braze alloy was provided in sheet form as 0.003 in. x 1.0 in. (0.008 cm x 2.54 cm) strips, specimens were prepared using one, two and three layers of braze alloy to yield bondline thicknesses of 3, 6 and 9 mils (0.008 cm, 0.015 cm and 0.023 cm).

Thermal cycle and helium pumping tests were performed with the result that the charcoal with the 6 mil (0.015 cm) bond layer yielded the best "overall" pumping performance, although the 3 mil (0.008 cm) bond layer had behaved comparably. These tests are described in Sections 6-9. A significant reduction in pumping performance was observed with the 9 mil (0.23 cm) braze layer. This behavior appeared to be caused by the shearing off of charcoal in the thickest bond layer during cool-down with the concomitant loss of charcoal on the surface.

A final sample (No. 110), was prepared which used the 6 mil (0.015 cm) braze to retain the charcoal, and which was made using the upgraded procedure described in Section 5. A retainer wall placed around the perimeter of the 4 in. (10.2 cm) diameter sample significantly improved braze and charcoal coverage. This sample produced the highest pumping performance of any braze-bonded charcoal to date.

4.2 OPTIMIZATION OF COPPER CEMENT BOND

Samples were prepared which used as-received copper cement (Nos. 108 and 109) and diluted copper cement, (No. 107), (see Table 2). Charcoal retention on sample No. 107 was poor and approximately 50% of the effective area was uncovered. One sample prepared with as-received cement (No. 109), indicated poorer performance than a previously produced specimen (Ref 1). A diluted copper cement was subsequently prepared and the resulting sample (No. 111), showed pumping performance which was slightly better than the best braze bonded sample. Based on these findings, it was decided to perform further investigations of copper cement bonded charcoal.

4.3 OPTIMIZATION OF CHARCOAL PARTICLE SIZE DISTRIBUTION

In the previous study (Ref 1), we showed that coconut charcoal particles exhibited helium pumping performance superior to that of other sources of activated charcoals. In addition, we showed that coconut charcoal in the finer size range (12x30 mesh), exhibited

superior helium pumping performance to larger sized coconut charcoal. These results suggest that even smaller sizes of coconut charcoal should exhibit better performance. To test this hypothesis, coconut charcoal granules in the size range 30x140 mesh and <325 mesh were bonded to a 4 in. (10.2 cm) diameter aluminum substrate using a silver-based epoxy adhesive. Pumping results are described in Section 9.

4.4 OPTIMIZATION OF COPPER SUBSTRATE THICKNESS

Three thicknesses of copper were evaluated for brazing compatibility: 0.125 in., 0.188 in. and 0.250 in. (0.32 cm, 0.48 cm and 0.64 cm). Three 4 in. (10.2 cm) diameter brazed specimens of each thickness were prepared using Sil-Fos braze and 12x30 coconut charcoal. Although all three substrate thicknesses showed excellent bonding of charcoal, significant differences were observed. The 0.125 in. (0.32 cm) thick copper substrate buckled slightly during the brazing operation, probably due to the large non-uniform thermal stresses induced in the plate during brazing and subsequent cool-down. The 0.188 in. (0.48 cm) and the 0.250 in. (0.64 cm) thick substrates showed little evidence of buckling. However, the 0.250 in. (0.64 cm) substrate required considerably greater heat input to melt the braze and bond the charcoal due to its larger mass. On this basis, the 0.188 in. (0.48 cm) thick copper substrate was used for scale-up procedures.

5 - SCALE-UP FOR TSTA OF OPTIMIZED BONDING AGENTS

Based on results obtained with the 4 in. (10.2 cm) diameter samples described in the previous section and in Ref 1, it was decided that the 12x30 mesh coconut (PCB) charcoal granules represented the optimum for helium cryopumping and the Sil-Fos braze alloy represented the optimum bonding agent for the charcoal. Further evaluation of copper cement is recommended for future effort. All substrates consisted of 3/16 in. (0.48 cm) thick copper plates. Because our objective was to produce a 16 in. (40.6 cm) diameter charcoal cryopanel for LLNL and TSTA, a stepwise increment in the effective area to be brazed was evaluated. Because a size limitation had not been observed during preparation of the 4 in. (10.2 cm) diameter panels, brazed specimens of 6 in. (15.2 cm) and 8 in. (20.3 cm) diameter were prepared. This increase represents up to a four-fold gain in the effective brazed area. Charcoal bonded specimens in these sizes were prepared using a 6 mil (0.015 cm) Sil-Fos braze alloy.

Although little difficulty was encountered in the preparation of a 6 in. (15.2 cm) diameter substrate, considerable difficulty was encountered in preparing the 8 in. (20.3 cm) diameter substrates. The braze alloy could not be maintained in the molten state across the entire area while charging the wetted surface with charcoal. This effect was due to the inability of the brazing torch to input sufficient heat to offset the large heat losses by radiation and conduction. Thus, while the braze at the center of the disc remained molten, the braze alloy at the edges of the disc remained unmelted.

It was concluded that a 6 in. (15.2 cm) diameter disc represented the largest size (i.e., 28 $in.^2$) that could effectively be brazed with conventional brazing equipment. Larger sizes could be made using specifically designed brazing equipment/special fixturing to reduce heat losses during brazing.

5.1 PREPARATION OF TSTA SPECIMEN

It was observed, while scaling up to a 28 $in.^2$ substrate, that a narrow (1/8 in. (0.32 cm) wide) rim region near the edge of the specimen was devoid of charcoal. The braze alloy layer was significantly thinner in this region because the braze tended to flow down the side of the disc. To compensate for this effect, a copper 30 mil (0.076 cm) sheet

metal retainer wall was constructed that fit tightly around the specimen. Although this retainer wall was effective in reducing the loss of the braze alloy, the Sil-Fos braze alloy reacted with the retainer wall and made the wall difficult to remove from the copper substrate. To reduce interaction with the braze alloy and the retainer wall, a preoxidized Kanthal A-1 strip was used as the wall material. When this alloy oxidizes, it forms an Al_2O_3 protective skin which was expected to resist wetting by the braze alloy. Figure 5-1 illustrates the Kanthal retainer wall. This approach eliminated any interaction with the braze alloy, and was effective in reducing the "edge effects" observed in the absence of the retainer wall.

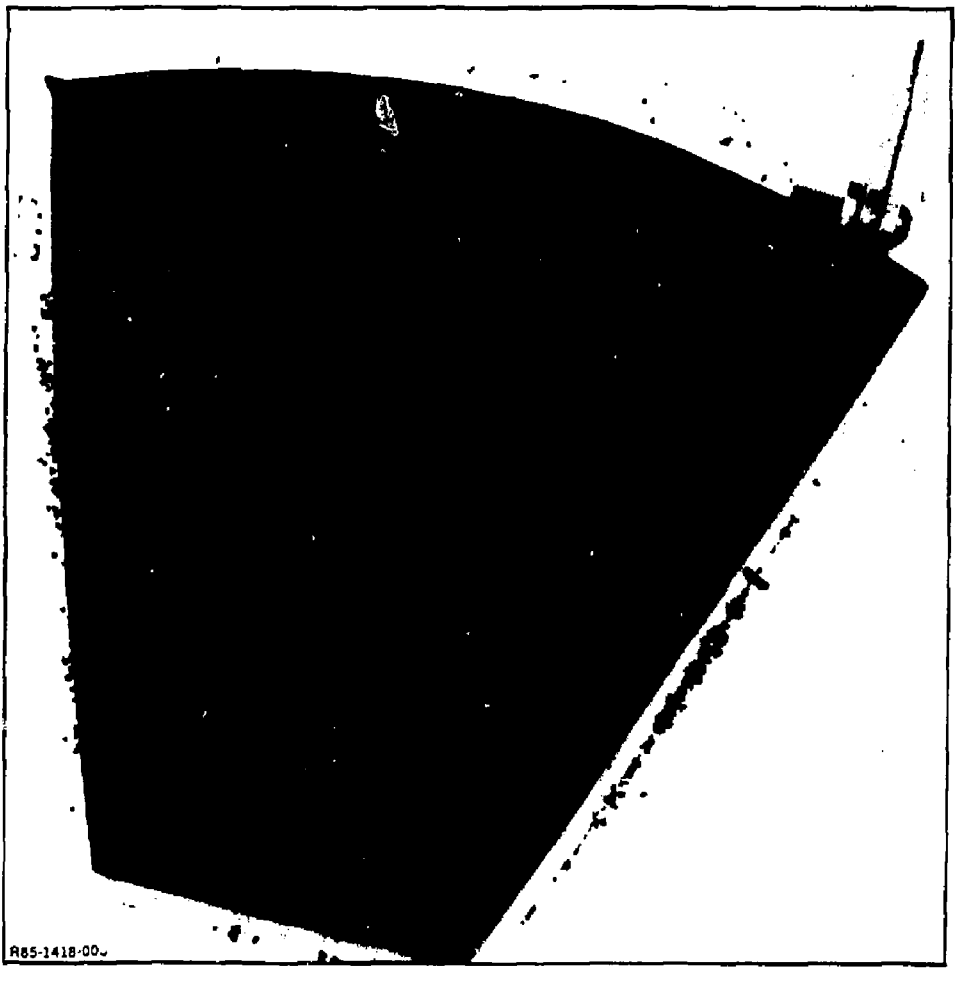


Fig. 5-1 Kanthal Retainer Shown in Place to Retain Charcoal During Brazing

5.2 DESIGN DEVELOPMENT OF 16 IN. DIAMETER TSTA TEST SPECIMEN

In order to produce a 16 in. (40.6 cm) diameter panel, it was decided to subdivide the area into a set of plates whose individual areas did not exceed the 28 in^2 (182 cm^2) limitation. Our initial layout featured a 6 in. (15.2 cm) diameter disc surrounded by eight annular plates with areas about equal to the central disc. This configuration, however, did not permit the construction of a retainer wall to confine the charcoal and braze alloy. A slight modification of this configuration was adopted in which a central octogonal piece was surrounded by eight truncated pieces fitting together to form the 16 in. (40.6 cm) diameter circle (see Fig. 5-2). Because machined plates required pre-drilling of attachment holes, it was necessary to fill the holes with a temporary filler to prevent the braze alloy from flow-

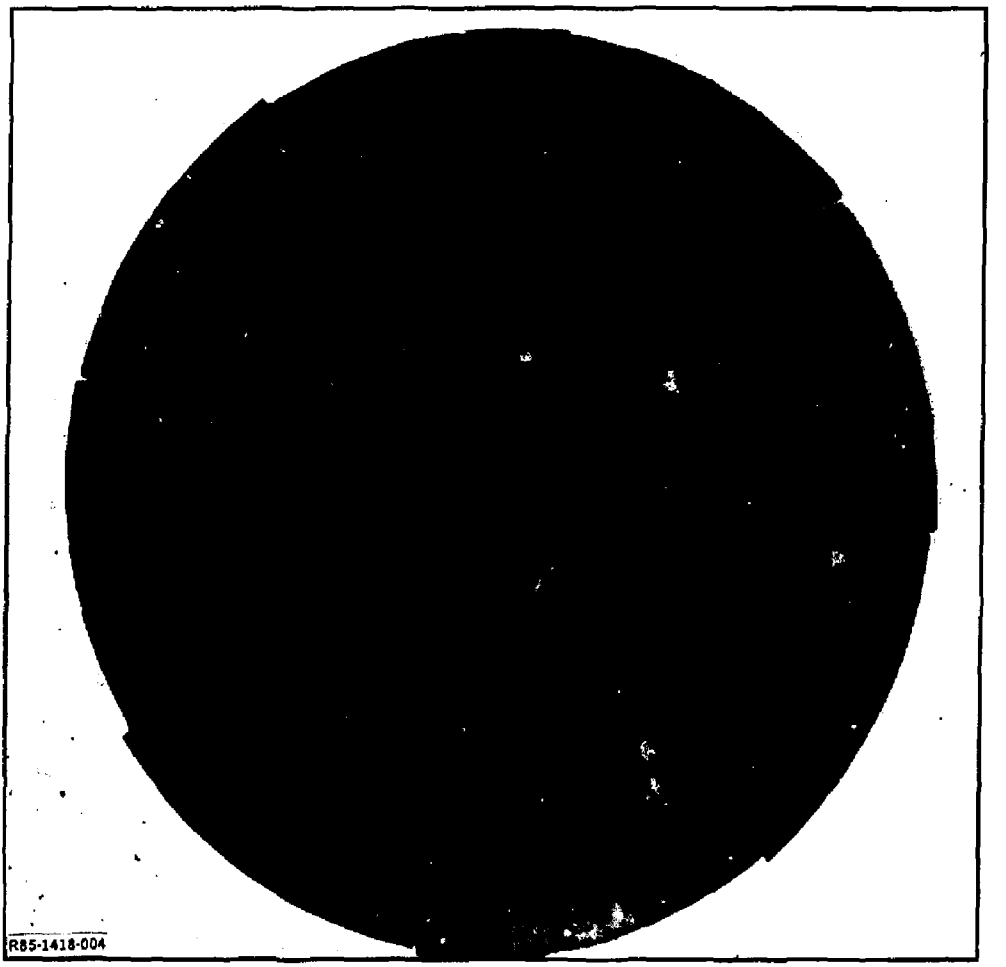


Fig. 5-2 Arrangement of Copper Plates for TSTA Pumping Panel

ing into the holes during the brazing operation. A BN (Carborundum Corp.) filler was identified in the form of a liquid coating that can be applied and cured at 50C (325K) in air. It was found that the BN filler was effective in preventing braze flow into the pre-drilled holes and was easily removed after the brazing operation.

Using the pre-formed Kanthal A-1 retainer, a 16 in. (40.6 cm) diameter charcoal cryo-pump panel (Fig. 5-3) was fabricated from a central octagonal piece plus eight annular pieces using 0.188 in. (0.48 cm) thick copper tiles, 12x30 mesh coconut charcoal granules and Sil-Fos braze alloy.

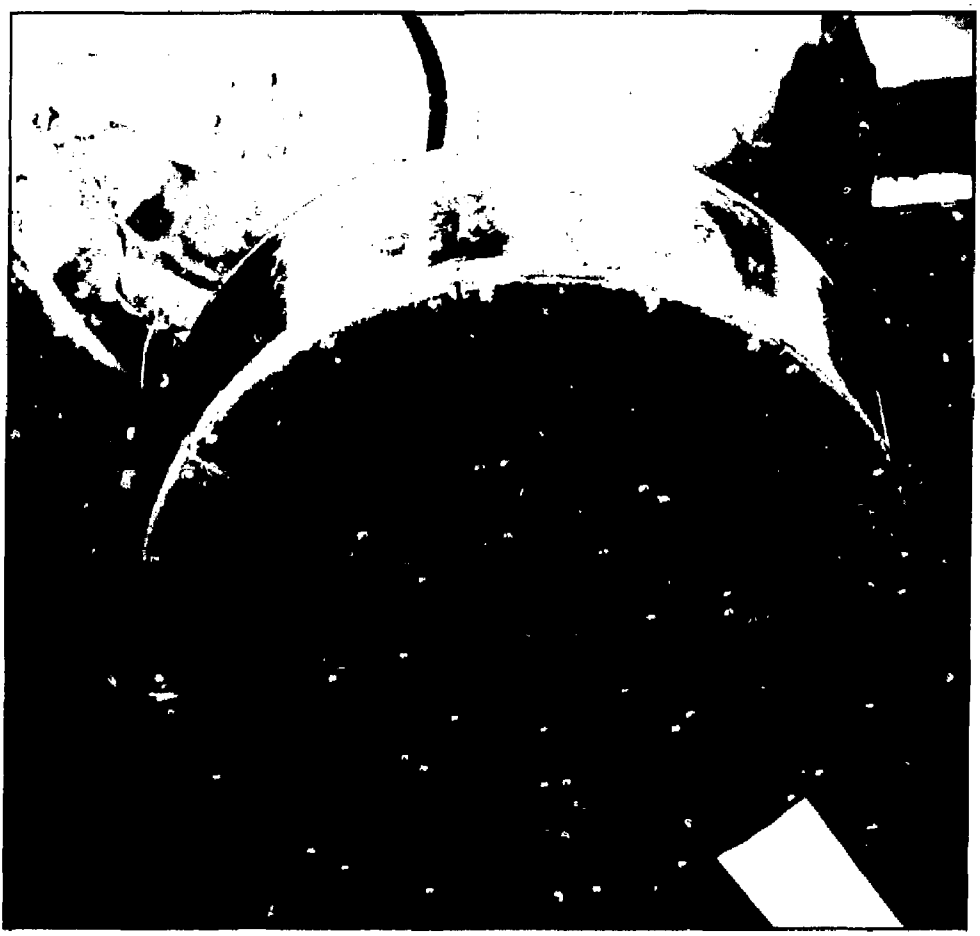


Fig. 9-4 Helium Pumping Performance of Coconut Charcoal Bonded with Undiluted Copper Cement

6 - TEST PROGRAM

The test program consisted of three parts. The first part continued an evaluation (see Ref 1), to determine the optimum charcoal particle size for pumping helium. The Ref 1 results had shown increased pumping capability with decreased particle size, so the first part of the test program evaluated finer particle sizes than those previously tested.

In the second part, charcoal test samples which featured either a braze or a copper cement to attach the charcoal to copper substrates, were subjected to performance and thermal cycle tests. Performance tests performed on the samples in their "as received" conditions were repeated following exposure to various environmental conditions.

The third part evaluated performance and integrity of the 16 in. (40.6 cm) diameter charcoal pumping panel installed on the helium dewar of the TSTA charcoal compound cryopump.

Performance comparisons — either between repeated test runs on a given sample or between runs on different samples—were based on measurement of specific pumping speed versus specific capacity for a constant helium throughput.

7 - TEST SET-UP

The vacuum test installation at the LLNL Vacuum Technology Laboratory, which was used for the 1983 program (Ref 1) was used for this program. The charcoal test articles were configured either as 4 in. (10.2 cm) diameter samples or as segments of the 16 in. (40.6 cm) diameter panel for the TSTA pump. The exposed size of the 10.2 cm samples was 3.5 in. (8.9 cm) diameter. The former were installed on a test pump appended to the main test tank, and the latter were mounted on the TSTA pump helium dewar installed in the main tank. The test equipment is described in the following subsections.

7.1 TEST APPARATUS

The system used in the study is illustrated in Fig. 7-1. The roots/mechanical system roughed the main chamber and the test pump, while a turbo-molecular pump provided final roughing to a pressure in the 10^{-7} range prior to introduction of liquid helium. Liquid nitrogen in the cryopump shield dewar or in the main tank shield contributed to the pump-down. In some tests, when the turbo-molecular pump was unavailable, the liquid nitrogen units performed final roughing. A shield in the main tank was installed for the TSTA panel tests only.

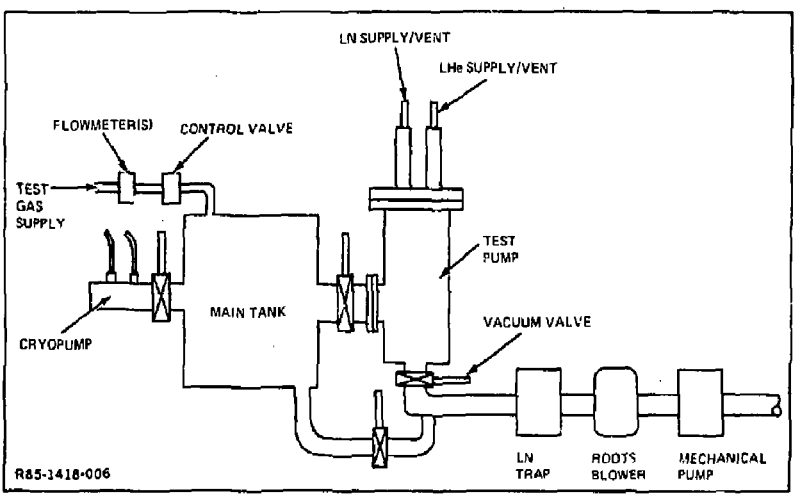


Fig. 7-1 Test System Schematic

The test gas helium fed into the main chamber was controlled by a servo-operated valve and shut-off valve. Flow for the 4 in. (10.2 cm) panel tests was held at a constant value of 6×10^{-4}, Torr liter s^{-1} cm^{-2}, within a range of ±4%. This value is representative of fusion reactor throughput rates. For the 16 in. (40.6 cm) panel tests the flow was 6×10^{-4} Torr liter s^{-1} cm^{-2}, or 3×10^{-4} Torr liter s^{-1} cm^{-2}.

7.2 TEST PUMP

The workhorse pump used in the Ref 1 tests was again the test bed for the 4 in. (10.2 cm) panel evaluations (Fig. 7-2 and 7-3). The pump's liquid helium reservoir which held the test panel was shielded by a four-sided chevron array. Access to the panel was through a demountable bottom plate which completed the shielding.

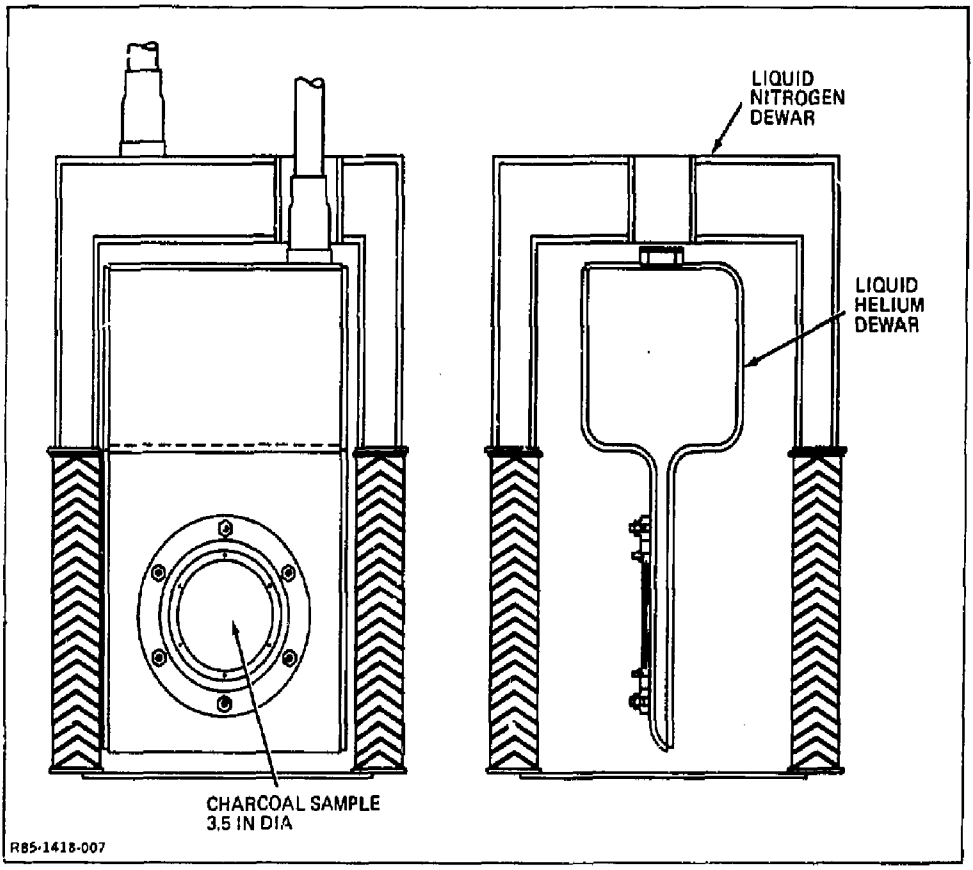


Fig. 7-2 Workhorse Cryopump for Charcoal Sorbent Tests

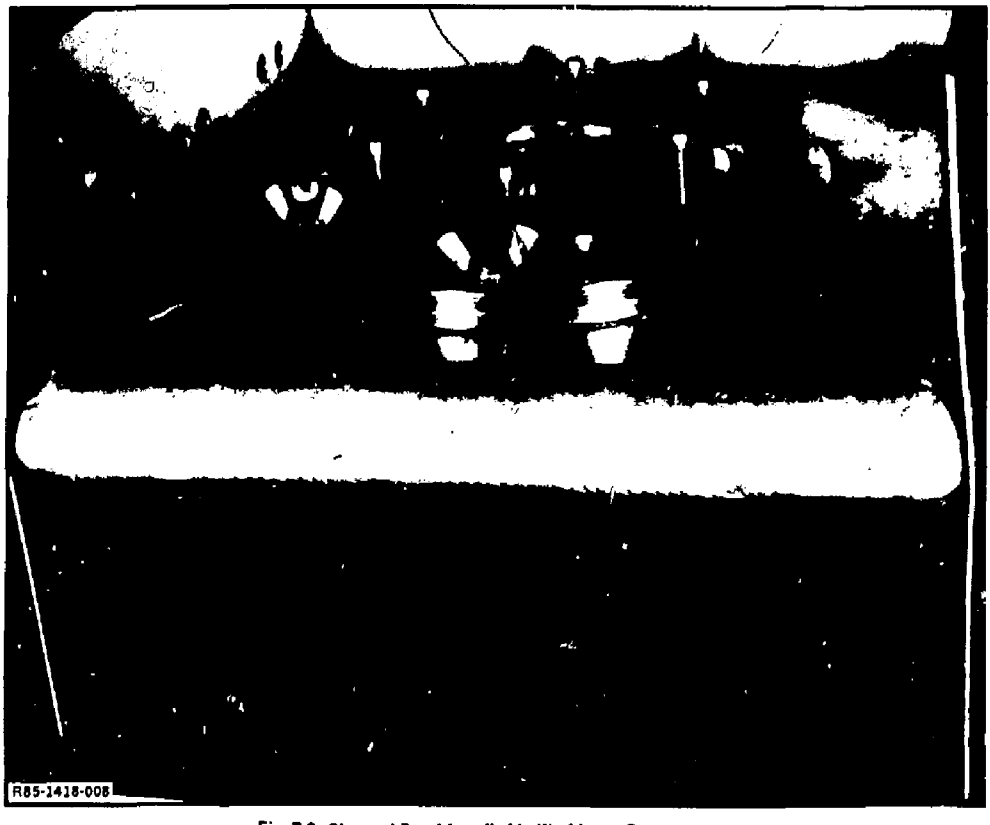


Fig 7-3 Charcoal Panel Installed in Workhorse Cryopump

7.3 TEST DEWAR

For the 16 in. (40.6 cm) panel tests, the panel was mounted on the liquid helium dewar of a TSTA compound cryopump (Ref 3). Los Alamos personnel had disassembled the cryopump, removed the existing pumping panel and shipped only the dewar to LLNL for preliminary evaluation of the charcoal panel. When the dewar is in the compound cryopump, it is surrounded by a liquid helium-cooled inner shield and chevron and a liquid nitrogen cooled outer shield and chevron (Fig. 7-4). In the test cell at LLNL, the helium dewar was suspended in the main test tank, surrounded by the liquid nitrogen-cooled shield mounted inside the tank wall (Fig. 7-5).

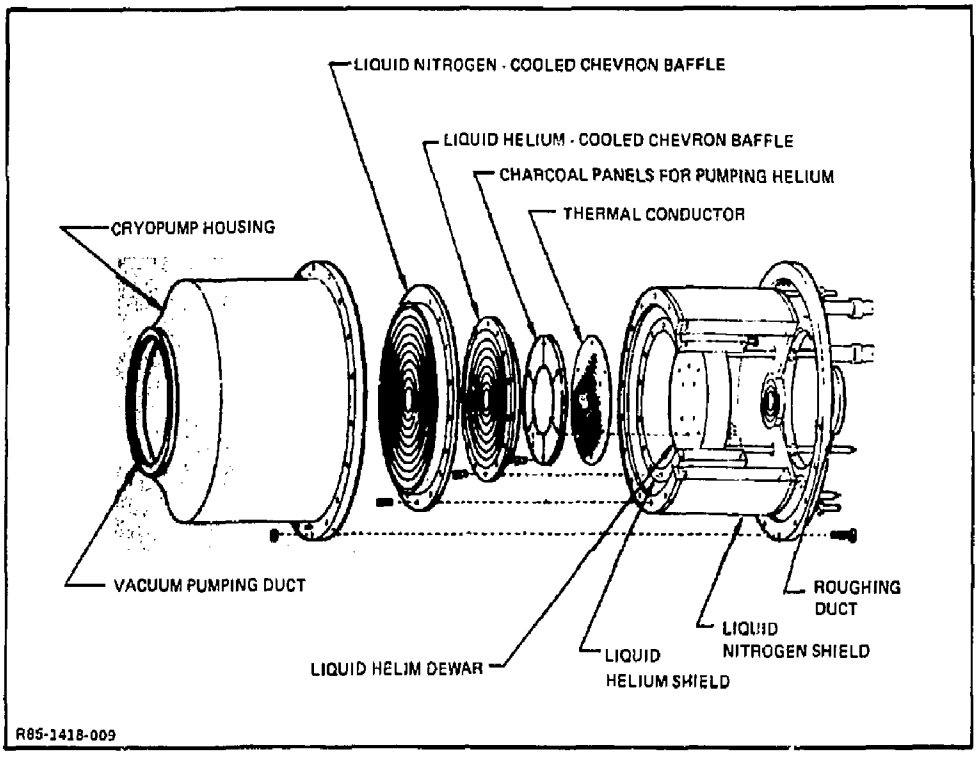


Fig. 7-4 Charcoal Compound Cryopump

7.4 TEST ARTICLE

Three types of test articles were used for the three parts of the test program cited in Subsection 7.3. For the evaluation of charcoal particle size, two samples were prepared. The samples were 4 in. (10.2 cm) diameter aluminum alloy plates with fine 30x140 mesh or <325 mesh charcoal attached by a thin layer of epoxy.

For the performance and thermal cycle tests, the charcoal (12x30 mesh) was attached to 4 in. (10.2 cm) diameter copper plates using either braze alloy or copper cement.

Each aluminum or copper plate was held by a retainer ring against the test pump's liquid helium dewar, with a copper mesh between the plate and the dewar to enhance thermal conductance.

For evaluation of the scaled-up 16 in. (40.6 cm) diameter charcoal test panel, the charcoal (12x30 mesh) was brazed to 3/16 in. (0.48 cm) thick copper plate. The test panel's nine pieces (Fig. 5-3) were mechanically attached to the helium dewar's flat lower surface, with a copper mesh between the panel and the dewar to effect good thermal contract.

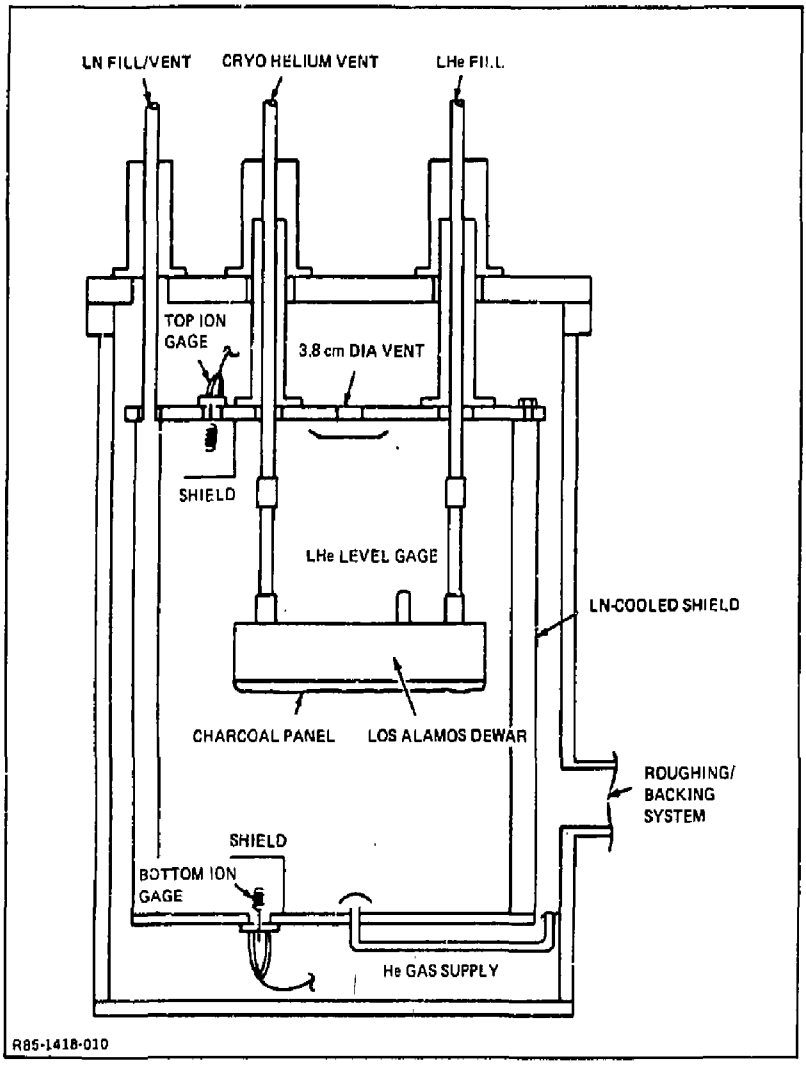


Fig 7-5 TSTA Dewar in the LLNL Vacuum Chamber

7.5 INSTRUMENTATION

Temperature, pressure and flow measuring instruments monitored the condition of the test articles and apparatus and provided data. Silicon diode sensors measured the temperature of the charcoal panels, the test pump helium dewar, the TSTA pump dewar, the test pump chevron and the main tank LN shield. The sensors were calibrated in liquid nitrogen or helium before the test program began. Nude ionization gages measured pressure in the main tank, at the bottom and top of the appended test pump container in the interspace out-

side the test pump's chevron baffles, and at the top and bottom of the volume formed by the LN shield in the main tank. The gages' relative sensitivity to helium was 0.143 based on manufacturer's data. A digital flow meter calibrated for air indicated gas flow into the test tank. The meter was calibrated in the laboratory for helium and the relative sensitivity for helium was 0.714.

8 - TEST METHOD

The test apparatus shown in Fig. 7-1 was used for the 4 in. (10.2 cm) diameter sample tests and for the 16 in. (40.6 cm) diameter panel test. Procedures are described for testing both types.

8.1 TESTING OF 4 IN. (10.2 CM) SAMPLES

The first sample was installed with the test tank up to air with the isolation valve between the tank and the appended test pump open. For subsequent installations, the main tank remained under vacuum. The LN-trapped roots/mechanical pump roughed the tank and pump. After ten minutes of roughing, the turbo-molecular pump isolation valve was opened and the roots/mechanical system was isolated. If test sample bakeout was specified, it was performed 20 minutes after roughing began by passing heated air for 20 minutes through the cryopump's helium dewar. At the end of bakeout (or when tank pressure indicated less than $5x10^{-5}$ Torr without bakeout), cooldown of the test pump's chevrons and shield with LN began. Liquid helium cooldown of the pump dewar began when the chevron temperature had dropped to 95°K, which occurred approximately 75 min after the start of chevron cooldown without bake, or 90 min with bake. At this time, tank pressure was on the order of $5x10^{-7}$ Torr.

Approximately one hour after the start of liquid helium transfer, the sample temperature stabilized and the helium test gas was fed into the main tank. The total elapsed time from the start of roughing until temperature stabilization was approximately 170 min without bakeout and 210 min with bakeout. Flow was increased until the steady state value was achieved, usually in less than two minutes when the accumulated helium on the sample was less than 0.07 Torr liter cm^{-2}.

Multiple test runs were made on some individual samples to evaluate the effect of prior conditions on the sample's performance. Therefore, the rough-down and cooldown sequences differed from the initial conditioning described above. These sequences are discussed as part of the test results (see Section 9).

Pressure (nitrogen equivalent), temperature and helium gas flow (air equivalent) were recorded at appropriate intervals during each test run. In addition, a strip chart recorded

pressure for run diagnostics. Pump and tank pressures were also recorded 5 to 10 minutes after the termination of helium gas flow at the end of a run.

8.2 TESTING OF 16 IN. (40.6 CM) DIAMETER PANEL

The TSTA pump dewar with the charcoal panel attached was installed in the liquid nitrogen shield. This assembly, was then suspended from the test apparatus cover, and placed in the main chamber (Fig. 7-5). The appended pump remained under vacuum throughout this test series but was not cooled down and did not function as a pump. The roots/mechanical system and turbo-molecular pump roughed the apparatus. The pump dewar was baked at 330°K for over one hour with the shield cooled, prior to introduction of the liquid helium. Data recording for this test was similar to that described above.

9 - TEST RESULTS

Pumping speed as a function of pumping capacity was determined for all samples that were tested. The test procedure for determining pumping speed was to use the constant flow method by introducing a fixed flow of helium gas into the main tank for the duration of a run. This flow was continued until a minimum of 20 min run time had elapsed. Because fusion reactor cryopumps will probably be regenerated after run periods of less than 20 min to minimize tritium inventory, data taken before 20 min elapsed time is of most interest.

Helium specific pumping speed S_S in liter s^{-1} cm^{-2} was determined from the relationship:

$$S_S = 2.61 \times 10^{-3} \times Qi/(PiA)$$

Here, A is the charcoal sorbent effective surface area in cm^2, Qi is the indicated helium flow in sccm (air), and Pi is the ionization gage pressure in Torr (indicated for nitrogen). The charcoal sorbent effective surface area for the 4 in. (10.2 cm) diameter panels was 9.3 $in.^2$ (60.1 cm^2), and for the 16 in. (40.6 cm) diameter panel, it was 189 $in.^2$ (1221 cm^2). The pressure Pi for the small sample tests was measured by the ionization gage at the top of the appended pump cavity in the volume between the pump housing and the pump's nitrogen cooled shield. The pressure Pi for the large panel tests was measured by the top ionization gage shown in Fig. 7-5. Helium specific capacity C_S in Torr liter cm^{-2} was determined from:

$$C_S = 1.09 \times V_i/A$$

where V_i is the cumulative volume of helium in scc (air).

9.1 EVALUATION OF CHARCOAL PARTICLE SIZE

A set of runs was included in this program to determine the performance of finer (30x140 mesh and < 325 mesh) PCB coconut charcoal than had been evaluated in the preceding program (Ref 1). The smallest mesh size charcoal (12x30) in that program had produced the highest pumping speed. The 30x140 mesh charcoal sample (No. 101), was tested in its as received condition (two runs) and in its condition following a 340K vacuum bake (one run).

All runs were terminated in less than 10 min, as pressure rose excessively with the $6x10^{-4}$ Torr liter s^{-1} cm^{-2} helium flow. The quantity of 30x140 mesh PCB charcoal per sq cm of panel surface area is insufficient to absorb the helium at the test flow rate. Consequently, the finer mesh (<325) charcoal (No. 100), was not tested. The best performing charcoal cited in Ref 1 (12x30 mesh PCB), is evidently the optimum charcoal particle size for helium cryopumping.

9.2 THERMAL CYCLE TESTS

These tests were performed to evaluate the integrity of charcoal/bond combinations after being subjected to temperature variations and to helium absorption and regeneration. The profiles were representative of conditions to be expected in operation. In general, the samples were cooled from ambient temperature to liquid helium temperatures as described in Section 8. Following a period of helium pumping (more than 20 min), the pump dewar was warmed to 85-90°K. The sample temperature reached 45-50 K. This warming operation effected sample regeneration; helium evolved quickly from the sample when the sample temperature was in the 18-25°K range. The sample was then recooled to the liquid helium temperature range and helium pumping was repeated. These helium pumping periods were performed three times on each sample except for one case in which poor performance would have made a third run unproductive.

Eight samples were prepared for the thermal cycle tests (see Table 2). Five used braze for attaching the charcoal to the copper substrate, comprising one 3 mil (0.008 cm) thick braze sample, two 6 mil (0.015 cm) braze samples, and two 9 mil (0.023 cm) braze samples. One sample of each braze thickness was tested. Three samples consisted of copper cement: two cements were applied in the as-received form and one was applied in a diluted form. One of the as-received cement samples was selected for test. The diluted cement failed to retain the charcoal; that sample was therefore discarded prior to testing.

All tested samples satisfactorily completed the thermal cycle tests. The charcoal remained bonded after the cycles. For tests in which the repeated pumping was done on a single day (Fig. 9-1 to 9-4), the pumping performance was consistent from one run to the next. The pumping performance, as indicated by the specific speed for helium, was considerably lower for the charcoal bonded by 9 mil (0.023 cm) thick braze (No. 105), than for the samples with thinner braze bonds (Nos. 102 and 103). For this reason, the third performance run on the 9 mil (0.023 cm) sample was omitted. The charcoal sample attached by undiluted copper cement (Fig. 9-4), yielded lower performance and the formulation was subsequently changed (sample No. 111), for later tests.

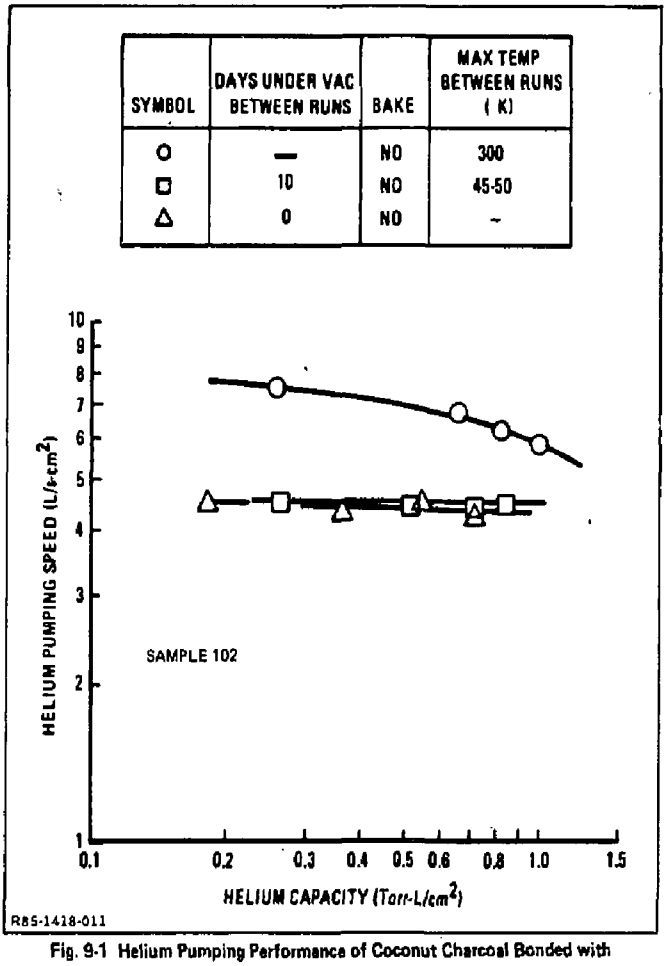


Fig. 9-1 Helium Pumping Performance of Coconut Charcoal Bonded with 3-Mil Sil-Fos Braze

For the sample having a 3 mil (0.008 cm) thick braze bond (No. 102) a ten day lapse occurred between the first and second runs. The second and third runs were made on one day. The pumping performance of these two runs was practically identical. However, it was measurably lower than that of the first run (see Fig. 9-1).

During the ten day lapse, the sample remained attached to the cryopump dewar in the test system. The liquid helium and nitrogen source was cut off and the sample gradually warmed to ambient temperature. The test system held under passive vacuum for the ten days before the roots/mechanical pumps were run and cryogens re-introduced for the second helium pumping run.

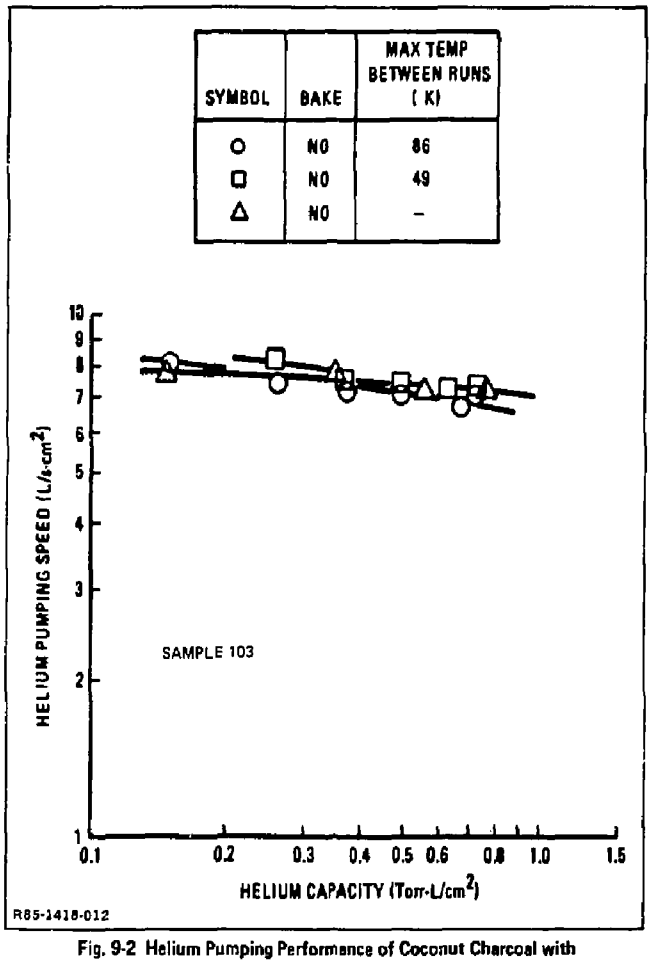


Fig. 9-2 Helium Pumping Performance of Coconut Charcoal with 6-Mil Sil-Fos Braze

It was observed that the charcoal panel's helium pumping performance was adversely affected by this exposure over a prolonged period to the vacuum chamber environment. As a result, an additional test series was included in the program to observe the effect of different conditions on the helium pumping performance.

9.3 ENVIRONMENTAL TESTS

Two samples were prepared for evaluation of pumping speed and capacity after exposure to conditions including warmup, exposure to air, bake-out and prolonged vacuum. Sample Nos. 110 and 111 were similarly configured to those evaluated in the Thermal Cycle

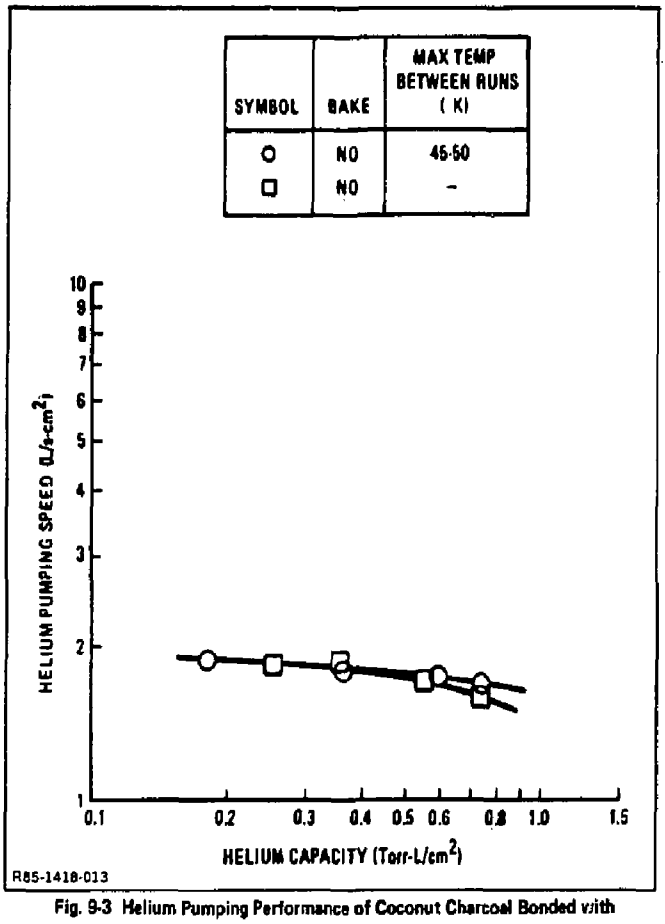


Fig. 9-3 Helium Pumping Performance of Coconut Charcoal Bonded with 9-Mil Sil-Fos Braze Layer

Tests, and comprised 12x30 mesh PCB coconut charcoal bonded to a copper substrate with either braze or copper cement. These samples were the result of improved fabrication techniques and had better charcoal retention than had been previously obtained.

Each sample in its as-received condition was installed in the vacuum system and cooled down in accordance with the previously outlined procedure. Helium throughput was nominally held at a 6×10^{-4} Torr liter s^{-1} cm^{-2} for all runs.

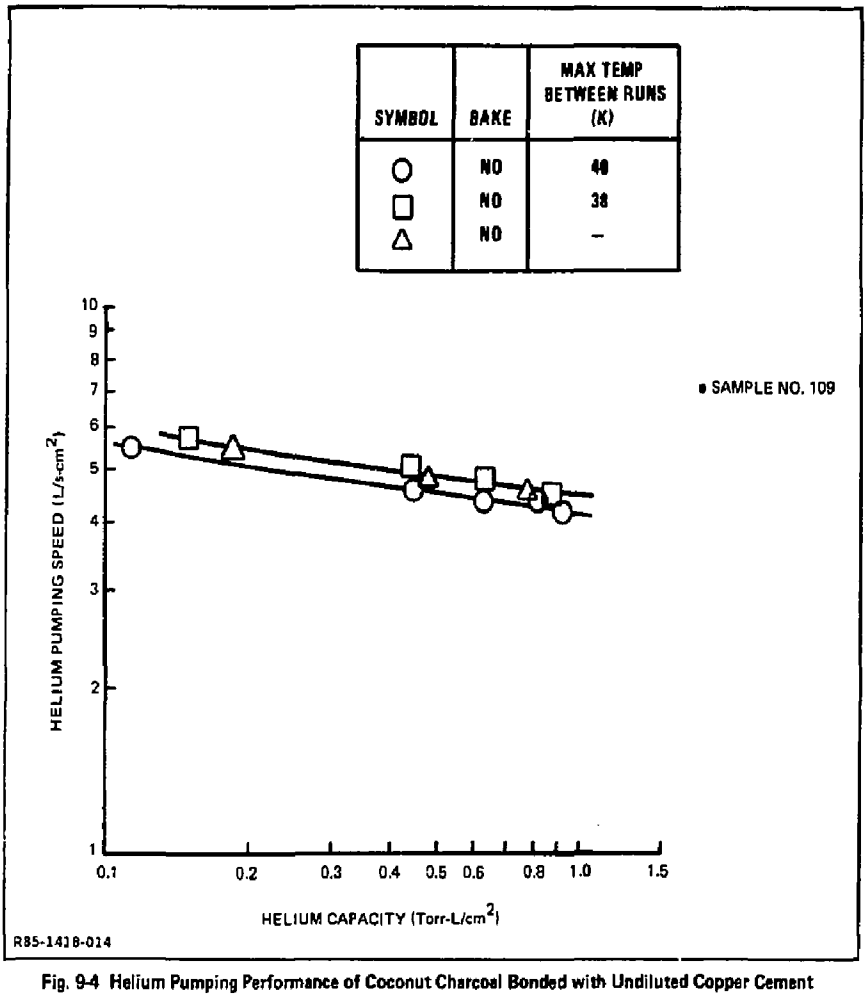
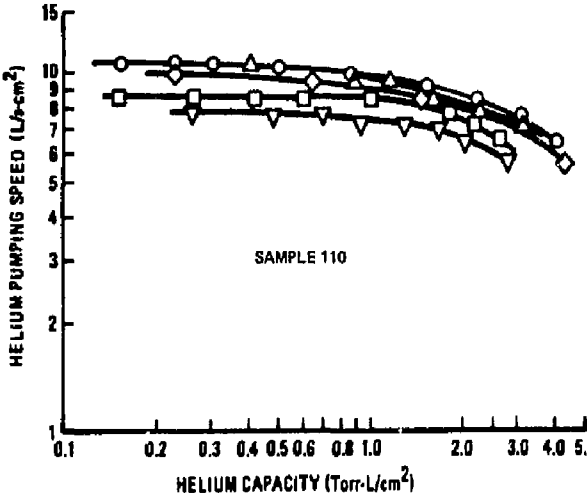


Fig. 9-4 Helium Pumping Performance of Coconut Charcoal Bonded with Undiluted Copper Cement

9.3.1 Charcoal Sample with Braze Bond (No. 110)

Five helium pumping runs were made with this sample. Figure 9-5 shows a measurable variation in helium pumping speed from over 10 liter s^{-1} cm^{-2} to under 8 liter s^{-1} cm^{-2}.

The best performance was obtained with the as-received sample tested in its unbaked condition. Specific pumping speed remained over 10 liters^{-1} cm^{-2} after more than 20 min of pumping at a pumping capacity of approximately 0.7 Torr liter cm^{-2} absorbed helium.



Exposure History of Charcoal Test Samples

RUN	AS RECVD	UP TO AIR BEFORE RUN	DAYS UNDER VAC BETWEEN RUNS	LN COOLED BETWEEN RUNS	LOW TEMP BAKE (330-340 K) BEFORE RUN
○	YES	YES	–	–	NO
□	NO	YES*	1	NO	YES
◇	NO	NO	1	YES	NO
△	NO	YES†	1	YES	YES
▽	NO	NO	3	NO	NO

†FOLLOWING VACUUM HOLD
**BEFORE VACUUM HOLD

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Fig. 9-5 Effect of Vacuum Exposure History on Helium Pumping Performance of Coconut Charcoal Bonded by 6-Mil Sil-Fos Braze

After the first run, the sample was warmed up, then held under vacuum overnight. The chamber was brought up to air for less than an hour before it was roughed down. The sample was vacuum baked at 330°K for 30 min before being re-cooled. Pumping performance during the second run showed a drop to under 9 l s^{-1} cm^{-2} helium pumping speed.

During the 16 hr period between the second and third runs, the cryopump's liquid helium dewar was emptied, but liquid nitrogen continued to cool the chevron shield. The helium dewar temperature did not exceed 95°K. Vacuum was continuously maintained and the sample was not baked before the third run. Pumping performance recovered to over 10 liter s^{-1} cm^{-2} at low capacity and was over 9 liter s^{-1} cm^{-2} after 20 min of steady state pumping.

The vacuum chamber was brought up to air following the third run, after which the vacuum was re-established and the pump chevrons cooled down for an overnight hold. The sample was vacuum baked at 330°K before liquid helium cooling. The high helium pumping performance of the first run was repeated in the fourth run.

Following the fourth run the cryogens were purged and the pump warmed up while vacuum was maintained in the system for three days. The pump was then cooled without sample bakeout and the ensuing run showed the poorest performance of all runs with a helium pumping speed of 8 liter s^{-1} cm^{-2} throughout the run. Although this performance was low compared to other results for this sample, it was considered a good performance compared to results obtained in Ref 1.

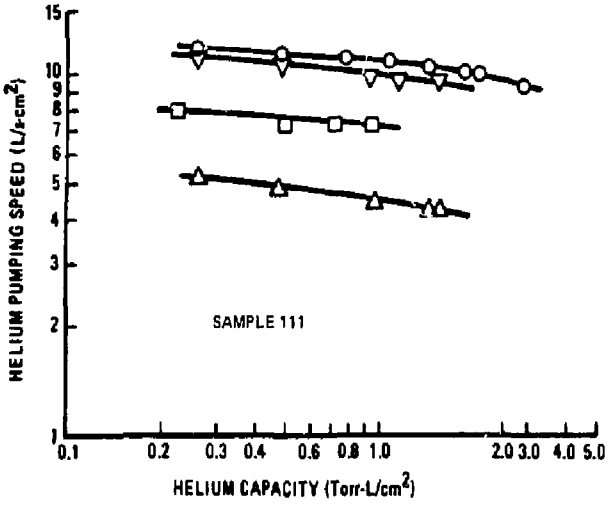
9.3.2 Charcoal Sample with Cement Bond (No. 111)

This sample was exposed to four helium pumping runs (Fig. 9-6). The first run after sample installation yielded the highest helium pumping speeds. The sample was kept under vacuum in the chamber for the 28 day duration of the tests.

The as received sample was vacuum baked at 330°K before cooldown. The measured helium pumping speeds were above 11 liter s^{-1} cm^{-2} until 20 min of steady state throughput had been absorbed by the charcoal.

The cryogens were purged out of the test pump after the first run, which warmed up and remained at ambient temperature under vacuum for five days before being baked at 330°K. Pumping performance in the second run, dropped below 7.5 liter s^{-1} cm^{-2} after 20 min of pumping.

The cryogens were purged and the pump allowed to warm up gradually to ambient temperature following the second run. The vacuum held for an additional 17 days before the sample was baked to 330°K and cooled down before the third run. The helium pumping



Exposure History of Charcoal Test Sample

RUN	AS RECVD	UP TO AIR BEFORE RUN	DAYS UNDER VAC BETWEEN RUNS	LN COOLED BETWEEN RUNS	LOW TEMP BAKE (330-340 K) BEFORE RUN	HIGH TEMP BAKE (380-400 K) BEFORE RUN
⊙	YES	YES	–	NO	YES	NO
⊡	NO	NO	5	NO	YES	NO
△	NO	NO	17 (22 CUM)	NO	YES	NO
▽	NO	NO	6 (28 CUM)	YES**	NO	YES (42.5 HRS)

**LN COOLED DURING DURATION OF BAKE (42.5 HRS)

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Fig. 9-6 Effect of Vacuum Exposure History on Charcoal Pumping Performance of Copper Cement Bonded Charcoal

speed after 20 min at the same helium throughput rate was reduced to approximately 4.5 l s^{-1} cm^{-2}, a 60% decrease compared to the performance of the as received sample.

The pump and sample were again allowed to warm to ambient while under vacuum, and held under vacuum for an additional six days. At 42.5 hours before Run No. 4, chevron cooldown with liquid nitrogen and sample bakeout was started. The LN cooling was continuous until completion of the run. Bakeout temperatures were increased to between 380° and 400°K for a continuous 42.5 hr period. The pump dewar was cooled with cryogenic he-

lium for 90 min before the pump and sample stabilized. Helium pumping performance recovered to within 5% of the value attained by the as received sample in the first run. Pumping speed after 20 min of steady state helium throughput was 10.5 l s^{-1} cm^{-2}.

9.4 TSTA PANEL TESTS

This test series consisted of performance runs in which the liquid helium cooled, 16 in. (40.6 cm) diameter panel was exposed to constant helium throughputs of 0.74 and 0.37 Torr liter sec^{-1}. These rates correspond to specific throughputs of $6x10^{-4}$ and $3x10^{-4}$ Torr l sec^{-1} cm^{-2}. The higher value corresponds to the specific throughput maintained in the tests of the 4 in. (10.2 cm) plates.

The panel was only vacuum baked prior to the first run while the shield surrounding the panel was maintained at liquid nitrogen temperature as long as the panel was in vacuum. Between the first and second run, the dewar was purged of liquid helium, warmed to 60°K to regenerate the helium gas, and recooled. The dewar was purged, allowed to warm up along with the liquid nitrogen shield, and recooled. The procedure between the third and fourth performance runs was the same as for the first run. No performance degradation was observed using the exposure methods described above.

Figure 9-7 shows the pumping performance of the TSTA panel for the two throughputs. Performance was unaffected by throughput for the range of values tested. The data are based on the top gage reading within the thermal shield (Fig. 7-6), and are corrected for helium gage sensitivity, though not for thermomolecular effect.

The performance of the panel in a two-chevron compound (TSTA) cryopump was estimated by correcting for the temperature effect as follows:

$$S = tS_{wo}\ To/T1$$

where S is the projected helium pumping speed of the two chevron pump; S_{wo} if the speed determined in these tests for the panel without baffle; To is the temperature of the incoming helium; T1 is the temperature in the cold space inside the shield; and t is the chevron transmissivity. Using a transmissivity based on Ref 4 of 0.15 for two chevrons in series, with T1 = 90°K for the cold space temperature, the estimated helium pumping performance for the compound pump is 4.9 l sec^{-1} cm^{-2}.

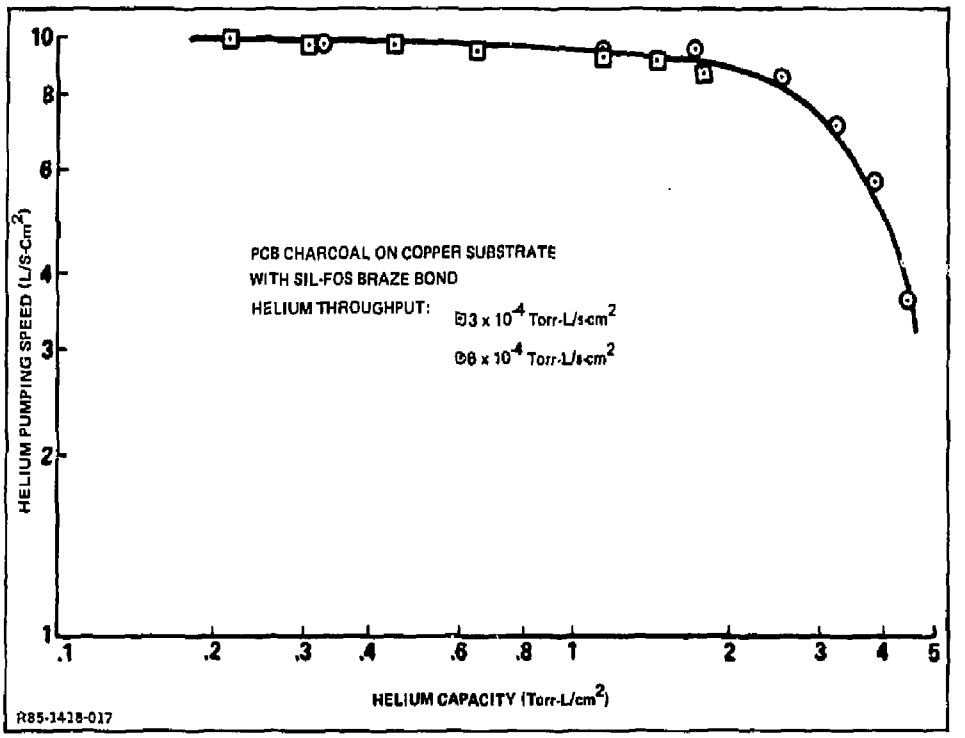


Fig. 9-7 Helium Pumping Performance of Scaled-up 40-cm Dia Charcoal Cryopump Bonded with 6-Mil Sil-Fos Braze

Examination of the apparatus after removal from the test chamber showed that some charcoal had dropped from the down facing panel. This condition is not expected to affect the panel's subsequent performance, as only the charcoal that was firmly attached to the bond and substrate contributed to the pumping capability.

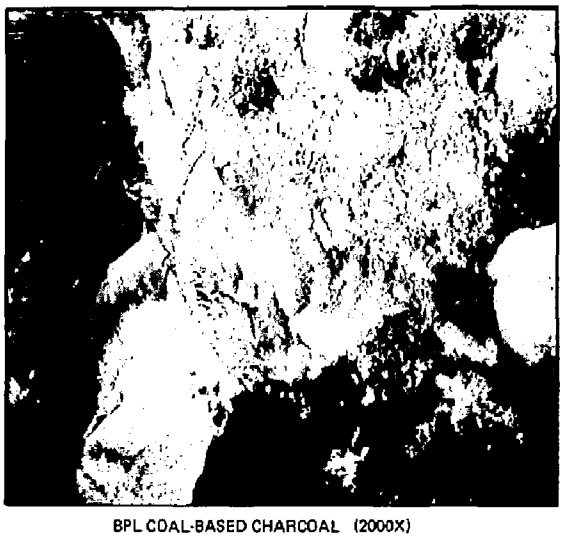
10 - CHARACTERIZATION OF CHARCOAL

In our prior work on charcoal characterization (Ref 1), it was shown that the PCB coconut charcoal exhibited superior helium pumping performance compared to the coal based charcoals BPL and Desorex. These differences are not explainable in terms of the atomic pore sizes of microscopic surface areas of each charcoal. To obtain a preliminary understanding of this phenomenon, both X-ray diffraction and SEM techniques were used to identify possible differences in crystallinity and/or morphology of the charcoals.

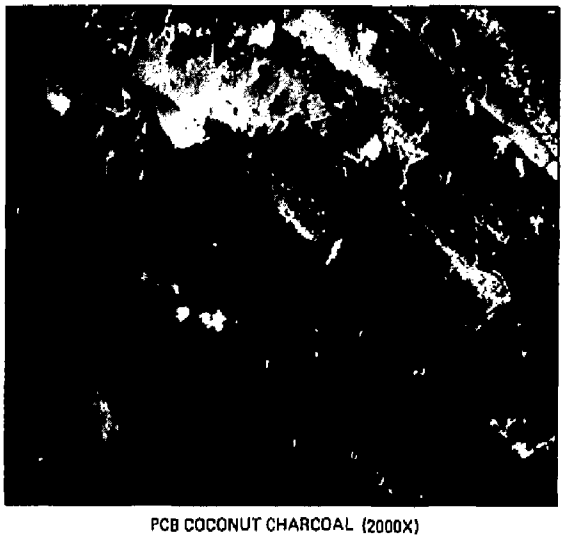
Specimens of PCB and BPL charcoal were ground to powder and subjected to a powder diffractometer using Cu Kα radiation. No crystalline peaks were observed in either powder sample. However, a broad enhancement of the background radiation was observed. This observation would indicate that both grades of charcoal are essentially amorphous. Therefore, no clear distinction between the two grades can be made in terms of crystallinity.

The SEM evaluation, however, revealed some differences in the morphology of the external surfaces of the charcoal. Figure 10-1 illustrates typical appearance (2000 X), of the PCB (coconut) and BPL (coal based) charcoals. As can be seen, the coconut charcoal contains significant areas of microporosity embedded in a lamellar-type surface structure. The visible pores are on the order of 5000 Å (5×10^{-5} cm), compared to the manufacturer's specification of 15-400 Å (1.5×10^{-7} to 10^{-6} cm), for the PCB and BPL charcoals which cannot be seen at 2000 X magnification. The BPL charcoal appears to contain more irregularly shaped areas with much surface debris. No evidence of the microporosity observed in the PCB charcoal can be seen in the BPL charcoal.

Although it is premature to clearly identify the reasons for differences in helium pumping speed based upon these cursory observations, it is possible that the greater degree of microporosity observed in the PCB charcoal could permit better helium access into and out of the interior of the charcoal. This may be the first clue to those factors in the charcoal that could affect helium pumping performance. More work on the microchemistries and pore size distributions of the charcoal is needed before the explanation based upon morphological differences can be supported.



BPL COAL-BASED CHARCOAL (2000X)



PCB COCONUT CHARCOAL (2000X)

H85-1418-018

Fig. 10-1 Microstructure of Charcoal Granules

11 - DISCUSSION OF RESULTS

On the basis of the results, it is clear that the original objectives of this program have been met. The substrate thickness, the braze alloy thickness and the charcoal particle size distribution have been optimized for the range of variables investigated to yield helium pumping performances that are reproducible and will meet anticipated fusion reactor requirements for helium cryopumping. We identified a copper cement bond/charcoal combination which produces comparable performance, and offers the manufacturing flexibility expected of a cement as compared to a braze. The choice of either brazing or copper cement for the bonding agent can be made as a matter of convenience, cost, availability and geometry.

Brazing techniques used in this program are suitable for producing flat charcoal pumping surfaces up to 28 $in.^2$ (180 cm^2). However, it is anticipated that scale-up to larger sizes is possible with improvements in brazing practice. Brazing methods would be difficult to apply to large complex shapes with curvilinear contours — as access and control of heat transfer to such shapes would be difficult to maintain. In such cases, the copper cement approach appears to be a preferred solution. Durability (i.e., adherence) of the charcoal particles on the substrate may be an issue for most bonding approaches. It was not possible to quantify the bond strength of the charcoal, although bond integrity was verified in a series of thermal cycle tests. The charcoal particles have quite irregular shapes and are held by a combination of shear and tension forces. The bonding interface between the charcoal and the bonding agent will influence the heat transfer characteristics as well as determine the durability of the charcoal cryopanel. One would anticipate that panels demonstrating high performance would also have good durability.

The effect on performance of cryopanel exposure to various environmental conditions has been demonstrated, but the cause and effect are not defined. Prolonged exposure of the charcoal to vacuum without liquid nitrogen shielding has an adverse effect on its subsequent helium pumping capability. However, use of liquid nitrogen shielding and bakeout restores the charcoal helium pumping performance. The fact that its capability can be restored is significant; the value of charcoal as a helium sorbent would be greatly diminished if the degradation were irreversible or unavoidable.

The results previously discussed are indicated by a limited number of tests and require additional investigation for verification. The helium pumping performance of charcoal is degraded if the charcoal at ambient temperature is exposed to vacuum for a prolonged time (from 16 hr to several days), before it is cooled for pumping. A low temperature bake (330°K) has no significant effect on improving the charcoal's performance. However, if the liquid nitrogen thermal shield surrounding the charcoal is maintained during the prolonged time between runs, helium pumping performance is subsequently high. Use of liquid nitrogen shielding with a 400°K bakeout restored the charcoal's high performance. The common element in maintaining high performance appears to be the continued use of liquid nitrogen shielding during prolonged charcoal exposure to vacuum. The phenomenon of charcoal performance degradation have require further evaluation. Although the nature and source of the degradation have not been identified, it is likely that hydrocarbon contaminant and water vapor from the residual vacuum are the source.

12 - CONCLUSIONS & RECOMMENDATIONS

The major conclusions of this program are summarized, as follows:

- The charcoal particle size distribution (12x30 mesh) which produced the best helium pumping performance was confirmed as the optimum particle size. Both finer and coarser particle sizes showed diminished pumping speed and capacity
- Thermal cycle tests showed that both the braze and cement bonded charcoal/bond systems exposed to temperature excursions between 4°K and 77°K (a typical regeneration profile), and between 4°K and ambient (shutdown profile) remained intact after these tests
- Systems consisting of charcoal bonded with copper cement or with braze exceeded the demonstrated capability of the charcoal/epoxy bond standard
- Scale up of charcoal/braze samples imposes no size limitation
- A 16 in. (40.6 cm) diameter charcoal cryopanel fabricated and installed in a TSTA compound cryopump showed high helium pumping performance with a projected specific speed of 4.9 l s^{-1} cm^{-2} for a double chevron shielded pump
- Repeated or prolonged exposure to vacuum of an ambient temperature sample with no bake or with only a low temperature (330°K) bake subsequently resulted in degraded pumping performance. Tests in which the sample was surrounded by a thermal shield held at liquid nitrogen temperature during vacuum exposure resulted in maintenance or restoration of the charcoal's pumping capability
- The superiority of coconut based charcoal may be related to the presence of micro-pores which could aid passage of helium into and out of the sorbent's interior. This type of pore was not present in the coal-based samples.

Based on the results of the cryopump development over the last two years, we recommend that the following tasks be performed:

- Characterization of various charcoal grades to identify microstructural correlation with helium pumping performance
- Metallographic study of charcoal/bond interface
- Evaluation of charcoal contamination
- Evaluation of aluminum substrates as an alternative to copper
- Scale up of copper cemented charcoal cryopanels
- Characterization of charcoal performance in co-pumping helium/hydrogen mixtures.

13 - REFERENCES

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