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DEVELOPMENT OF CHARCOAL SORBENTS FOR HELIUM CRYOPUMPING

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1 - SUMMARY

The performance of charcoal as a sorbent for pumping helium in vacuum chambers is well demonstrated. The charcoal typically is bonded to a cryogenically cooled substrate using an epoxy binder; however, the epoxy is considered incompatible with the fusion environment. This program demonstrated helium pumping speed versus capacity for constant throughput for cryogenically cooled charcoal test samples prototypical of elements which could find applications in fusion reactors. The two-phase program first established the optimum charcoal, based on grade and particle size, using an epoxy binder, and then used this optimum charcoal to evaluate inorganic bonding materials and attachment methods. The fusion-compatible (inorganic) samples were compared to the best performing sample in the first phase, which was coconut charcoal PCB 12x30 mesh. The bonding agents included brazes, metal cements and ceramics. One added sample had mechanically restrained charcoal.

Two attachment methods showed promise for future development: A copper cement and an Ag-Cu-P braze produced helium pumping speeds at low capacity ranging from 7 to 10 liter/(s.cm²) compared to the best epoxy sample (8.1 liter/(s.cm²)). The throughputs ranged from 4.7×10^{-4} Torr-liter/(s.cm²) to 6×10^{-4} Torr-liter/(s.cm²).

The effect of different levels of throughput and the effect of pumping an 80-20 deuterium-helium mixture were also evaluated. Reducing the 6×10^{-4} Torr-liter/(s.cm²) by 50% had no significant effect, indicating insensitivity to flow in this throughput range. The mixture run showed the capability to pump both gases although the pumping speed dropped more rapidly with increased capacity than with helium alone.

These results suggest that inorganically bonded charcoal-based helium cryopumps offer significant promise for meeting helium pumping requirements for advanced fusion reactors.

2 - INTRODUCTION

Production of helium in a deuterium-tritium fusion reactor represents a vacuum system throughput of unprecedented proportions. The rate is more than 15 Torr-liters per second for each 1000 MW of fusion power, so that pumping speeds for power reactors can be in excess of one million liters per second. One method for achieving these high pumping speeds is cryopumping.

Alternative methods of helium cryopumping have been studied (Ref. 1-6), including the use of cryotrapping with working fluids such as argon gas, and use of cryogenically cooled sorbents, such as molecular sieves and charcoals. At the outset of this program, the use of charcoal as an alternative was no longer being pursued although it was felt that this method could be improved significantly within the limits of fusion compatibility by improving the coverage of the charcoal on the pumping surface.

Charcoal sorbent cooled to between 4.2 and 18K has demonstrated effective pumping of helium (Ref. 1,5,7); however, the charcoal requires intimate thermal contact with a cryogenically cooled substrate. Possible methods of maintaining contact include chemical and mechanical bonding, and mechanical retention. Chemical bonding with epoxy is commonly used in commercial devices, however, use of epoxy resin binders is questionable in the fusion radiation environment. Mechanical bonding with a solder has been evaluated (Ref. 7); however, solder bonding precludes high density coverage of sorbent.

This program was conducted to develop methods for improving the helium pumping performance of charcoal cryopumping compared to that demonstrated in previous evaluations.

The test samples were prepared at Grumman and the 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.

3 - PROGRAM OBJECTIVE

The program objective was to evaluate in a limited test series the helium cryo-sorption capability of charcoal without using organic binders. Inorganic bonding had been used previously (Ref. 1,7) in response to the need for a fusion-compatible cryopump for the Tritium Systems Test Assembly (TSTA) at Los Alamos. In this case, the binder was a low melting point tin-silver alloy into which the charcoal was embedded while it was liquid. Although the coverage appeared to be only about 60 to 70% charcoal in the alloy, the pump met the TSTA objectives.

This program evaluated methods of improving the coverage of charcoal on the cryogenically-cooled substrate with the purpose of improving pumping speed and capacity. In order to reduce any variability introduced by the test apparatus when comparing results, the complete program included an initial phase in which a single binder (silver-epoxy was used) and a single substrate (aluminum) were tested with the charcoals. The single charcoal size and type producing the best results in the first phase was used throughout the second phase, to satisfy the program objective.

The information derived from this work included the effect on helium pumping performance of:

- Charcoal type, size and mixture
- Throughput rate
- Braze type and processing
- Mechanical attachment of sorbent
- Inorganic binders
- A deuterium-helium mixture.

4 - SURVEY OF CHARCOAL SORBENTS

Although the effectiveness of charcoal as a cryosorbent for pumping helium at low temperatures has been reported (Ref. 1,5,7), no systematic study has been undertaken on the effects of charcoal type and/or particle size distributions on the helium pumping speed and helium pumping capacity. The purpose of this phase of the screening program was to identify several commercial grades of charcoal that could yield the highest helium pumping speeds and to evaluate the effect of particle size distribution on helium pumping characteristics.

Three grades of charcoal were selected for this survey: two coal-based and one coconut-based. One coal-based charcoal and the coconut-based charcoal were available in three irregular particle size distributions, while the other coal-based charcoal was available in a pelletized form. Table 4-1 indicates the characteristics of each charcoal type as reported by the manufacturer. The charcoal is shown in the macrograph of Fig. 4-1. Although the macroscopic porosity of the charcoals is evident, the microporosity of each charcoal could not be resolved. Thus, no clear differences between the various charcoal grades could be determined.

Table 4-1 Description of As-Delivered Charcoal Grades

TYPE	SOURCE	PORE DIAMETER, (Å)	SURFACE AREA (m ² /g)	BULK DENSITY (g/cc)	PARTICLE DENSITY** (g/cc)	MESH SIZE (US SIEVE)
PCB	COCONUT	15-400	1150-1250	0.44	0.85	4x10 6x16 12x30
BPL	COAL	15-400	1050-1150	0.48	0.85	4x10 6x16 12x30
DESOREX**	COAL	-	1300	0.40	-	2 mm DIA x 7 mm CYLINDER
*Hg DISPLACEMENT						
**PROVIDED BY LURGI-UMWELT, FRANKFURT, GDR

Fig. 4-1 Sem Micrograph of Coconut Charcoal Showing Macroscopic Porosity

To further evaluate the effects of charcoal particle size on volume packing of particles (i.e., bulk density), samples of each particle size were poured into a 25 cc container and weighed. Table 4-2 presents the results of these measurements. No significant differences in bulk density were observed for the various particle size distributions.

Table 4-2. Bulk Density for BPL & PCB Charcoal Grades

PCB	Mass (g)	Volume (cc)	Density (gm/cc)
4x10	13.197	25	0.528
6x16	12.992	25	0.520
12x30	12.969	25	0.519
BPL	Mass (g)	Volume (cc)	Density (gm/cc)
4x10	13.169	25	0.526
6x16	15.129	25	0.605
12x30	13.817	25	0.552

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Table 4-3 illustrates the measurement of the average mass/particle obtained by weighing a representative number of particles from each batch. Table 4-3 also indicates that largest charcoal particles are a factor of 6 to 20 times greater than the smaller particles.

Table 4-3. Average Mass of Charcoal Particle

Type	Mesh size	No. of particles	Total wt (g)	Unit weight (mg/particle)
PCB	4x10	103	1.61475	15.7
PCB	6x16	30	0.07465	2.48
PCB	12x30	30	0.01825	0.61
BPL	4x10	61	1.41622	23.22
Desorex	—	70	1.17455	16.78

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To evaluate the effects of mixing various charcoal grades on the bulk density of the mix, nine different combinations of the three particle sizes within each grade were mixed. Tables 4-4 and 4-5 illustrate the results of these measurements for PCB and BPL charcoals. A 40% volume mix of 4x10 (U.S. standard mesh size) with 6x16 particles results in the greatest bulk packing for PCB charcoals, while a 20% volume mix of 12x30 with 6x16 particles results in a maximum particle density for BPL charcoals. No clear reason for this result is apparent; however, differences in particle shape and size of the two charcoals may be responsible for the observed results.

Table 4-4. Density Analysis of PCB Charcoal Mixtures

No.	Mix	Mass (gm)	Volume (cc)	Density (g/cc)
1	5cc (4x10) + 20cc (12x30)	15.530	23	0.675
2	10cc (4x10) + 15cc (12x30)	15.378	23	0.668
3	15cc (4x10) + 10cc (12x30)	15.668	22	0.712
4	5cc (4x10) + 20cc (6x16)	15.114	24	0.629
5	10cc (4x10) + 15cc (6x16)	15.439	21	0.735
6	15cc (4x10) + 10cc (6x16)	15.313	225	0.680
7	5cc (12x30) + 20cc (6x16)	15.099	22	0.686
8	10cc (12x30) + 15cc (6x16)	14.729	22	0.669
9	15cc (12x30) + 10cc (6x16)	14.890	215	0.692

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Table 4-5. Density Analysis of BPL Charcoal Mixtures

No.	Mix	Mass (gm)	Volume (cc)	Density (g/cc)
1	5cc (4x10) + 20cc (12x30)	16.514	22	0.728
2	10cc (4x10) + 15cc (12x30)	16.539	23	0.719
3	15cc (4x10) + 10cc (12x30)	16.116	22	0.732
4	5cc (4x10) + 20cc (6x16)	17.270	24	0.719
5	10cc (4x10) + 15cc (6x16)	15.439	21	0.735
6	15cc (4x10) + 10cc (6x16)	16.409	22	0.745
7	5cc (12x30) + 20cc (6x16)	16.829	21.5	0.783
8	10cc (12x30) + 15cc (6x16)	16.627	22	0.755
9	15cc (12x30) + 10cc (6x16)	16.397	22.5	0.728

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To evaluate the differences between the volume density and areal density of these particles, pieces of sticky tape (50 mil width) were used to bond the mixtures of charcoal particles and the areal density (g/cm^2) was measured. Table 4.6 indicates that the 4x10 particles yield the highest mass density per unit area for both PCB and PBL particles while the 12x30 particles yielded the lowest areal densities. To evaluate the effects on areal density of mixing various particle size distributions, the equivalent particle size mixtures used in Tables 4-4 and 4-5 were spread on to sticky back tape. Tables 4-7 and 4-8 illustrate the results of these measurements. Tables 4-6 to 4-8 clearly illustrate that the mixing of particles reduces areal density, with the largest unmixed particles (4x10) presenting the greatest areal density. One might expect that the highest helium pumping speeds and capacities would be associated with the largest particles because they have the greatest internal pore area per unit area exposed to helium flow.

Table 4-6. Areal Densities of Charcoals (Pure Materials) on Adhesive Tape

Charcoal type	Particle size	Mass of charcoal (mg)	Area (mm ²)	Density (mg/mm ²)
PCB	(4x10)	3307.62	2700	1.225
PCB	(12x30)	833.15	2400	0.347
PCB	(6x16)	1864.08	2937.5	0.634
BPL	(4x10)	3149.86	2462.5	1.279
BPL	(12x30)	1006.11	2550	0.394
BPL	(6x16)	2764.73	2600	1.063
Desorex	—	3737.00	3500	1.067

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Table 4-7. Areal Density Calculations for Charcoal Mixtures of PCB Charcoal

Mixture no. (See table 4-4)	Mass of charcoal (mg)	Area (mm ²)	Density (mg/mm ²)
1	1307.53	2950	0.443
2	1401.09	3100	0.452
3	1606.71	3525	0.456
4	1984.63	3400	0.584
5	1372.68	2550	0.538
6	2168.45	3000	0.723
7	1304.95	2850	0.458
8	1359.43	2900	0.469
9	1480.88	3125	0.474

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Table 4-8. Areal Density PBL Charcoal Mixtures

Mixture no. (See Table 4-5)	Mass of charcoal (mg)	Area (mm ²)	Density (mg/mm ²)
1	1329.57	3062	0.434
2	1887.25	2800	0.674
3	2058.10	2700	0.762
4	2787.01	2775	1.01
5	2523.50	2700	0.935
6	2818.51	2650	1.063
7	1914.96	2525	0.758
8	1746.30	2700	0.647
9	1354.54	2625	0.517

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5 - BONDING & RETENTION STUDIES

5.1 EPOXY BONDING

A silver-based epoxy bonding agent was used for all particle size distribution tests, which were run to evaluate the effects of charcoal grade and particle size distribution on helium pumping speed. A thin layer (approximately 0.003 - 0.005 in.) of epoxy (with hardener) was spread on a 4 in.-diameter aluminum alloy (6061-T6) plate and the charcoal particles were spread uniformly on the uncured layer. All specimens were cured two hours at 140°F. Curing produced a well bonded uniform distribution of charcoal on all surfaces. This is illustrated in Fig. 5-1.

Fig. 5-1 Charcoal Particles Embedded with Epoxy Adhesive on Aluminum Plate

5.2 INORGANIC BONDING AGENTS DEVELOPMENT

Since organic bonds are unacceptable in a fusion neutron environment because of their eventual degradation, several approaches to achieving a thermally conductive bond using inorganic bonding agents were evaluated. These include 1:

1. Brazing
2. Low temperature curing ceramic adhesives
3. Low temperature curing metallic adhesives
4. Mechanical attachments.

Table 5-1 summarizes the various material combinations using epoxy and inorganic bonds that were tested.

Table 5-1. Description of Charcoal Test Specimens

Sample no.	Charcoal type	Mesh size	Bonding agent	Substrate
1	Coconut (PCB)	4x10	Silver-epoxy	6061-Al
2	Coconut (PCB)	6x16	Silver-epoxy	6061-Al
3	Coconut (PCB)	12x30	Silver-epoxy	6061-Al
4	Coal (BPL)	4x10	Silver-epoxy	6061-Al
5	Coal (BPL)	12x30	Silver-epoxy	6061-Al
6	Coal (Desorex)	-	Silver-epoxy	6061-Al
7	Coconut (PCB)	$\begin{Bmatrix} 4 \times 10 \\ + \\ 12 \times 30 \end{Bmatrix}$	Silver-epoxy	6061-Al
8	Coconut (PCB)	$\begin{Bmatrix} 6 \times 16 \\ + \\ 12 \times 30 \end{Bmatrix}$	Silver-epoxy	6061-Al
9	Coconut (PCB)	12x30	Ag-Cu braze + Ti sheet	Nickel
10	Coconut (PCB)	12x30	Mechanical (See text)	
11	Coconut (PCB)	12x30	Ag-Cu-P braze	Cu
12	Coconut (PCB) Baked to 900C	12x30	Silver-epoxy	Al
503	Coconut (PCB)	12x30	Al_2O_3	SS-Flame sprayed on Al
516	Coconut (PCB)	12x30	ZrO_2	SS-Flame sprayed on Al
571	Coconut (PCB)	12x30	Cu	SS-Flame sprayed on Al
543	Coconut (PCB)	12x30	Cu	SS-Flame sprayed on Al
543A	Coconut (PCB)	12x30	Cu	Cu
543B	Coconut (PCB)	12x30	Cu	Al
565	Coconut (PCB)	12x30	Stainless steel	SS-Flame sprayed on Al

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5.2.1 Brazing

One brazing method involved the use of a titanium wetting agent and a silver-copper braze alloy. Brazing operations were conducted in a vacuum-brazing oven at 10^{-6} Torr and 900°C for 10-30 minutes. The configuration consisted of a 4 in.-diameter x 1/8 in. thick nickel plate, which was machined with a 1/4 in. wide lip around the edge. A 0.001 in. layer of Ti foil was sandwiched between two 0.003 in. layers of Ag-Cu braze alloy. Brazing produced well bonded charcoal particles with braze alloy apparently wetting both charcoal and substrate. This is illustrated in Fig. 5-2. Evaluation of the interface between the charcoal and the braze alloy indicated that the braze had infiltrated into the charcoal pores (Fig. 5-3). These

Fig. 5-2 Coconut Charcoal Brazed to Nickel Substrate

Fig. 5-3 Section of Braze Showing Wetting & Penetration of Coconut Charcoal

observations are consistent with the tendency of the dissolved titanium in the braze to form a carbide at the interface and lower the interfacial energy. All specimens survived cooldown to liquid N_2 temperatures with no loss of charcoal.

Because the vacuum-brazed charcoal showed low performance (Subsection 9.2.1), an additional sample was made using Ag-Cu-P braze, which was applied in air to a 1/8-in. copper substrate. In this case, the charcoal was not subjected to the high braze temperature but was pressed into the molten braze. This sample, proposed and prepared by Livermore personnel, was included because the Ag-Cu-P had an apparent higher viscosity than the Ag-Cu braze, with the prospect that the braze would not infiltrate the charcoal pores. The charcoal was well attached to the braze and the method produced good coverage.

5.2.2 Low Temperature Curing Ceramic Adhesives

Another approach to achieving a bond involved the use of low temperature curing ceramic bonding agents. Substrate surfaces consisted of 6061-T6 aluminum with a flame sprayed stainless steel, pure copper and aluminum. All ceramic adhesives were obtained from commercial sources and consisted of one- or two-part mixtures that could be readily spread or trowled with a spatula in a layer on to the substrate. Charcoal particles were spread on the uncured ceramic layer to form a dense uniform layer and allowed to cure at room temperature for one hour. Final curing consisted of placing the partially cured specimens into an oven at room temperature and slowly raising the temperature to 250°F for two hours followed by a slow cool to ambient. Excess or unbonded charcoal was brushed off the surfaces. All specimens were cooled in liquid N_2 to evaluate compatibility of charcoal and substrate at low temperatures.

5.2.3 Low Temperature Curing Metallic Adhesives

Two commercial adhesives consisting of stainless steel and copper based compositions were evaluated. Both adhesives were evaluated on aluminum substrates flame sprayed with stainless steel, while the copper adhesive was also evaluated on pure copper and pure aluminum substrates. The metal substrates were chemically cleaned in nitric acid and rinsed in distilled water. These one-part adhesives were spread in a thin layer over the metallic surfaces and a layer of charcoal was placed on to the uncured adhesive. The adhesive was allowed to cure at room temperature for two hours. The composite was then placed in a curing oven for 2.5 hr at 250°F and

allowed to cool slowly to ambient. Excess or unbonded charcoal was brushed off and the specimens were cooled in liquid N_2 . They appeared to survive the liquid N_2 cooldown with little loss of charcoal. Figure 5-4 illustrates the typical appearance of a sample.

Fig. 5-4 Optical Micrograph of Copper-Bonded Charcoal

5.2.4 Mechanical Attachments

Multiple layers of charcoal were considered a way of increasing pumping speed by increasing coverage. A metallic honeycomb was bonded to an aluminum substrate and charcoal particles were poured into the honeycomb cells and tamped into form several layers of sorbent. The charcoal was retained in the honeycomb cell using a stainless steel 20 mesh screen epoxy-bonded to the outer edges of the honeycomb. Honeycomb cell size was $1/8$ in., and the honeycomb material was titanium.

6 - TEST PROGRAM

The two-phase test program to evaluate charcoal pumping performance comprised a charcoal screening test phase, using a single method of bonding the charcoal to a substrate, and a bonding or retention test phase. The test specimens are described in Table 5-1.

The charcoal sample defined by type, size and mixture, showing the best performance in the charcoal screening test phase was selected for the bonding test phase. This phase to evaluate fusion-compatible attachment methods included various bonds such as inorganic metallic cements, ceramics and brazes, and several substrates including aluminum, stainless steel, copper and nickel. An additional test article was prepared to test a mechanically attached charcoal sample.

The comparison among samples was based on the measurement of specific pumping speed versus specific pumping capacity for a constant helium throughput.

7 - TEST SETUP

The vacuum test installation at the LLNL Vacuum Technology Laboratory was used for the test program. Each charcoal test sample was characterized for helium pumping speed versus capacity using the equipment and procedures described below.

7.1 TEST APPARATUS

The system used in the study is shown in Fig. 7-1 and schematically in Fig. 7-2. The test pump was an appendage to the main tank and held each charcoal test sample. A Roots/mechanical pump roughed the main chamber and test pump, while a Displex cryopump provided final roughing to a pretest pressure in the 10^{-7} Torr range. In later tests the cryopump was removed, and the liquid nitrogen cooled chevrons of the test pump performed final roughing.

One flow meter or two meters in series were used to measure helium throughput, which was bled into the main chamber through a servo-controlled valve. Samples were tested with a constant helium throughput of 0.037 Torr-liter/second, which was selected as being representative of fusion reactor throughput rates scaled to the size of the sample.

Two additional conditions were tested. Two tests were made at half flow to evaluate the effect of this reduction on pumping speed, and one used an 80-20 mixture of deuterium and helium to show the capability for pumping these gases.

7.2 TEST PUMP

The test pump used in this program was a rugged workhorse cryopump designed by Livermore for ready demounting and servicing. The pump illustrated in Fig. 7-3 and 7-4 featured a liquid helium reservoir within a four-sided rectangular array of chevrons. This single set of chevrons was maintained at approximately 80°K by conduction to a liquid nitrogen reservoir. A plate on the bottom, which completed the shielding of the helium reservoir, was attached to the chevron array by screws for ready disassembly. This assembly was suspended in a cylindrical vacuum vessel having a top flange for servicing.

Fig. 7-1 Test Apparatus Showing Main Tank & Appendage Test Pump

Fig. 7-2 Test System Schematic

Fig. 7-3 Workhorse Cryopump for Charcoal Sorbent Tests

Fig. 7-4 Workhorse Test Pump Removed from Pump Housing

7.3 TEST ARTICLE

Each test article consisted of a 4-in. diameter metal plate with charcoal particles bonded to it or mechanically restrained as described in Section 5. The metal plate was held by a retainer ring against the pump's reservoir, with a copper mesh between the plate and the reservoir to enhance thermal conductance (Fig. 7-5).

A silicon diode temperature sensor was attached at the center of each test article except for the mechanically restrained sample.

Fig. 7-5 Charcoal Sample Installed inside Test Pump

7.4 INSTRUMENTATION

Two flow meters, three temperature sensors and three pressure gages were used to monitor the condition of the test equipment and to provide data. The test series was started using a 0-50 sccm (air) flow meter, which had to be used at 4% of its full scale rate to satisfy the required helium flow. The meter was calibrated (Fig. 7.6) in the laboratory. A 0-5 sccm (air) meter made available in the first test set was added series to provide a crosscheck.

Fig. 7-6 Calibration of Flowmeter for Helium

Silicon diode sensors measured temperature on the test pump chevron, the pump helium reservoir and the test article. The sensors were calibrated in liquid helium or liquid nitrogen prior to their use. Bayard-Alpert-type ionization nude gages measured pressures in the main tank, and at the top and bottom of the test pump container in the interspace outside of the chevron array. The indicated pressure of these gages (calibrated for air) was multiplied by seven for helium based on manufacturer's data.

The test control center is shown in Fig. 7-7.

Fig. 7-7 Control Station for Charcoal Sorbent Tests

8 - TEST METHOD

Each test article depicted in Table 5-1 was assembled to the test pump and fitted with a diode sensor at its center prior to installation in the vacuum facility. Each test consisted of the following procedure.

The system was rough pumped using the Roots blower/vacuum pump, and hot air was passed through the test pump helium dewar to desorb residual water vapor from the charcoal. The Displex cryopump attained high vacuum conditions, however, it was not available for the entire test series. In its absence, the test pump shielding filled with liquid nitrogen effected the final pumpdown. When the chevron shields were cold, transfer lines between a helium storage dewar and the test pump allowed filling of the pump's liquid helium reservoir. Temperature of the test article was stabilized to 7.5K prior to the start of the run. The quantity of liquid helium in the test pump reservoir was maintained between 90 and 100% full during the tests.

After thermal stabilization, the test gas (either helium or an 80-20 mixture of deuterium and helium) was fed into the main tank using a servo-controlled flow valve. Flow was increased until the steady state value was achieved within a few minutes (usually less than 2 minutes). Each run was continued until the test pump pressure indicated on the ionization gage rose to approximately 10^{-4} Torr. In cases where the initial pressure was close to this value, the run was terminated when sufficient data was taken to characterize the sample. Pressure, temperature and test gas flow were recorded at appropriate intervals during each test. In addition, a strip chart recorded pressure for run diagnostics. When the run was terminated by stopping the test gas flow, the pump pressure in the system was recorded 5 to 10 minutes afterward.

9 - TEST RESULTS

The pumping speed as a function of capacity was determined for all cryosorbent samples under investigation. The test procedure was to use the constant flow method in which a fixed flow of test gas was introduced into the main chamber of the test apparatus. This flow was continued until the sample reached its capacity as indicated by a noticeable increase in the appendage pump pressure which was the reference pressure for data reduction. Runs were generally continued until the reference pressure increased to 10^{-4} Torr (indicated for N_2).

The helium pumping speed S was determined from the relationship to the throughput Q :

$$S = Q/P$$

where P was the pressure measured at the top of the appendage pump cavity in the volume between the pump housing and the pump's nitrogen-cooled thermal shield.

Although the speed of the cryosorption surface is larger than the value derived using the above relationship, by virtue of the conductance limitation of the pump chevron, the procedure used was appropriate since the objective of the program was to compare performance among different cryosorbent samples.

It was generally observed that those samples exhibiting the highest helium sorbent capacity also had the highest helium pumping speeds. It should be noted for reference that when the sample has absorbed $0.75 \text{ Torr}\cdot\text{l}/(\text{cm}^2)$ of helium with a throughput of $6 \times 10^{-4} \text{ Torr}\cdot\text{l}/(\text{s}\cdot\text{cm}^2)$, the total pumping time is 20 minutes. Because cryopumps in fusion reactors will probably be regenerated within this time to minimize tritium inventory, the data taken prior to this time is of more interest.

The test logs and reduced data tabulation sheets are tabulated under separate cover and are on file at Grumman.

9.1 CHARCOAL SORBENT TEST

9.1.1 Coconut Charcoal on Silver Epoxy - Various Mesh Sizes

This set of runs evaluated the performance of the coconut charcoal (PCB) in mesh sizes 12x30, 6x16, and 4x10 to determine for PCB the best performing particle size. Samples were exposed to the same helium throughput, 6×10^{-4} Torr-liter/(s.cm²).

Figure 9-1 shows that the charcoal performance varied with particle size, with the smallest size producing the best performance. Although the largest particle size had the greatest total internal pore area per unit area exposed to the helium flow, it appears that intimate thermal contact of the charcoal to the cooled substrate was the controlling factor.

Fig. 9-1 PCB Charcoal on Epoxy Bond - Comparison of Various Mesh Sizes

9.1.2 Coal Charcoal on Silver Epoxy - Various Mesh Sizes

This set was similar to that reported above except that coal-based charcoal (BPL) was used here. Performance was in the same order (Fig. 9-2) with the smaller particle sizes performing better; however, the BPL performance was poorer than the PCB. The intermediate size particles (6x16) were not tested after the BPL demonstrated significantly poorer performance.

Fig. 9-2 BPL Charcoal on Epoxy Bond - Different Particle Sizes

9.1.3 Comparison of Charcoal Types

Figure 9-3 compares the best performing coal and coconut charcoals to each other and to a pelletized charcoal (Desorex). This very limited evaluation showed that the coconut charcoal has a decided advantage over the other grades in this application. Although Desorex particles were larger, their performance was poorer than the largest coconut charcoal particle (PCB 4x10).

Fig. 9-3 Comparison of Various Charcoal Types

9.1.4 Comparison of Samples with Mixed Charcoal Mesh Sizes

A set of tests was run to evaluate the effect of mixing two different charcoal sizes in a single sample, to improve the helium pumping capability by filling the spaces between the larger particles. Two samples were tested: a mix of 12x30 and 4x10 mesh PCB, and a mix of 12x30 and 6x16 mesh PCB (Fig. 9-4 and 9-5). The single mesh size PCB 12x30 remained the highest performing sample in the group.

Fig. 9-4 PCB Charcoal on Epoxy Bond - Various Particle Sizes

Fig. 9-5 PCB Charcoal on Epoxy Bond - Comparison of Particle Size Mixture

9.1.5 Comparison of Helium Throughputs

Additional runs were included to evaluate the effect of reduced helium flow on speed and capacity. The helium throughput was reduced to 50% of the nominal value for a PCB 12x30 mesh sample and for a PCB 6x16 sample. It was expected for a given surface coverage of helium that higher helium pumping speeds would result, however, the results (Fig. 9-6 and 9-7) show little difference in performance for the two flow conditions and in fact show the inverse result. The inverse is not explainable other than test variability, but the small change in performance is indicative of a lack of sensitivity in the range of flows being tested.

Fig. 9-6 Comparison of Different Flow Rates - 12x30 Mesh PCB Charcoal

Fig. 9-7 Comparison of Different Flow Rates - 6x16 Mesh PCB Charcoal

9.1.6 Helium-Deuterium Mixture Test

Cryopumps for fusion applications must pump both helium and hydrogen mixtures. A run was included in the test program to show the effect of an 80-20 mixture of deuterium and helium on the ability to pump helium with the PCB 12x30 mesh sample. The total throughput was adjusted so that the helium throughput was held at the prevalent value of 6×10^{-4} Torr-liter/(s·cm²).

This test showed that the helium pumping performance is degraded as seen in Fig. 9-8. Apparently, the deuterium blocks or competes with helium for the charcoal pore sites and reduces helium pumping speeds by about 30% after 20 minutes of pumping. It must be emphasized however, that the test pump has a large (approximately 1350 cm²) area for pumping deuterium compared to the surface available for pumping helium (60 cm²), therefore the results are specific to the configuration that was tested.

Fig. 9-8 Relative Pumping Speed of Helium in Deuterium/Helium Mixture

9.2 CHARCOAL TESTS WITH INORGANIC BONDING AGENTS

Since the PCB 12x30 mesh charcoal was shown to produce the highest helium pumping performance among the configurations described in Subsection 9.1, all charcoal bond tests used this single charcoal grade throughout.

9.2.1 Charcoal Brazed to Substrate

The two samples prepared in significantly different ways (Subsection 5.2.1) showed significantly different results (Fig. 9-9 and 9-10). The sample with Ag-Cu-P braze alloy demonstrated the highest helium pumping performance of any sample in the test series. Although the Ag-Cu-Ti braze alloy sample showed excellent wetting and bonding of charcoal to substrate, its performance was poor.

Fig. 9-9 PCB 12x30 Charcoal - Silver Copper Braze to Titanium

Fig. 9-10 PCB 12x30 Charcoal-Ag-Cu-P Braze to Copper

Two possible contributing factors were: the braze had penetrated the charcoal pores and clogged a significant number of potential sites for pumping helium, and the charcoal had been modified by the extended exposure at 900°C. To verify this latter hypothesis, a sample which consisted of charcoal baked at 900°C in vacuum and then applied to the silver-epoxy binder after cooldown was tested. Although the performance improved (Fig. 9-11) as compared to the brazed sample, it was significantly poorer than a comparable sample (Fig. 9-1, 12:30 mesh) for which the charcoal was used in as-received condition. The baking of the charcoal thus has an adverse, unexplained effect. This observation appears to be consistent with the performance of the Ag-Cu-P braze sample.

Fig. 9-11 PCB 12x30 Charcoal on Epoxy - Oven-Baked at 900°C

9.2.2 Charcoal Attached by Inorganic Cements

Figure 9-12 presents the results of testing samples made with inorganic binders. The copper cement, which was used to make two samples on aluminum substrate, produced satisfactory performance and showed that a useful inorganic bonding material was available for fusion applications. Other samples in this category produced poor performance indicating the importance of good thermal properties of the bonding material, e.g., a sample using ZrO_2 as binder produced no pumping of helium.

Fig. 9-12 PCB 12x30 Charcoal with Metal Cement Bonds

The copper cement was also used on a copper substrate to allow comparison of different substrates. An aluminum substrate consisted of a 1/4 in. thick aluminum substrate which was flame sprayed with stainless steel on one side to which the charcoal was bonded. The exposed surface area was 3.5-in. dia (60 cm²). The copper substrate consisted of 1/8-in. thick copper plate and the surface area covered with charcoal was 4.0-in. dia (79 cm²). Although the outer 1/4-in. radius (30%) was masked by the locating ring (Fig. 7.3), the full area was used for calculation of speed and capacity. This would lead to a conservative estimate of pumping performance. Although the total throughput was equal for both tests, the specific

throughput differed because of the different surface areas. The comparison of the pumping performance of these substrates (Fig. 9-13) showed negligible differences. This would indicate that the thermal conductivity of the bond was more influential than the substrate. However, due to the many variables mentioned, this conclusion is tentative.

Fig. 9-13 PCB 12x30 Charcoal with Copper Cement Bond

9.2.3 Mechanical Attachment

The sample consisting of the charcoal held in the honeycomb structure by a wire mesh produced no measurable pumping capability. The steady throughput was not established before the 10^{-4} Torr (N_2 equivalent) pressure limit was reached. The need for a conductive bond between the charcoal and the substrate was further reinforced by this test.

10 - EVALUATION OF BONDS (POST TEST)

10.1 BRAZE ASSEMBLY

Titanium-modified brazes exhibited some infiltration of the braze alloy in the pores of the charcoal. This phenomenon is probably due to TiC formation at the charcoal-braze interface and is the reason for the excellent wetting and bonding of the charcoal particles to the substrate. However, the reduced helium pumping characteristics of the brazed charcoals are partially related to the pore infiltration by the braze as well as to the effects of the elevated temperatures on charcoal stability. Approaches to brazing charcoal to a cryosupport must be based upon shorter heating and cooling cycles and upon a braze which will wet slightly but not infiltrate the charcoal pores.

10.2 LOW TEMPERATURE CURING ADHESIVES

All low temperature curing adhesives are essentially weakly bonded to the charcoal, because the low curing temperatures combined with the low chemical reactivity of charcoal do not permit strong chemical bonding between charcoal and adhesive. There is no evidence of pore infiltration in these bonded charcoals; however, both weak chemical and mechanical interlock forces may explain the adhesion observed. The systematic effects of bond line thickness and thermal properties of the bonding agent have not been investigated; however, the wide variations observed in helium pumping speeds suggest that these parameters are important to the helium pumping performance and will require further investigation.

10.3 MECHANICAL ATTACHMENTS

Honeycomb-restrained charcoal performed poorly in helium pumping experiments and clearly show that intimate thermal contact between the charcoal layer and the cryosupport is essential for high helium pumping speeds. Mechanical retention of the charcoal particle is simply inadequate to provide the interface contacts needed to cool the particles to liquid helium temperatures.

10.4 DISCUSSION

These results clearly show that the thermal properties of the interface bonding agent are critical to the successful performance of charcoal based helium cryopumps. The bonding agent must be thin enough to minimize thermal gradients across the interface zone yet thick enough to provide bonding interfaces for good adhesion to the substrate.

The thermal conductivity of the bonding agent should be as large as possible to minimize thermal gradients. These conclusions would tend to favor silver and copper based adhesives for this application. The adhesive must provide an intimate physical contact with the charcoal; however, penetration of adhesive into the internal pore structure of the charcoal is detrimental to helium pumping characteristics. Substrate preparation may also be important. Although flame sprayed stainless steel on an aluminum support would tend to increase the "effective" surface area for contact, the additional "thermal resistance" created by this layer may not compensate for the increased surface area. It thus appears that clean, smooth substrates made of copper, nickel and/or aluminum are preferred for this application.

The effect of charcoal particle size on helium pumping speeds indicates, contrary to expectation, that smaller particles are preferred to larger particles. Although it would be very difficult to perform experiments to identify the precise reasons for this observation, it is suggested that temperature of the smaller particles more closely approaches the liquid helium temperature than the larger particles. Given the measured sensitivity of helium pumping speed on charcoal temperature, a significantly larger portion of each smaller particle is available to pump helium. Since our results have not optimized fully the effects of particle size on helium pumping speed and capacity, it may be possible to obtain even higher pumping speeds using smaller particles than those used in this study.

11 - CONCLUSIONS

Testing of the cryogenically cooled charcoal using fusion-compatible binders for pumping helium has shown promising results. The program demonstrated comparable or improved performance with these binders compared to the same charcoal (type and size) using an epoxy binder. The program objective has been satisfied.

Our major conclusions are:

- Of the three charcoals tested, (i.e., PCB coconut, BPL coal and Desorex), the PCB performed best
- Charcoal particle size influenced pumping capability. The smaller particle resulted in better helium pumping performance. For the pump configuration tested, the best charcoal, PCB 12x30 mesh on epoxy, pumped 7.8 liter/(s.cm²) at low capacity and maintained above 7.1 liter/(s.cm²) for approximately 20 minutes with a helium throughput of 6×10^{-4} Torr.liter/(s.cm²)
- A copper cement (with PCB 12x30) showed promise with pumping speed above 5.5 liter/(s.cm²) after 20 minutes. Some variability between different samples using the same copper binder pointed out the need for better process control. No satisfactory ceramic binder was found among the limited samples tested. Their performance was consistently poor compared to the copper cement
- Samples using different brazes produced dramatically different results. An Ag-Cu-Ti braze applied to titanium to enhance charcoal wetting showed about 2 liter/(s.cm²) pumping speed at 20 minutes. An Ag-Cu-P braze, heated in air to avoid high temperature charcoal baking, showed no visible bronze migration (30x) and exhibited helium pumping speeds above 8.5 liter/(s.cm²) after 20 minutes.

The program showed the promise of using metallic bonds for charcoal attachment schemes in helium cryopumps as well as the direction for future work which form the following recommendations.

- Show reproductivity of the best cement and braze bonds
- Evaluate performance of smaller PCB charcoal particles
- Evaluate production techniques and prepare test samples
- Test with helium and helium/hydrogen mixtures
- Prepare pump for use in TSTA.

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