Phil
274
5/24/67
LA-3664

MASTER

LOS ALAMOS SCIENTIFIC LABORATORY
of the
University of California
LOS ALAMOS • NEW MEXICO

Preparation of High Density Graphite
by Hydrostatic Pressing

UNITED STATES
ATOMIC ENERGY COMMISSION
CONTRACT W-7405-ENG. 36

DISTRIBUTION OF THIS DOCUMENT IS UNLIMITED.
129

DISCLAIMER

This report was prepared as an account of work sponsored by an agency of the United States Government. Neither the United States Government nor any agency Thereof, nor any of their employees, makes any warranty, express or implied, or assumes any legal liability or responsibility for the accuracy, completeness, or usefulness of any information, apparatus, product, or process disclosed, or represents that its use would not infringe privately owned rights. Reference herein to any specific commercial product, process, or service by trade name, trademark, manufacturer, or otherwise does not necessarily constitute or imply its endorsement, recommendation, or favoring by the United States Government or any agency thereof. The views and opinions of authors expressed herein do not necessarily state or reflect those of the United States Government or any agency thereof.

DISCLAIMER

Portions of this document may be illegible in electronic image products. Images are produced from the best available original document.

LEGAL NOTICE

This report was prepared as an account of Government sponsored work. Neither the United States, nor the Commission, nor any person acting on behalf of the Commission:

A. Makes any warranty or representation, expressed or implied, with respect to the accuracy, completeness, or usefulness of the information contained in this report, or that the use of any information, apparatus, method, or process disclosed in this report may not infringe privately owned rights; or

B. Assumes any liabilities with respect to the use of, or for damages resulting from the use of any information, apparatus, method, or process disclosed in this report.

As used in the above, "person acting on behalf of the Commission" includes any employee or contractor of the Commission, or employee of such contractor, to the extent that such employee or contractor of the Commission, or employee of such contractor prepares, disseminates, or provides access to, any information pursuant to his employment or contract with the Commission, or his employment with such contractor.

This report expresses the opinions of the author or authors and does not necessarily reflect the opinions or views of the Los Alamos Scientific Laboratory.

Printed in the United States of America. Available from
Clearinghouse for Federal Scientific and Technical Information
National Bureau of Standards, U. S. Department of Commerce
Springfield, Virginia 22151

Price: Printed Copy \$3.00; Microfiche \$0.65

CESTI PRICES

LA-3664

UC-25, METALS, CERAMICS,
AND MATERIALS

TID-4500

H.C. \$ 3.00; MN .65

LOS ALAMOS SCIENTIFIC LABORATORY
of the
University of California
LOS ALAMOS • NEW MEXICO

Report written: December 8, 1966

Report distributed: May 11, 1967

Preparation of High Density Graphite
by Hydrostatic Pressing

by

D. H. Schell

and

L. A. Wertz

LEGAL NOTICE

This report was prepared as an account of Government sponsored work. Neither the United States, nor the Commission, nor any person acting on behalf of the Commission:

A. Makes any warranty or representation, expressed or implied, with respect to the accuracy, completeness, or usefulness of the information contained in this report, or that the use of any information, apparatus, method, or process disclosed in this report may not infringe privately owned rights; or

B. Assumes any liabilities with respect to the use of, or for damages resulting from the use of any information, apparatus, method, or process disclosed in this report.

As used in the above, "person acting on behalf of the Commission" includes any employee or contractor of the Commission, or employee of such contractor, to the extent that such employee or contractor of the Commission, or employee of such contractor prepares, disseminates, or provides access to, any information pursuant to his employment or contract with the Commission, or his employment with such contractor.

THIS PAGE
WAS INTENTIONALLY
LEFT BLANK

PREPARATION OF HIGH DENSITY GRAPHITE

BY HYDROSTATIC PRESSING

by

D. H. Schell

L. A. Wertz

ABSTRACT

Graphitic pieces were made by hydrostatically pressing a thermosetting plastic bonded mix and processing the pressings through a 2500°C graphitizing treatment. Pressings made with 85 parts graphite flour and 15 parts Thermax bonded with 27.5 pph Varcum had densities around 1.8 g/cc and compressive strengths around 9000 psi.

This investigation included the use of different filler materials, binders, and processing variables in an attempt to optimize density and strength.

1. INTRODUCTION

Graphites may be fabricated in a number of ways such as extrusion, molding, and hot pressing with each method producing graphites with unique properties. Hydrostatic pressing is one molding method which has not been investigated to any great extent. The graphite so formed is expected to be isotropic and to possess relatively uniform properties in both the transverse and longitudinal directions. This method may be especially interesting when a thermosetting binder is used.

The purpose of this program was to investigate hydrostatically pressed graphites made with a variety of filler materials; various binder and filler levels, additives and processing variables; and, to some extent, different binders.

2. MATERIALS

2.1 Composition of the Pressing Mix

Hydrostatic pressings were made with a mixture of graphite flour and Thermax bonded with Varcum, a liquid thermosetting resin. The usual mix consisted

of 85 parts graphite flour to 15 parts Thermax. The amount of binder varied considerably but was usually around 27.5 pph of the dry carbon ingredients.

2.2 Graphite Flours

Most pressings were made using one of two types of graphite flour. One, designated 1008-S, was obtained from Great Lakes Carbon Company, New York, N. Y., and was given a LASL identifying number, M-2. The analysis of the high purity flour is presented in Table I. The flour was ground by the vendor to meet the nominal particle size distribution noted in Table II.

TABLE I
CHEMICAL ANALYSIS OF GRAPHITE FLOURS

Element*	M-2 Flour, ppm (range)	S-97 Flour, ppm
Ash	100 - 300	100
Iron in ash	15 - 60	70
Silicon in ash	25 - 100	30

*All other elements analyzed were below the detection limits of spectrochemical (semiquantitative) analysis.

TABLE II
PARTICLE SIZE DISTRIBUTION OF M-2 AND
S-97 GRAPHITE FLOURS

Mesh Size	M-2 Flour, Weight Percent	S-97 Flour, Weight Percent
+65	3	
-65 +100	5	
-100 +150	11	
-150 +200	26	2*
-200 +270	10	8
-270 +325	15	20
-325	30	70
*+200 mesh		

The graphite flour was received in 55-gal drums, each containing 300 lb. The drums of flour were rolled on a 3/4-hp, variable speed drum roller manufactured by U. S. Stoneware. A sample was taken from each drum with a sampling tube and transferred to a plastic tube for radiographic examination to detect high-density impurities.

The flour from eight drums which had passed radiographic evaluation was crossblended into nine drums. The flour in each of the nine drums was screened through a 60-mesh screen in an Abbe No. 2 Turbo Sifter; the tailings were passed through the sifter three times to achieve a net recovery of 98%. The flour discharged from the sifter over an Eriez Model D-2 magnetic grate, and was collected in 55-gal drums. Each drum was then rolled at 40 rpm for 8 hr to homogenize the contents.

The second graphite flour used was obtained from Speer Carbon Company, St. Mary's, Penn., and was designated S-97. This was also a high purity flour having an impurity level as noted in Table I. The particle size of the S-97 flour was considerably smaller than that of the M-2 flour as noted in Table II.

A few pressings were made using a graphite flour from Union Carbide, Carbon Products Division, which had a high coefficient of thermal expansion (CTE). This flour was made by grinding graphitized stock made from a spheroidized coke. The coke was produced by the Carbon Products Division using an air-blowing process during the final stages of solidification. The flour had a particle size distribution similar to the M-2 flour and was slightly less pure.

2.3 Thermax

Thermax is a carbon black obtained from Thermax Carbon Company, Sterlington, La., and has an average particle size of 0.9 μ . The chemical analysis is shown in Table III.

TABLE III
CHEMICAL ANALYSIS OF THERMAX

Element*	ppm (range)
Ash	700 - 2000
Silicon	100 - 620
Iron	30 - 50
Calcium	10
Magnesium	5 - 10
Sodium	5 - 10
Aluminum	3 - 10
Manganese	1 - 5

*All other elements analyzed were below the detection limits of a spectrochemical (semiquantitative) analysis.

Thermax was received from the vendor in 50-lb bags. Five bags full were passed through the 60-mesh screen in the Abbe sifter, passed over the magnetic grate, and packed into a 55-gal drum. The loaded drum was not rolled, however, since rolling would form undesirable spherical agglomerates.

2.4 Binders

The binder used in the pressing of the carbonaceous ingredients was Varcum 8251, a thermosetting resin, obtained from the Varcum Chemical Division of Reichhold Chemicals, Inc. Varcum is a partially polymerized furfuryl alcohol having viscosity-temperature characteristics as shown in Table IV. The liquid Varcum was stored in 55-gal drums at 10°C prior to use to minimize aging. Maleic anhydride was used as the catalyst for the resin in the amount of 4 grams of maleic anhydride to 100 cc of Varcum.

TABLE IV
VISCOSITY CHARACTERISTICS OF VARCUM 8251 RESIN

Temperature, °C	Viscosity, Centipoises
20	306
30	136
40	75
50	46

During the manufacture of the Varcum resin, its viscosity was adjusted step-wise with furfuryl alcohol additions. Samples of resin of various viscosities were obtained and used as binders for some pressings.

The pitch binder used was Barrett Vac-30, a common coal tar pitch used in the graphite industry.

A second thermoplastic binder used was acenaphthylene which in many respects is similar to pitch. The acenaphthylene, designated 90% pure, was obtained from Henley and Company, New York, N. Y.

3 EQUIPMENT

3.1 Blenders

A Patterson-Kelly 8- or 16-qt twin-shell blender equipped with an intensifier bar and liquid addition apparatus was used to mix dry ingredients and to blend dry ingredients with the liquid binder.

A 16-qt blender and mix in the blender are shown in Fig. 1.

A few batches were mixed in a 1-gal capacity Baker-Perkins sigma blade mixer, MD-1, shown in Fig. 2. This mixer was equipped with facilities to heat the mixing chamber and to pull a vacuum on the chamber for solvent removal.

3.2 Chopper

A Hobart Model 4532 meat chopper made by the Hobart Manufacturing Company, Troy, Ohio, was used to homogenize and densify the pressing mix. This chopper is shown in Fig. 3.

Fig. 1. Patterson-Kelly 16-qt twin-shell blender.

Fig. 2. Baker-Perkins sigma blade mixer.

Fig. 3. Hobart chopper shown with mix.

3.3 Extrusion Press

The mix for certain hydrostatic pressings was homogenized, de-aired, and further mixed by being extruded. The extrusion was done using a 100-ton horizontal extrusion press, shown in Fig. 4, equipped with a 6-in.-diam by 18-in.-long extrusion chamber.

Fig. 4. 100-ton extrusion press.

The chamber was normally held at 45°C through the use of electric cartridge heaters in the chamber wall. The extrusion die had a reduction ratio of about 65 to 1 and produced a multiple-hole shape.

A sliding vacuum sleeve was fitted to the ram of the extrusion press so that the extrusion chamber and the charge could be evacuated prior to and during extrusion. A Kinney KC-5 vacuum pump was used for this purpose. The pump was protected from dust and volatiles with a Dollinger 10-25 filter and a liquid nitrogen cold trap.

3.4 Vibrating Table

The loose material from the meat chopper was tamped into the pressing sacks using a Syntron VP-55 vibrating table. The table had a 20 by 20 in. vibrating surface and operated at 1458 cycles/min.

3.5 Pressing Sacks

The pressing sacks and lids were furnished by the Plastics Section of the Los Alamos Scientific Laboratory (LASL). They were formed by dipping a

mandrel into Plastisol and curing to 175°C for $3/4$ hr. Most sacks were 5 in. in diam and about 7 in. high, although other sizes were used.

3.6 Hydrostatic Press

The filled sacks were pressed by hydrostatic oil pressure in a pressure chamber 8 in. i.d. and 24 in. deep (Fig. 5). The oil was pressurized using an air operated pump (Hydraulic Engineering Corporation Model SC-10-600-30). The normal compaction pressure was 20,000 psi.

3.7 Curing Oven

The pressed pieces were cured in an electrically heated, circulating air oven capable of operating at 450°C . The oven, Model FO 962430, was made by the Cooley Electric Manufacturing Corporation and is shown in Fig. 6 along with the containers for the pressings. A cam-operated Wheelco controller was used to provide the desired heating cycle, usually to 250°C .

Fig. 5. Hydrostatic pressing chamber.

Fig. 6. Curing oven and pressing containers.

3.8 Baking Furnace

The cured pieces were loaded into a stainless steel can and baked to 800°C in a vacuum baking furnace designed and built at LASL (Fig. 7).

The furnace was heated by resistance heating elements controlled by a cam-operated Wheelco controller. The hot zone of the furnace was 16 in. in diam and 60 in. long; the furnace normally operated under a pressure of 10 torr or less. The vacuum system consisted of a two-stage Nash Hytor pump: two Model TS-10 pumps connected in series but powered by a single 10-HP electric motor. An air operated valve in the vacuum line prevented air from being drawn into the furnace through the vacuum pump in case of a power failure. Also, a self-opening trap prevented water from the vacuum pump from entering the furnace.

3.9 Graphitizing Furnace

The baked pieces were placed in a graphite crucible and graphitized in an induction heated graphitizing furnace designed and built at LASL. The induction coil was powered by a 175-kW, 960-cycle MG set. The 82-in.-long coil consisted of flat copper strips, 6 in. wide, bent and silver-soldered together to form a cylinder. Three parallel copper tubes to carry cooling water were silver-soldered to the outside of the coil. The crucible containing the pressings was located within a graphite susceptor 18.75 in. o.d. and 16.62 in. i.d. The susceptor, in turn, was surrounded by a 3-in.-thick layer of carbon black insulation, a 1-in.-thick ceramic liner, a layer of Fiberfrax, and the induction coil. The temperature in the furnace was measured optically through a sight port in the top of the furnace that sighted into a hole in the graphite crucible. A helium flush kept the sight port open and prevented the graphite in the furnace from oxidizing. The graphitizing furnace is shown in Fig. 8.

4. PROCEDURE

4.1 Preparation of Materials

When pressings were made using a standard mixture of 85 parts graphite flour to 15 parts Thermax, the dry ingredients were blended together in large quantities prior to blending with the binder in the P-K blender. The proper quantities of graphite flour and Thermax to make a 100-kg batch were weighed and placed in a 55-gal mixing drum containing a baffle of expanded metal to facilitate mixing. The loaded mixing drum was rolled at 40 rpm for 8 hr, and samples were taken for radiographic examination and chemical analysis.

Fig. 7. Vacuum baking furnace.

Fig. 8. Induction graphitizing furnace.

When pressings were made using nonstandard ratios of graphite flour to Thermax, the individual dry constituents were placed in the P-K blender and blended for 5 min using the intensifier bar prior to the liquid addition.

The binder resin was stored in 55-gal drums. The drum being used was rolled in the cold storage area at 40 rpm for 2 hr to homogenize the material prior to withdrawal. The quantity of resin required for a day's work was withdrawn from the drum the evening before use and placed in an oven at 35°C overnight.

The maleic anhydride catalyst was ground and screened to -20 mesh prior to use, and 4 grams of catalyst per 100 cc of resin was added to the resin. The mixing was performed in 1-gal batches; the mix was heated to 40°C and stirred actively for approximately 20 min prior to use. Maleic anhydride is a slow-acting catalyst at 40°C so the processing times were not critical.

4.2 Blending

The dry ingredient (graphite flour-Thermax) charge for the 8-qt P-K blender was 3 to 3.5 kg; the charge for the 16-qt blender was 6 kg.

The dry ingredients for a mix were weighed and placed in the blender. If the ingredients had not been premixed, they were blended for 5 min using the intensifier bar. The proper amount of catalyzed resin was weighed and placed in a glass reservoir which was wound with a heating tape and had a tube leading to the liquid addition apparatus of the P-K blender. The resin was heated to approximately 40°C, then, over a period of 15 to 20 min, was added slowly under 1-psi air pressure to the dry mix through the intensifier bar of the blender. After the resin addition was completed, the blender was stopped, the lids were opened, and the lids and blender walls were scraped to remove adhering material. The blender was then sealed and rotated for another 5 min with the intensifier bar operating. The blended material had the consistency of wet sand.

The Baker-Perkins sigma blade mixer was used in mixing batches bonded with a thermoplastic binder. The dry carbon ingredients were placed in the mixer and blended. The binder (pitch or acenaphthylene) was dissolved in trichloroethane and slowly added to the blending dry ingredients. Mixing proceeded for 10 min, and then the mix was heated slightly and evacuated to remove the solvent. The mix was screened through a 14 mesh screen before pressing.

4.3 Chopping

The meat chopper was used to homogenize and heat (by friction) the pressing mix if the mix was to be extruded. The normal procedure was to pass the mix through the chopper twice before, and once after, extrusion. The ground mix was the material loaded into the pressing sacks.

Some pressings were made using mix which was not extruded but only ground. Such a mix was passed through the meat chopper four times before being loaded into the pressing sacks. Normally the meat chopper is fitted with a chopper plate having 3/16-in.-diam holes. A plate with 1/8-in.-diam holes was used for mixes which were ground but not extruded.

4.4 Extrusion

The extrusion procedure was as follows:

1. The material was loaded by hand into the extrusion chamber of the extrusion press, and the ram was

advanced until the vacuum sleeve engaged the extrusion chamber.

2. The extrusion chamber, die, and extrusion mix were evacuated to an absolute pressure of 1 mm or less. This vacuum was held for 3 min prior to extrusion.

3. The mix was extruded at a rate of 20 ft/min at a pressure of approximately 3000 psi.

4. The extrusions were ground in a meat chopper, and the ground material was loaded into the pressing sacks.

4.5 Hydrostatic Pressing

The parts which comprise a pressing sack are shown in Fig. 9. These include the Plastisol sack and lid, clamping ring, Aero Seal clamp, weight, hypodermic needle, sponge (shown in the mirror), and mix. The pressing sacks were filled with the loose blended or ground mix, and the mix was packed lightly by tapping the filled sacks on the bench top. More loose material was added, and the procedure was repeated several times until no compaction was noted. The sack full of material was then placed on a vibrating table, a 1500-gram, 5-in.-diam weight was placed on top of the material, and vibration was started. The duration of the vibration was usually 5 min. The sack was then removed from the vibrator and the weight extracted.

The sack lid was pierced with a No. 16 hypodermic needle, and a 7/8-in.-diam by 1/2-in.-thick sponge was affixed to the needle point. A clamping ring 4-3/4 in. o.d., 4-1/2 in. i.d., and 1 in. wide was placed around the lid, and the lid was seated in the sack. The sack was sealed mechanically using an "Aero Seal" hose clamp. The sack and its contents were evacuated through the protruding hypodermic needle (Fig. 10), usually for 1 hr. The needle was then extracted from the lid, and the hole was sealed with a solution of polyvinyl-chloride in equal parts of methyl ethyl ketone and acetone.

The loaded and evacuated sacks were placed in the pressure chamber, oil was added, and the chamber was closed. The air-operated oil pump was started, and the chamber was pressurized to the required pressure. The pressure was immediately released and the cham-

Fig. 9. Parts comprising a pressing sack.

Fig. 10. Filled pressing sacks being evacuated.

ber opened. Oil was removed from the exterior of the sacks with hexane before they were opened.

4.6 Curing

The green pressings were placed in a stainless steel can having a loose fitting lid, and the loaded can was placed in a Cooley oven. The curing operation consisted of gradually heating the pressings to polymerize the thermosetting binder. The heating cycle required 63 hr to go from 45° to 250°C. The heating rate was especially slow, 1°C/hr, from 90° to 110°C while many of the volatiles from polymerization were released. After curing, the pressings were strong enough to be handled without danger of cracking.

4.7 Baking

The cured pressings were then baked to carbonize the binder and further remove gases and volatiles. Baking consisted of heating the 250°C-cured pressings from room temperature to 800°C in a controlled cycle totalling 48 hr. The pressings were baked in a stainless steel can with a loose fitting graphite lid.

Pressings which were bonded with a thermoplastic binder were sand baked to 650°C prior to graphitizing. These pressings were placed in a stainless steel can, with foundry sand packed around them and heated in a vertical tube furnace to 650°C over 48-hr.

4.8 Graphitizing

The baked pressings were placed in a graphite crucible which, in turn, was placed in the induction heated graphitizing furnace. The pressings were heated to 2500°C over 4-1/2 hr and held at temperature for 1 hr. A helium atmosphere was maintained in the furnace during this period and during the cooling cycle which took about 1-1/2 days.

4.9 Further Processing

The pressings were machined into right cylinders, measured, and weighed, and their densities calculated. They were then sent to the Nondestructive Testing Group where they were radiographed. Compression samples, 3/4 in. diam by 3/4 in. long, were then cored from the pressings. One radial and one tangential sample were extracted from each pressing tested at room temperature.

5. DISCUSSION OF RESULTS

The first pressings were made in various sized pressing sacks, and were at times cracked or laminated. Also, different sack loading techniques produced some variation among pressings made with similar mixes. Despite these variables, some of the early data are interesting and therefore are tabulated in relation to the major variable introduced in the mix without reference to minor variables such as sack size or tamping method. All densities and properties are for material which has been graphitized at 2500°C.

The pressings tabulated in Tables V and VI were made with S-97 and M-2 graphite flour mix containing various amounts of wood flour and binder. For comparison one batch may be listed more than once in a table or in more than one tabulation.

A series of M-2 pressings containing 2.5 pph wood flour was made in which the Varcum binder was diluted with a solvent, Freon TF, prior to being mixed with the dry carbon ingredients. The solvent

TABLE V
EFFECT OF WOOD FLOUR ON GRAPHITE PRESSINGS

Batch No.	Varcum Binder, pph	Wood Flour, pph	Density, g/cc	Compressive Strength, psi	
				I	II
S-97 Graphite					
1937AA	18	1.25	1.497	2640	2315
1939AA	18	2.5	1.471	2125	2135
1933AA	18	4.0	1.396	1685	1715
M-2 Graphite					
1928AA	20	0	1.694	5360	5430
1967AA	20	1.25	1.661	4720	4845
1948AA	20	2.5	1.605	3555	3547
1926AA	20	4.0	1.383	1380	1710

TABLE VI
EFFECT OF BINDER CONTENT ON GRAPHITE PRESSINGS

Batch No.	Varcum Binder, pph	Wood Flour, pph	Density, g/cc	Compressive Strength, psi	
				I	II
S-97 Graphite					
1936AA	16	4.0	1.388	1450	1545
1933AA	18	4.0	1.396	1685	1715
1925AA	20	4.0	1.400	1900	1990
M-2 Graphite					
1950AA	16	2.5	1.507	1300	1520
1948AA	20	2.5	1.605	3555	3547
1953AA	22	2.5	1.612	4650	4590
1984AA	30	2.5	1.681	5580	5570

plus binder content for all mixes was 20 pph; the data are reported in Table VII.

TABLE VII
EFFECT OF BINDER AND SOLVENT CONTENT
ON M-2 PRESSINGS

Batch No.	Varcum Binder, pph	Freon TF, pph	Density, g/cc	Compressive Strength, psi	
				I	II
1956AA	10	10	1.383	315	373
1954AA	12	8	1.411	415	470
1964AA	16	4	1.529	1957	2010
1962AA	18	2	1.564	2175	2595

Pressings were made using the M-2 graphite flour mix bonded with a thermoplastic binder, either acenaphthylene or coal tar pitch. The batches were solvent mixed in the Baker-Perkins sigma blade mixer, and at the end of the mixing cycle the mixing chamber was evacuated to draw off the solvent. These pressings were not cured, but were sand baked to 650°C prior to graphitizing. Data for these batches are shown in Table VIII.

TABLE VIII
PROPERTIES OF M-2 PRESSINGS BONDED WITH
A THERMOPLASTIC BINDER

Batch No.	Binder	Binder, pph	Density, g/cc	Compressive Strength, psi	
				I	II
1988AA	Acenaphthylene	25	1.338	465	440
1990AA	"	30	1.348	1800	1600
1991AA	Pitch	25	1.357	310	385
1992AA	"	35	1.425	1363	1615

It can readily be seen that these early experiments did not produce dense or strong pressings. The density was purposely lowered in the pressings with the wood flour additions to produce a porous body. The wood flour did produce porosity but seemed to reduce the compressive strength disproportionately. It was also evident that the binder content greatly influenced the strength.

It was noted that the best pressings were produced if the compacted length-to-diameter ratio was less than 1. Also, a uniform sack loading procedure described earlier was adopted for subsequent pressings.

These procedures, along with a consistent mixing, pressing, and heat treating technique, made systematic investigation of known mix and processing variables possible.

One of the first items investigated was the effect of binder content on a standard 85 wt/% M-2 graphite flour, 15 wt/% Thermax mix. The results presented in Table IX showed that the density increased as the binder content increased, but that the strength was greatest at ~ 30.0 pph binder and then decreased. A similar binder series was made using a mix consisting of 85 wt/% S-97 graphite flour and 15 wt/% Thermax, and the data are tabulated in Table X.

TABLE IX
EFFECT OF BINDER CONTENT ON M-2 PRESSINGS

Batch No.	Binder, pph	Density, g/cc	Compressive Strength, psi	
			I	II
2225AA	20.0	1.636	3625	3618
2226AA	22.5	1.658	5127	4627
2227AA	25.0	1.673	5960	5970
2228AA	27.5	1.693	6285	5770
2229AA	30.0	1.694	6085	6017
2230AA	32.5	1.716	5567	5757
2231AA	35.0	1.722	5443	5400

TABLE X
EFFECT OF BINDER CONTENT ON S-97 PRESSINGS

Batch No.	Vaccum Binder, pph	Density, g/cc	Compressive Strength, psi	
			I	II
2232AA	20.0	1.553	3652	3662
2233AA	22.5	1.536	3240	3145
2234AA	25.0	1.574	4430	4270
2235AA	27.5	1.605	5245	5218
2236AA	30.0	1.617	5500	5465
2237AA	32.5	1.642	5885	5880
2238AA	35.0	1.687	7570	7600

As with the M-2 mix, the density increases with binder content. However, in contrast to the M-2 mix, the strength increases continuously with increasing binder content.

M-2 graphite flour had been used for various purposes with the result that different ratios of flour to Thermax had been investigated. The 85-to-15 ratio was found to yield a product having the highest density. Such an investigation had not been made with the S-97 graphite flour. Consequently, a series of pressings was made in which the Thermax content was varied. All pressings were bonded with 30 pph Vaccum, and gave the results shown in Table XI.

It is interesting that both the density and strength tend to increase with increasing Thermax content even to relatively high Thermax levels.

The densities of hydrostatic pressings prepared with standard mixes were very low compared with the densities of extrusions made with the same mix.

TABLE XI
EFFECT OF THERMAX CONTENT ON S-97 PRESSINGS

Batch No.	Thermax, pph	Density, g/cc	Compressive Strength, psi	
			I	II
2239AA	0	1.493	3200	3360
2240AA	2.5	1.504	3725	3675
2241AA	5.0	1.518	3900	3975
2242AA	7.5	1.546	4060	3985
2243AA	10.0	1.589	4825	4910
2244AA	12.5	1.585	4970	4870
2236AA	15.0	1.617	5500	5465
2245AA	17.5	1.632	4865	4855
2246AA	20.0	1.665	5325	5340
2247AA	22.5	1.688	6945	6895
2248AA	25.0	1.713	6310	6295

The improved densities were apparently caused by the evacuation of the mix prior to extrusion and the working of the mix during extrusion. Test samples were prepared by extruding the mix and passing the extrusions through a meat chopper prior to hydrostatic pressing to determine if the improved properties of the extrusion process could be realized in hydrostatic pressings. The results are shown in Table XII.

TABLE XII
EFFECT OF EXTRUDING A PRESSING MIX

Batch No.	Processing	Density, g/cc	Compressive Strength, psi	
			I	II
G-97 Flour Mix, 30 pph Binder				
2236AA	Not extruded	1.617	5500	5465
2250AA	Extruded	1.782	7240	7445
M-2 Flour Mix, 27.5 pph Binder				
2228AA	Not extruded	1.693	6285	5770
2274AA	Extruded	1.800	9215	8995

These data show that extruding a mix prior to pressing produces a dramatic increase in density and compressive strength.

The possibility of obtaining a more uniform pressing mix by adding a small amount of solvent to the carbon mix or binder to improve the wetting characteristics of the binder was explored. All of the mixes

were extruded and reground before being pressed; the solvent was probably removed when the mix was evacuated prior to extrusion. The solvent was added directly to the dry carbon ingredients in some batches and was mixed with the Varcum binder in others. Three different solvents were used: Philsolv (Ph), Chlorothene (Ch), and Freon TF (Fr). The results of these experiments are shown in Table XIII.

Using a solvent to achieve a better mix did not result in a clear-cut improvement. There was, possibly, a trend toward stronger pressings with a small solvent addition at the expense of reducing the density slightly. Strengths were definitely higher when the solvent was added to the binder rather than to the dry ingredients.

It has been shown that extruding and regrinding a mix prior to pressing substantially increased the density and strength of the pressing. The extrusion operation was quite time consuming so we wished to determine if the pressing mix could be densified by grinding alone. The meat choppers can be fitted with breaker plates having different hole sizes, thus determining to some extent the amount of work put into the mix.

Pressing mixes were made using the M-2 graphite mix and 27.5 pph Varcum binder. These mixes were passed through the chopper four times prior to being pressed. The first mix, 2609AA, was ground and then reblended in the P-K blender for several minutes before being pressed. The data on ground mixes are presented in Table XIV.

Grinding a pressing mix several times prior to pressing appears to produce pressings having strengths and densities similar to those produced with an extruded mix. Reblending the mix after grinding did not improve compressive strength.

At the start of this program, pressings were made at various hydrostatic pressures. Pressure seemed to have little effect once a reasonably high pressure was obtained. A reasonable pressure was based on previous die molding work on similar graphite mixes which indicated that the product density varied little with pressures above 7000 psi. Of course pressures which would crush the graphite particles would affect the product density. The 20,000-psi

TABLE XIII
EFFECT OF A SOLVENT ON M-2 PRESSINGS.

Batch No.	Solvent	Solvent, pph	Solvent Added to Dry Mix	Solvent Added to Binder	Density, g/cc	Compressive Strength, psi	
						I	II
2300AA	Ph	1		X	1.796	8865	8715
2275AA	"	2		X	1.791	9675	9595
2276AA	"	"	X		1.791	8715	8815
2601AA	"	3		X	1.800	8600	8600
2602AA	"	4		X	1.782	8485	8430
2279AA	"	5		X	1.804	8455	8710
2280AA	"	10		X	1.779	5995	8660
2278AA	Ch	2		X	1.787	9310	9250
2277AA	"	"	X		1.786	9045	8835
2273AA	Fr	"		X	1.791	9405	9290
2272AA	"	"	X		1.805	8700	8815

TABLE XIV
EFFECT OF GRINDING A PRESSING MIX

Batch No.	Breaker Plate Hole Size, in.	Density, g/cc	Compressive Strength, psi	
			I	II
2609AA*	3/16	1.792	6495	6680
2608AA	3/16	1.782	8005	7950
2610AA	1/8	1.810	8005	8210

*Reblended in the P-K blender after grinding.

hydrostatic pressure selected for most of this work was felt to be high enough to produce reasonable densities but low enough not to tax the capability of the equipment which had a pressure limit of 30,000 psi.

Two pressings were made at pressures other than 20,000 psi using the M-2 graphite mix and 27.5 pph Varcum binder. The results are tabulated along with data on a standard pressing (Table XV) and show that the compressive strength parallel to the pressing direction increases with pressing pressure. However, the data indicate that the maximum compressive strength perpendicular to the pressing direction is obtained near 20,000 psi pressure. It should be remembered that the table shows the results of only one pressing at each pressure. Because the strengths at the higher pressure are all quite similar, a repeat of this experiment may produce results which show a different trend.

TABLE XV
EFFECT OF HYDROSTATIC PRESSURE ON M-2 PRESSINGS

Batch No.	Pressure x 1000 psi	Density, g/cc	Compressive Strength, psi	
			I	II
2605AA	10	1.787	8290	8650
2274AA	20	1.800	9215	8995
2604AA	30	1.800	8980	9140

One of the graphite fillers investigated briefly in relation to hydrostatic pressing was Carbon Products' high CTE flour described previously. Pressings which contained 85 parts flour to 15 parts Thermax were made using different binder contents.

These pressings were considerably stronger than those made from any other flour (see Table XVI). Although this flour is considerably different in character from the other flours, there is no obvious explanation for the higher strengths.

TABLE XVI
HIGH CTE GRAPHITE FLOUR PRESSINGS

Batch No.	Varcum Binder, pph	Density, g/cc	Compressive Strength, psi	
			I	II
2283AA	20.0	1.792	13950	14285
2284AA	25.0	1.820	12715	13265
2281AA	27.5	1.809	13725	14015

The CTE of batch 2284AA was measured by the LASL Materials Evaluation Group over a range of 25° to 2150°C. The average transverse CTE was $5.3 \times 10^{-6}/^{\circ}\text{C}$, and the average longitudinal CTE was $5.6 \times 10^{-6}/^{\circ}\text{C}$. The CTE of an M-2 pressing, batch 2274AA, was $4.0 \times 10^{-6}/^{\circ}\text{C}$ transverse and $4.2 \times 10^{-6}/^{\circ}\text{C}$ longitudinal.

Furfuryl alcohol base resins which were more viscous than the standard Varcum became available, and M-2 graphite pressings bonded with these resins were made using the extrusion mixing procedure.

The more viscous binders would not feed through the liquid addition apparatus of the P-K blender. Therefore, the dry ingredients and the binder were mixed together by hand and then blended for 10 min in the P-K blender using the intensifier bar.

Although the more viscous resins were hard to work, they did produce pressings having superior properties (Table XVII).

Hydrostatic pressings of M-2 graphite mix bonded with 27.5 pph Varcum were made to evaluate the

method of adding the Varcum to the P-K blender.

The procedure followed in making most of the pressing mixes was to blend at room temperature taking about 15 min to add the liquid binder to the dry ingredients. The Varcum and mix temperatures and the Varcum addition time were varied with the results shown in Table XVIII; all mixes were extruded prior to pressing.

These results are inconclusive but certainly indicate that the mixing conditions represent a variable that can no longer be ignored and suggest that further experiments would be worthwhile. The results are opposite to what might be predicted in that short addition times at room temperature should not produce as thorough a mixing action as longer addition times with warm ingredients, yet the densities and strengths are higher for the short addition times at room temperature. Compared to other pressing experiments, the densities are high but the strengths are not exceptional.

TABLE XVII
PROPERTIES OF EXTRUSIONS MADE WITH VISCOUS VARCUM

Batch No.	Designation	Varcum, pph	Varcum Viscosity, cps	Density, g/cc	Compressive Strength, psi	
					I	II
2264AA	063X07	25	461	1.786	8755	8970
2263AA	063X06	25	600	1.787	8840	8615
2262AA	063X04	27	1120	1.785	8925	8995
2261AA	063X02	27	2690	1.796	9245	9500
2260AA	063X01	30	21000	1.859	10595	10280

TABLE XVIII
EFFECT OF VARIATIONS IN VARCUM ADDITION CONDITIONS

Batch No.	Variable	Density, g/cc	Compressive Strength, psi	
			I	II
2625AA	Mix and Varcum heated to 45°C before blending.	1.778	7040	7380
2629AA	Varcum heated to 50°C while blending. Added in 9 min.	1.794	6350	7350
2628AA	Mix at RT. Varcum added in 10 min.	1.815	7900	7900
2626AA	Varcum added in 9 min 20 mil shims*	1.821	8400	8050

*Shims determine the opening in the intensifier bar through which the Varcum sprays. These are normally 10 mils thick.

Another variable which might affect the quality of the pressings is the length of time that the filled pressing sacks are evacuated prior to pressing. Times other than the normal 1 hr were tried for standard M-2 pressing with the results shown in Table XIX.

TABLE XIX
EFFECT OF EVACUATION TIME ON M-2 PRESSINGS

Batch No.	Evacuation Time	Density, g/cc	Compressive Strength, psi	
			I	II
2620AA	0	1.777	7040	6830
2621AA	0	1.768	7325	7180
2619AA	30 min	1.786	8055	8025
2622AA	2 hr	1.786	7380	7475

Again the results are inconclusive. Both densities and strengths are lower than for a standard batch, but a standard batch made at this particular time might have given different results.

6. SUMMARY

Hydrostatic pressing has been shown to be a satisfactory technique for fabricating fine grained, isotropic graphite. The composition of the pressing mix, especially the wood flour and binder content, has been shown to have a large effect on the properties of the pressing. Processing variables, primarily mixing procedures, also greatly affect the properties.

It is quite obvious that reproducibility of pressings made with similar mixes but at different times is not so good as it should be and that there are unknown variables which have an appreciable effect on the pressing quality. However, a series of pressings made at one time for a particular experiment does show definite and valid characteristics.

7. ACKNOWLEDGEMENTS

The authors wish to thank J. M. Taub and W. W. Martin for guidance in this work. We also wish to thank the Plastics, Ceramics, and Physical Metallurgy Sections of CMB-6 and the Shops Department for supporting work; W. C. Erickson for help in preparing the report; and J. D. Ellis for typing.