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A STUDY OF SOME MANUFACTURING
PARAMETERS AS THEY RELATE TO THE "C"
DIRECTION THERMAL CONDUCTIVITY OF INSULATING TILE

MASTER

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A STUDY OF SOME MANUFACTURING PARAMETERS
AS THEY RELATE TO THE "C" DIRECTION
THERMAL CONDUCTIVITY OF INSULATING TILE *

INFORMATION CATEGORY

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ABSTRACT

Fourteen tests were conducted for evaluating the effect of bow, and taper on the "c" direction thermal conductivity of insulating tile manufactured from pyrolytic graphite. It was established that no change resulted on tilting the basal planes at an angle of $0^{\circ}45'$. Increasing the angle to $1^{\circ}15'$ resulted in a 28% increase, and an angle of $2^{\circ}30'$ increased the conductivity by 44%. This data permits a relaxation of the present bow and taper limitations as follows. If the bow of the first deposited surface is less than .008", the taper may be up to 20% of the blank thickness. If the bow is .008" - .015" the taper must be less than 10%. The taper of a "blank" having a bow of .015" - .020" should not exceed 7.5%.

Two thermal conductivity tests were conducted for studying the effects of spalled first deposited surfaces, which displaces the tile to the last deposited portion of a tile blank. It was established that on fine grain continuously nucleated material this displacement could result in a 30% increase in the "c" direction thermal properties. On this particular furnace run, HM-14, acceptance tests on which coupons were taken from the first deposited portion were well within the limits of the specification. The 30% increase, resulting from displacing the coupon to the last deposited portion, would have exceeded the specification. It is recommended that no change be made to the present spalling limitations of fine grain continuously nucleated material.

A review of past supplier performance and their manufacturing methods disclosed that 7200 insulating tile, represented by 170 thermal conductivity tests, manufactured from the last deposited portion of six furnace runs of tile blank had met the thermal requirements of the specification. In the case of fine grained continuously nucleated only 75% of the furnace runs met these requirements. Since the substrate nucleated structure appears to be more uniform within a blank, and controllable from furnace run to

furnace run, it has been recommended that only substrate nucleated pyrolytic graphite be utilized for manufacturing insulating tile.

The above combination of relaxed bow and taper limitations, the deletion of spalling limitation on substrate nucleated material, and its simplicity of manufacturing utilizing either deposited surface for basal plane orientation should result in a substantial cost reduction.

INTRODUCTION

It is generally agreed that the thermal conductivity properties of an insulating tile are the prime consideration. The requirements for basal plane parallelism in the finish component is, therefore, recognized as a most important factor in guaranteeing that the basic raw material insulating characteristics have not been weakened during manufacturing. Since basal plane orientation is extremely difficult, if not impossible, to measure, it has been necessary to insure proper orientation of the layers by the combination of raw material procurement and the manufacturing method. This has been accomplished by purchasing tile blank in which the plane of the first and last deposited surfaces are fairly flat and parallel, and obtaining from the first deposited portion of this, an insulating tile, the basal plane orientation of which is referenced parallel to the first deposited surface. The purchasing of a physically suitable tile blank for this approach to manufacturing has been a major problem.

Pyrolytic graphite is produced by passing a hydrocarbon gas, such as methane, over a hot surface held at temperatures of 3500°F to 4500°F under reduced pressure. The carbon atoms are removed from the gas stream by a thermal decomposition process and are deposited on the hot surface (mandrel) in a manner similar to a vacuum plating process. As the material is deposited, the graphite crystals are positioned in such a manner that their "c" axis (cross crystal direction) is perpendicular to the surface and their "a" axis is parallel to it. This orientation of basal planes parallel to the deposition surface results in the material having a very high degree of anisotropy in such properties as mechanical strength, electrical and thermal conductivity, and thermal expansion. The material perpendicular to the deposition surface is an excellent thermal and electrical insulator and mechanically weak, while parallel to the surface, it is a good thermal and electrical conductor, and for graphite, at least, exhibits excellent mechanical properties. In order to obtain the maximum benefit of this anisotropy of properties in a particular shape of pyrolytic graphite, it is necessary to have a mandrel of the desired shape on which to carry out the deposition process.

A major problem in the deposition of flat plate for insulating tile results from the thermal expansion anisotropy. In the direction perpendicular to the surface the material contracts about 20 times greater on cool down from deposition temperature than it does in the parallel direction, which results in the introduction of very high residual stresses. Also, during deposition, the initial layers are exposed to a high temperature for the longest period of time and grow, subjecting the subsequent layers to a tensile stress along the planes and a shear stress perpendicular to them. This places the first deposited surface in compression, and the last deposited surface in tension. The cumulative effect of these residual stress are always sufficient to distort the layer planes, and result in bowed deposits. Machining flat surface from bowed deposits result in the cutting of basal planes which reduces the effective thickness of the insulating properties of the material by heat flow short circuiting along the basal planes.

In conjunction with the above process problem there is generally a severe non-parallelism of the first and last deposited surface. This results from the gas chemistry of the deposition process. The progressive change in concentration and reactivity of the feeder gas as it is depleted and diluted with the products of reaction, deposits carbon in a patterned varying rate. This non-parallelism is a progressive distortion of the layers from the first to the last deposited surface. The degrees of non-parallelism, or taper, is a progressively increasing angle of inclination of the layer planes as deposit thickness increases. Machining of tapered deposits result in the cutting of basal planes and the reduction of the materials effective insulating characteristics.

In addition to the above bow and taper of tile blanks, there is normally a partial or complete spalling of the first deposited surface on about 40% of a furnace load of plate stock. This results from the physically breaking away of the deposited plate from the mandrel. The magnitude of the spalled area is random and can be up to the cleavage of the entire first deposited surface. Partial spalling of over one half of the as deposited surface makes it extremely difficult to reference the first deposited surface for machining

parallel to the basal planes, accordingly, limitations have been placed on the depth and surface area of spalling.

The pyrolytic graphite industries price and delivery schedules are basically dependent on furnace time and as a result is primarily a function of the yield to a customer's requirements. The lower the yield, the higher the unit price. The extreme sensitivity of this relationship is graphically illustrated by a proposal resulting from a price negotiation with one of the supplier's, varying only the bow and taper call outs slightly as follows:

<u>Alternative Proposal</u>	<u>Description</u>	<u>Unit Price</u>
1	15 mil bow on last deposit surface - 10% thickness variation from mid-point	\$20.70 ea.
2	15 mil bow on substrate side - 10% thickness variation	\$18.10 ea.
3	15 mil maximum bow on substrate side - 15% thickness variation	\$11.90 ea.
4	15 mil maximum bow on last deposited side - 15% thickness variation	\$13.50 ea.
5	15 mil bow on substrate side - 20% thickness variation	\$12.90 ea.
6	15 mil maximum bow on last deposited side - 20% thickness variation	\$12.90 ea.
7	10 mil maximum bow on substrate side - 20% thickness variation	\$11.20 ea.
8	10 mil maximum bow on substrate side - 15% thickness variation	\$12.40 ea.

In support of a manufacturing effort to utilize to the fullest the yield of tile blank from a furnace load of pyrolytic graphite plate, a program of evaluation was initiated to establish qualitatively, the relationship of degree of bow and taper to the "c" direction thermal conductivity. In addition, the program was to include evaluation of obtaining the insulating tile from the last deposited surface. By this manufacturing

technique, the degree of spalling of the first deposited surface, would no longer be a consideration in blank procurement.

TEST PROGRAM AND RESULTS

PART I

Evaluation of the Angle of Inclination of Layer Planes (Bow and Taper) to "c"

Direction Thermal Conductivity

A. Forty-eight (48) tile blanks from furnace load HM-14 were machined to .125" x .5" x 2.8" at angles of 0°, 0°45', 1°15', and 2°30' by the following machining sequence, and were tested for thermal conductivity by Southern Research Institute.

1. Using the first deposited surface as a reference, machine last deposited surface flat.
2. Using the flat surface obtained in Step 1 as reference, finish the first deposit surface flat to various angles by removing a minimum amount of material, angle of inclination to be as follows:

<u>Angle</u>	<u>Inches of Inclination/Linear Inch</u>
0°	0.
0°45'	0.013
1°15'	0.022
2°30'	0.044

3. Using the machined surface obtained in Step 2 as a reference, finish the last deposited surface flat to desired thickness.

B. Results of Thermal Conductivity Testing

See Table II (page 18)

TEST PROGRAM AND RESULTS

PART II

Description of Test Program and Results of a Manufacturing Procedure for the Deletion of Spalling as a Procurement Parameter.

- A. Four insulating tile thermal conductivity test specimen .125" x .5" x 2.8" were obtained from the last deposited portions of tile blanks in excess of .250" thickness from furnace load HM-14 by the following machining sequence, and were tested for thermal conductivity by Southern Research Institute:
1. Using the last deposited surface as a reference, machine the first deposited surface flat.
 2. Using the flat surface obtained in Step 1, as a reference, machine flat the last deposited surface removing a minimum of material.
 3. Using the machined surface obtained in Step 2, as a reference, finish machine the first deposited surface to desired thickness.
 4. Finish specimen width.
 5. Finish specimen length.
- B. Results of Thermal Conductivity Testing
See Table III (page 17)

DISCUSSION

1. Angle of Inclination (Bow and Taper)

Table I contains all of the available thermal conductivity data obtained from the first deposited portion of HM-14 tile blank. This consisted of the initial ten (10) acceptance tests and three (3) reruns. Table II includes four (4) tests at 0° and the results of blanks machined at three (3) angles of inclination.

No increase in thermal conductivity resulted from inclining the layer planes at an angle of 0° to $0^\circ 45'$ of the first deposited surface. See Fig. 1. The calculated increase for this change in the angle of inclination should have been about 70%, as illustrated in Fig. 2.

The probable explanation for this no change in thermal properties up to an angle of $0^\circ 45'$ is the equal contribution of the bowed and tapered layer planes to the orientation at both angles. Apparently, a similar actual angle of inclination existed.

As the angle of inclination increased beyond $0^\circ 45'$ the thermal conductivity increased as shown in Fig. 1. At $1^\circ 15'$, it had jumped 28% and at $2^\circ 30'$ it had increased 55%. In both cases the increase was only 25% of that which should be expected had the basal plane orientation been inclined to the calculated angle, as shown in Fig. 1. This again is probably due to the bow and taper contribution to the angle of distortion of the as deposited layer planes. From Figure 3, it can be seen that for an average furnace run of plate stock, a 1° inclining of the layer planes (.018 inch/inch) should not significantly effect the "c" direction thermal conductivity. At this angle, 44% of the layers should have been intersected resulting in a 100% increase in thermal conductivity. Actually, there was only 15% increase in thermal conductivity. This indicates that the actual heat short circuiting by the intersection at a 1° angle is probably only a $15'$ effective angle

of inclination.

The above consideration permits the construction of Fig. 4, which graphically illustrate the parameters of bow and taper as expressed in mils bow and percent nonparallelism of the opposite faces of tile blanks. Experience has indicated that it is rare indeed that bow is in excess of .020" in a 3" tile blank and also that taper in such a short length rarely exceeds 20%, or 13 mils/inch. This permits simplification of the bow and taper limitations as denoted by the arrows, namely, if the bow of the first deposited surface is less than .008", the taper may be up to 20% of the blank thickness. If bow is .008" - .015" the taper must be less than 10%. The taper of a "blank" having a bow of .015" - .020" should not exceed 7.5%.

2. Thermal Conductivity of First and Last Deposited Material (Spalled Blanks)

Table II contains the thermal conductivity of the four additional specimen machined from the last deposited portion of HM-14 blanks 250 - 300 mil thick. Fig. 5 illustrates that the thermal conductivity of tile obtained from the last deposited surface is approximately 30% higher than for those obtained from the first deposited surface. As noted in Fig. 6, HM-14 was one of those furnace runs of material having exceptionally low thermal conductivities, however, when the test specimen was obtained from the opposite portion of the blank, this excellent heat of material exceeded requirements of the specification. The relationship of thermal conductivity to specimen location in the blank is probably explained by the structure variation shown in Fig. 7. The first deposited surface displays all of the characteristics which probably contribute to maximum insulating characteristics as a low rate of nucleation and gross micro cracking. The last deposited surface, however, has a very highly regenerative structure with very few if any micro cracks, both of which tend to increase the thermal conductivity. Actually there appears to be three zones of structural difference across this cross section, going from a coarse grained regenerative structure with numerous cracks to a fine grain regenerative structure with a few micro cracks to finally, a fine grained regenerative structure with no visible cracks.

In the past it has been generally observed that thermal conductivity in the "c" direction decreases with decreasing regeneration. This is graphically illustrated in Fig. 6, which is a summary of the averages and the ranges of thermal conductivity testing conducted at Southern Research Institute on tile for WANL. It is obvious that the generally fine grained, highly regenerative structure of HTM is frequently in trouble as to thermal conductivity, while the substrate nucleated structure of Super Temp and the coarse regenerative of G.E.'s is generally lower and within limits. The typical microstructures of these manufacturers are shown in Fig. 8. It is most apparent that the likelihood of micro cracking increase as the structure coarsens, and it is probable that these cracks act as effective thermal dams, at least at the temperature of the testing. This it is believed contributes effectively to the inherently lower thermal conductivity of the latter two (2) types of structures.

It has been established fairly well that the regenerative type materials strength are significantly higher than substrate nucleated, the strength increasing as the grain size decreases. It is believed that during deposition the initial layers deposited, which are exposed to temperature for the longest period of time, grows permanently in the "a" direction, and contracts in the "c" direction inducing growth stresses in the first deposited portion. As stated earlier this contributes to the bowing of the plate deposits. Cracking appears to be also dependent on the magnitude of these growth stresses. In general, the coarser the grain, the weaker the material the greater the degree of micro-cracking, and the lower the thermal conductivity. This is probably true to a degree at any deposition temperature which is related most importantly to thermal conductivity. The lower the temperature of deposition, the lower the thermal conductivity. There are of course, practical limitations to decreasing the temperature, as the possibility of sooted deposits increase as the temperature decreases.

From the above it appears that excessively deep spalling, which would move the obtaining of an insulating tile to the last deposited portion of a highly regenerative tile blank is not permissible. Accordingly, spalling depth should be restricted to about 15% of the tile blanks thickness, and no more than half the surface area.

Referring to Fig. 6, it is observed that six furnace runs of substrate nucleated plate stock have been tested for thermal conductivity. In no instance did the average of the ten (10) tests per furnace run exceed the limits of the specification at the 2000° test temperature.

These specimens and the 7200 insulating tile they represented were obtained from the last deposited portion of the tile blank, having been manufactured from plate stock by referencing the last deposited surface for basal plane orientation, removing only a minimum of stock, from this surface, and finishing to desired thickness by machining the first deposited surface.

This certainly appears to establish that the thermal properties of substrate nucleated plate are not sensitive to specimen location. On this type of structure, the degree or magnitude of spalling should not be a consideration, as the above demonstrates.

3. Economic Consideration

Relaxation from the present .015" bow and 10% taper limitations to the suggested sliding scale of up to .020" bow and 20% taper should result in a substantial cost reduction as illustrated by the price negotiation related in the introduction, the cost of tile blanks is extremely sensitive to furnace yield. A small change in acceptance criteria having a dramatic effect on price.

In addition, several economic advantages should result from the suggested change of microstructure from the present continuously nucleated to the substrate nucleated as

follows:

- A. Deletion of the limitation on spalling of the first deposited surface to the extent that the minimum section thickness of a spalled blank is within the drawing tolerance should result in a substantial increase in furnace yield.
- B. A definite advantage to manufacturing may be anticipated by permission to use that deposited surface which is a continuous deposit for basal plane orientation referencing.
- C. The change of microstructure should place tile blank procurement on a competitive basis. Presently, only one supplier has production experience in producing continuously nucleated plate. In addition, production processes have been successfully developed for producing the substrate nucleated plate in up to 36" furnaces yielding 7000 square inches per furnace run as compared to only 4000 square inches from 29" furnaces for the continuous nucleated structure. The above favorable combination of competition and large furnace processing should result in a cost reduction.

CONCLUSIONS

1. This evaluation indicates that for small angles of inclination, the sensitivity of of the "c" direction thermal conductivity to the basal plane orientation is about 25% of the theoretical value. An angle of inclination of 1° does not significantly effect the "c" direction insulating characteristics.
2. For a highly regenerative type microstructure, the thermal conductivity may increase significantly from the first deposited portion of a tile blank to the last deposited portion. For the furnace lot of material used for this evaluation (HM-14), the first deposited portion had exceedingly low and acceptable thermal conductivity. In the last deposited portion, it increased 30%, exceeding the limits of the specification.
3. A review of the thermal conductivity testing of substrate nucleated insulating tile indicates no significant change in the first or last deposited portions of tile blanks. The specification was not exceeded during the testing of 170 specimen in the manufacture of 7200 insulating tile from the last deposited portion of six furnace runs of substrate nucleated plate. This permits deletion of the spalling limitation if this type of material is utilized.
4. This review also established that the thermal conductivity of the substrate nucleated material is more uniform from furnace lot to furnace lot than the highly regenerative structure.

RECOMMENDATIONS

It is recommended that:

1. If the bow of the first or last deposited surface of a tile blank is less than .008", their non-parallelism (taper) should not exceed 20%.
2. If the bow is over .008" but not greater than .015", the taper should not exceed 10%.
3. If the bow is over .015" but no greater than .020", the taper should not exceed 7.5%.
4. Insulating tile should be machined from the first deposited portion only of fine grained continuously nucleated tile blank, referencing the first deposited surface. Spalling of this surface should not be over half the surface area and depth should be less than 15% of the blanks thickness.
5. Consideration be given to manufacturing insulating tile from substrate nucleated material.

HM-14 ACCEPTANCE TEST DATA (0° INCLINATION)
REFERENCED TO FIRST DEPOSITED SURFACE

TABLE I

<u>Test</u>	<u>Test Identity</u>	<u>Test Temp °F 1000°</u>	<u>Test Temp °F 2000°</u>
1	T1	.53	.40
2	T2	.70	.47
3	T3	.65	.45
4	T4	.66	.41
5	T5	.57	.44
6	B1	.55	.40
7	B2	.71	.48
8	B3	.71	.45
9	B4	.64	.39
10	B5	.58	.44
11	T1-2	.71	.53
12	T5-2	.71	.49
13	B3-1	.60	.49
Average		.64	.45

THERMAL CONDUCTIVITY OF SPECIMEN OBTAINED FROM
AND REFERENCED TO LAST DEPOSITED PORTION OF TILE BLANK

TABLE III

<u>Test No.</u>	<u>Test Temp °F 1000°</u>	<u>Test Temp °F 2000°</u>
1	.89	.61
2	.84	.60
3	.83	.66
4	.89	.67

INFLUENCE OF "A" DIRECTION CONTRIBUTION TO "C"
DIRECTION THERMAL CONDUCTIVITY WHEN "A" LAYERS ARE INCLINED

TABLE II

<u>Bow</u>	<u>Angle</u>	<u>Test No.</u>	<u>Test Temp °F</u> <u>1000°</u>	<u>Test Temp °F</u> <u>2000°</u>
.017 - .019	0°	1	.73	.59
.013 - .010	0°	2	.70	.59
.014 - .011	0°	3	.71	.56
.006 - .005	0°	4	.69	.55
.006 - .004	0°45'	5	.70	.51
.003 - .000	0°45'	6	.61	.49
.019 - .015	1°15'	7	.91	.69
.015 - .007	1°15'	8	.87	.68
.009 - .012	1°15'	9	.75	.68
.024 - .012	1°15'	10	.78	.68
.016 - .014	1°15'	11	.92	.61
.006 - .004	1°15'	12	.87	.58
.008 - .003	1°15'	13	.87	.60
.003 - .000	1°15'	14	.81	.58
.003 - .004	2°30'	15	.96	.75
.013 - .016	2°30'	16	.95	.76

Figure 1. Influence on "c" direction Thermal Conductivity when "a" Layers are Inclined at Three (3) Angles

Figure 2. Influence of "a" Direction Contribution to Thermal Conductivity of Pyrolytic Graphite when "a" Layers are Inclined (Calculated)

Courtesy Southern Research Institute

Figure 3. Angle of Inclination Vs. Thermal Conductivity

Figure 4. Allowable Bow vs Taper (Based on an allowable $0^{\circ}45'$ angle)

Figure 5. "C" Direction Thermal Conductivity as Related to First and Last Deposited Portion of a Tile Blank

THERMAL CONDUCTIVITY
BTU" s - Ft/Hr - Ft²

Figure 6. Supplier Thermal Conductivity Performance Chart (at 2000°F)

FIRST DEPOSITED SIDE

WANL-TME-833

Low Rate of Nucleation
Numerous Cracks
Lower Resultant
Thermal Conductivity
Maximum "c" Orientation

High Rate of Nucleation
Few continuous Cracks but
intracone Cracks Evident

Lines not normal to Cones are Result
of Overlay of smaller Photomicrographs

High Rate of Nucleation

Very few continuous and intracone
Cracks Evident

Higher Conductivity Due to Absence
of Cracks and Contribution of "a"
Layers

0.29"
Approximately

LAST DEPOSITED SIDE

Figure 7. Photomicrograph at about 30X Magnification of Cross Section of Pyrolytic Graphite Tile
from High Temperature Materials Run HM-14 - Courtesy Southern Research Institute

Substrate Nucleated with
Microcracks (G.E.)

Substrate Nucleated with Microcracks
(STC)

Continuously Nucleated Fine Grain (HTM)

Continuously Nucleated with Microcracks (HTM)

Figure 8. Manufacturer's Typical Plate Microstructures (75X)