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# GRAPHITE TRIPLE POINT AND SOLIDUS-LIQUIDUS INTERFACE EXPERIMENTALLY DETERMINED UP TO 1000 ATMOSPHERES

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# GRAPHITE TRIPLE POINT AND SOLIDUS-LIQUIDUS INTERFACE EXPERIMENTALLY DETERMINED UP TO 1000 ATMOSPHERES

By Glen J. Schoessow

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## FOREWORD

This report describes research related to advanced nuclear rocket propulsion. The work was performed under NASA Contract NAS3-10412 with Charles C. Masser, Nuclear Systems Division, NASA Lewis Research Center as Technical Manager. The program was helped materially and schedule-wise by Los Alamos Scientific Laboratory which donated the LANG grade test specimens, all machined to dimension and carefully documented.

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## SUMMARY

A pressure cell with a resistance-heated graphite test specimen in a helium atmosphere was used to experimentally determine the triple point of graphite and the solidus-liquidus interface to 1000 atmospheres absolute.

Four grades of graphite were tested for triple-point pressure and temperature. They were Union Carbide AGKS, AGSR, AGOT and Los Alamos Nuclear Grade, LANG. The triple-point pressure was determined to be 103 atmospheres absolute for the four grades. The temperature varied for each grade as follows: $7529 \pm 19^{\circ}R$ for AGOT, $7606 \pm 60^{\circ}R$ for LANG, $7629 \pm 33^{\circ}R$ for AGSR and $7738 \pm 45^{\circ}R$ for AGKS.

The solidus-liquidus interface from the triple point to 1000 atmospheres was experimentally determined for the LANG grade of graphite. The melting temperature increased with increasing pressure from $7645^{\circ}R$ at 103 atmospheres to $7750^{\circ}R$ at 1000 atmospheres absolute based on a linear least-squares fit.

The criteria for determination of melting were the coincidence of visual indication of melt by a post-examination of the sectioned specimen under a microscope, and the occurrence of a rapid voltage rise across the specimen which resulted from an increase in specimen electrical resistance upon initiation of melting.

## INTRODUCTION

In order to evaluate the specific impulse limits of graphite solid core and liquid or gaseous core nuclear rocket propulsion reactors, it is necessary to acquire appropriate information regarding the triple-point pressure and temperature of graphite and the solidus-liquidus interface to 1000 atm. This need is indicated in studies made by Ragsdale, Kascak, and Donovan (Ref.1); by Ragsdale (Ref. 2) and by Ragsdale and Rom (Ref. 3). Since the specific impulse of any rocket is directly proportional to the square root of the exhaust temperature, it is desirable to operate the rocket at the maximum possible temperature. The operating temperature is limited by material considerations. If accurate high-temperature data is available on the materials present in the reactor, the reactor can then be operated closer to its material limits; i.e., at a higher temperature.

Quite apart from the nuclear rocket propulsion application, graphite has several other demanding applications at high temperatures; such as, ablative materials, structural materials, power reactor fuel elements, etc. For these applications it is desirable to have reliable information on the pressure-temperature relationship, the triple point for graphite, and on any variations that might exist between several different grades of graphite.

Investigators seeking experimental data on the graphite melting temperature are faced with many difficulties. Several approaches have been tried in order to maintain the desired pressure and to accurately measure the melting temperature (Refs. 4,5,6,7, and 8).

While it is desirable and necessary to present a brief review of the work of previous investigators, it is obviously impossible to summarize in a few lines the expertise of the investigators. The fine work of these scholars will be briefed and used for comparison purposes. The author's apologies are extended for any unintentional inaccuracies in the interpretations of their results.

Basset's work with the graphite phase diagram (Ref. 4) was very well done, but several inaccuracies were inherent in his equipment and procedures. He had a relatively small power supply of 300 amps at 12 to 30 volts which limited him to a specimen 15 mm long with a reduced test section 2 mm diameter by 6 mm long. With such a small specimen he could not provide an interior cavity to obtain true blackbody temperature measurements. An error was thus introduced by using a pyrometer which had been calibrated for a black body to measure a graphite surface temperature. He reported that convection currents in the high-pressure argon presented a considerable problem to his optical high-temperature

measurements. He referred to "fleeting" glimpses of the graphite at high temperatures and pressures. He used the specimens' failure in arc as the primary indication that the melting temperature had been reached. Basset stated that the instant of failure probably involved significant overpressures and transient temperatures and these caused noticeable variations in appearance in specimens even though run at the same pressure. He reported a triple point of 7200°R at 102 atm.

Jones (Ref. 5), using improved equipment and procedures, performed basically the same experiments as Basset. He electrically heated a specimen with a test section 0.125 in. x 0.200 in. x 0.625 in. long. The specimen was mounted in a high-pressure cell filled with argon and heated to failure. The specimen had a shallow hole into the hottest region and was surrounded by a graphite sleeve which was electrically insulated from the test specimen. The sleeve and hole allowed black-body conditions to be more closely approached, but the shallow hole precluded perfect black-body conditions. He made no corrections for carbon vapor effects stating that his experiments indicated such effects were negligible. Since he employed a specimen rupture technique, however, he experienced the same overpressure and transient temperature problems that Basset had observed. Jones reported a triple point of 7250°R at 100 atm.

Noda performed a similar type of experiment (Ref. 6). He used a direct current power supply to melt and fail a 4 mm diameter by 30 mm long graphite rod with a smaller center section. This specimen rupture technique is about the same as the one used by Basset and Jones and it involved the same problems. Noda used a pressure cell filled with argon and a calibrated optical pyrometer. He reported a triple-point pressure of 100 to 110 atm and the triple-point temperature of 7235 $\pm$ 90$^{o}$R.

Bundy (Ref. 7) employed a flash-heating capacitor discharge method using small rods of graphite 0.04 in. diameter x 0.28 in. long. He surrounded the graphite rod with combinations of diamond powder, boron nitride, and pyrophyllite sleeves, and he obtained high-pressure environments by compressing the sleeves between large pistons. He used an energy balance method to estimate the temperature. His main effort, however, was to establish the solidus-liquidus interface from Basset's triple-point data to the diamond triple point. He reported a maximum melting temperature of 8280$^{o}$R at about 65,000 atm.

Fateeva (Ref. 8) heated a specimen 0.8 mm diameter with alternating current until the specimen ruptured. A two-color pyrometer method was used for temperature measurements. The quartz window was extended close to the specimen surface which greatly reduced problems with convection

currents. Although the work appears to have been carefully done, the rupture method was employed with its associated problems as mentioned above. Two-color methods, moreover, assume equal specimen emissivities at the two wave lengths involved, and it was assumed that any absorbing material of quartz, argon, and carbon vapor present between the specimen and the instrument had equal coefficients of absorption at the two wave lengths being employed. Neither of these assumptions is correct. Fateeva reported a triple point of 8370°R. The triple-point pressure was not reported.

The methods used in the present work eliminate or greatly reduce the sources of error present in some of the works mentioned above.

A large power supply with a capability of 3000 amps at 20 volts made it possible to use a specimen with a test section 0.45 in. diameter by 1.0 in. long. This specimen was large enough to permit construction of a nearly ideal black-body cavity in the hottest central volume as shown in Fig. 2. A black-body calibrated monochromatic pyrometer was then used for all temperature measurements. Helium gas was used to pressurize the test cell shown in Fig. 1. An optical tunnel was used to prevent specimen image distortion at the higher pressures as shown in Fig. 3. The center of these large specimens was melted while the outer surface remained solid as shown in Fig. 6. The central cavity temperature

was calibrated against power at temperatures below the threshold for significant amounts of carbon vapor. This calibration, together with the accurately measured power at which melting began, was then used to determine the melting temperature. Use of these procedures largely eliminated the effects of carbon vapor on the measurements.

## EXPERIMENTAL APPARATUS

The pressure cell is shown in Fig. 1 with all cooling water, gas and power connections. The flexible power and water tube shown is necessary for two reasons: it transmits the power and cooling water to the specimen clamp; and, at the same time, provides the necessary flexibility for mounting the brittle graphite specimens in the rigid pressure cell. The specimen clamps and the top and bottom power and water fittings are internally cooled by the water.

Instrumentations included a disappearing filament manual optical pyrometer calibrated by NBS; two balancing-bridge-type recorders to record voltage across and current through the pressure cell; a 3000 amp, 50 mv 0.25% accuracy shunt; calibrated pressure gauges, 15 to 2015 psia and 15 to 15,015 psia; and other instruments associated with the various system's proper operation. In addition, at regular intervals during the test program, an optical pyrometer comparison system was used to check the stability and calibration of the pyrometer used in this work.

## EXPERIMENTAL PROCEDURE: TRIPLE-POINT PRESSURE

The power input to the graphite specimen was increased from a starting power of about 3 kilowatts to the melting power. The power was increased in a smooth, reproducible, programmed manner by using a motor-driven power programmer. The typical test time was seven minutes. Each test was allowed to continue until the recorder measuring the voltage across the pressure cell showed an inflection point and subsequent very rapid rise. This was due to a specimen resistance increase and the current source nature of the power supply used. Extensive operating experience has shown that the voltage inflection point (VIP) represents the initiation of melting and/or sublimation at the graphite specimen's center. That is, if the specimen was removed before the VIP had occurred, no evidence of melt or sublimation was found. If the VIP was allowed to occur, examination of the specimen showed evidence of melting and/or sublimation depending upon the operating pressure.

The triple-point pressure was located by repeating the experiment at various pressures and then visually examining the sectioned specimen under a 120x microscope. The necessary evidence for melting was taken to be the presence of a dendritic-appearing melted region and/or of spheres of graphite which form from melted phase and which are due to the

action of surface tension on a liquid as shown in Fig. 6. These filled spheres formed from the liquid phase are entirely different from the hollow spheres which can be formed from a vapor phase. The two separate types of spheres must not be confused. Basset (Ref. 4) presents a more complete discussion of the various graphite forms present after sublimation has occurred and after melting has occurred. Noda (Ref. 6) presents a discussion of the 1 to 3 mm diameter filled spheres he observed. A specimen that did not show evidence of melting is shown in Figure 7.

The specimen geometry used for these tests is shown in Fig. 2 except that the hole plug and vent hole shown were not added until tests above the triple-point pressure. This geometry was necessary to insure that melting occurred under a known pressure. If the holes had not been present or if they had become plugged during a test, any melting observed inside the specimen occurred under the measured gas pressure plus an unknown vapor pressure plus an unknown pressure from restrained thermal expansion. All tests reported herein were terminated with the radial and axial holes still open to provide pressure relief.

## EXPERIMENTAL PROCEDURE: TRIPLE-POINT TEMPERATURE

These tests used the same specimen geometry as the triple-point pressure tests described above. The radial-hole, axial-hole combination formed a vent system which

served to sweep away much of the otherwise troublesome graphite vapor. The power input was increased by using the same power programmer used for the triple-point pressure tests. The disappearing-filament optical pyrometer was positioned to look into the graphite specimen's 0.090 in. diameter radial hole shown in Fig. 2. The pyrometer was successively set to twelve different temperatures from 4090 to 7380 R. When the radial hole image behind the pyrometer filament became bright enough to cause the pyrometer filament to "disappear," the pyrometer operator pressed a switch which marked the voltage and amperage recorder charts. In this manner a power-temperature calibration was obtained for each specimen.

During some tests, small amounts of a dark-appearing vapor would be observed in the radial hole during the last temperature measurements. This vapor would absorb some of the emerging black-body radiations thus lowering the readings and eventually partially plugging the radial hole. The readings thus affected are not included in the data.

After this dark vapor appeared, the power increase was allowed to continue until the voltage inflection point (VIP) was observed. The power supply would then be turned off before specimen failure occurred. The power input at the instant of the VIP was taken as the melting initiation power. The power-temperature data was then fitted using

a least-squares technique on a digital computer. The equation $P = a_1T^4 + a_2T + a_3$ was used. The $a_1T^4$ term accounts for radiative power transfer, and the $a_2T$ term accounts for conductive and convective losses. The $a_3$ term accounts for the fact that power is not zero at zero temperature, but is zero at room temperature. Hence the equation form can be theoretically justified. Above about 6000°R, the data shows a smaller and smaller temperature increase for a given power increase as shown in Fig. 4. This is because radiative heat transfer begins to assume a significant role above that temperature, and power radiated is directly proportional to $T^4$. Proper choice of the $a_1$ and $a_2$ terms by the curve-fitting least-squares code allows this effect to be properly taken into account.

Two curve fits were made for each set of data. One fit used all data obtained. The other fit did not consider those points which appeared to be low due to carbon vapor. Figures 4 and 5 show a typical set of data and the two least-squares fits. The Fig. 5 procedure was used for all data. The number of points omitted varied from 0 to 3. The final step was to place the measured value for the melting initiation power in the fitted equation and solve for the temperature. This was done by solving $P_{melt} = a_1T^4 + a_2T + a_3$ for its appropriate root using a digital computer polynomial root-solving code.

## EXPERIMENTAL PROCEDURE: SOLIDUS-LIQUIDUS INTERFACE

The pressure tests from 200 to 1000 atm were conducted with a specimen geometry exactly as shown in Fig. 2. The hole plug and vent hole were added to control the amount of chimney-effect flow. At higher pressures the flow tended to cool the specimen center unless a smaller vent hole was used. The test procedure and data procedure and data processing for each test was the same as for the triple-point temperature tests.

Above 200 atm, significant problems were encountered with gas convection currents. The radial temperature observation hole appeared to "float" in the pyrometer field of view due to the convective flow of the hot helium. To correct this problem, the optical tunnel shown in Fig. 3 was installed. It allowed measurements to be made up to 1000 atm with only minor convection problems about equal to the 100 atm tests without the tunnel.

## SELECTION OF GRAPHITE GRADES

Three grades of graphite commercially available from Union Carbide Company and one grade of special graphite supplied by Los Alamos Laboratory were selected for the program as outlined below:

AGOT - This is the normal nuclear grade graphite used for graphite-moderated reactors. All neutron absorbing elements, including boron, have been removed to very low

levels during the manufacturing process.

AGSR - This is an inexpensive grade of graphite. It was chosen because of its possible use as a structural material in a nuclear rocket engine.

AGKS - This is a spectroscopic grade graphite. The main criterion in its manufacture is maintaining high purity by careful choice of raw materials and manufacturing procedures. It is a relatively pure material available at reasonable cost.

LANG - This is a nuclear rocket grade graphite donated by Los Alamos Scientific Laboratory. It contained 70 ppm U-235 and other minor impurities. It is manufactured from 60 mesh high-purity graphite flour base mixed with Thermax sub-micron carbon-black particles and a small amount of 40 to 50 mesh pine-wood flour. A partially polymerized furfuryl alcohol called Varcum 8251 was used as the binder. After blending of the above materials and extrusion on a horizontal press, four thermal cycles were performed on the extrusions for a combined heat-up time of 127 hours. The final operation was graphitization at 4720$^{o}$R for four hours.

## TRIPLE-POINT PRESSURE RESULTS

The results obtained from the triple-point pressure tests are given in Table 1. The pressure at the melting portion of the graphite is accurately known for the tests listed. Many tests are not included because swelling

occurred and/or all vent holes to the melting portion were plugged. This indicates a buildup of internal pressure and hence the total pressure is unknown for such tests. All tests listed had vent holes to the melt which remained open throughout the test.

The criteria for determination of melting was the coincidence of two conditions:

1. The presence of filled spheres, since surface tension in a liquid causes such a formation, or other visual dendritic structures such as shown in Fig. 6.

2. The sudden and rapid rise in electrical resistivity of the test specimen resulting in a rapid rise in voltage across the test specimen.

These tests indicate that 1515 psia is the threshold pressure for graphite melting to occur and is therefore the indicated triple-point pressure. No evidence of melt was observed in any location where the pressure was below 1515 psia or 103 atm.

## TRIPLE-POINT TEMPERATURE RESULTS

The results obtained for the triple-point temperature are given in Table 2.

Since the temperature at which the voltage inflection point (VIP) occurred did not appear to be a strong function of pressure for pressures near the triple point, tests run at slightly above and below the triple-point pressure

are reported in these results. Below the triple-point pressure, no filled spheres were observed, but the sublimation temperature measured in such tests at a few pounds per square inch below the triple-point pressure would be only an undetectable amount different from the triple-point temperature.

From Table 2, the triple-point temperature results are AGOT grade, 7529 $\pm$ 19°R; LANG grade, 7606 $\pm$ 60°R; AGSR grade, 7629 $\pm$ 33°R; AGKS grade, 7738 $\pm$ 45°R. These values are the mean value for the experiments listed plus or minus one standard deviation of the mean.

## SOLIDUS-LIQUIDUS INTERFACE RESULTS

The results obtained for the melting temperature of LANG grade graphite for pressures of 200 through 1000 atm are given in Table 3.

Five or more specimens were run at each pressure. Only those experiments which contained no observable difficulty, however, are reported in Table 3; i.e., if a test was unsatisfactory because of specimen misalignment, improper functioning of the optical tunnel, or other recognizable physical difficulties, the data was not used.

The data and one standard deviation are shown in Fig. 8. A linear least-squares fit to the solidus-liquidus interface data from 200 to 1000 atm for LANG grade graphite yields $T = 0.1166\ P + 7633$, where P is absolute atm pressure

and T is $^{o}R$. This equation yields 7750 at 1000 atm pressure and 7656 at 200 atm. Fig. 6 shows a typical specimen after melting above the triple-point pressure.

## DISCUSSION OF PROCEDURES AND POSSIBLE ERRORS

The optical pyrometer used for these measurements was calibrated at NBS in June, 1967. Additional checks of this calibration were performed on the actual experimental setup by measuring the melting point of gold, molybdenum and tungsten. The results of these tests verified the accuracy of the pyrometer as it was used on this particular apparatus and indicated that the temperatures reported are very close to absolute values. It is estimated that the abscissa of Fig. 8 could be labeled minus zero plus $50^{o}$.

Geometry effects due to the optical path through the pressure cell were investigated. A $4660^{o}R$ brightness-temperature tungsten filament, a mockup of the cell geometry including the safety shield, optical tunnels, etc., present on the actual equipment was alternately placed between the pyrometer and the filament and then removed. The same temperature reading was obtained in both cases, demonstrating that no errors were introduced because of cell geometry.

Errors due to changes in the calibrated pyrometer were eliminated by periodically comparing the calibrated pyrometer to an older pyrometer of long-proven stability. A pyrometer comparison instrument was available for this

purpose. The older pyrometer was stored except to periodically recheck the calibrated pyrometer. No significant variations were noted.

As a further check on the pyrometer calibrations, a different pyrometer was used for one otherwise standard test. For test number 124, a Micro-Pyrometer was used. The results of this test can be found in Table 2. The results were essentially the same as results obtained with the calibrated manual pyrometer.

Absorption in the helium was investigated by passing a light beam from a 4660°R brightness-temperature tungsten filament into one window of the pressure cell and reflecting it out the other window with a mirror. With the tungsten temperature held constant, pyrometer readings were made on the emerging beam while the cell pressure was varied from vacuum to 140 atm. No differences were noted in the pyrometer readings. This procedure is the equivalent of testing to 280 atm since twice the normal optical path is involved inside the cell.

Absorption in the quartz window was measured experimentally. Alternate temperature readings were made of a 4660° brightness-temperature tungsten filament while looking through the quartz and then removing the quartz from the optical path. Above that temperature, the sun and combinations of grey filters were used. Grey filters were placed

between the pyrometer and the sun until a brightness temperature reading in the desired range was obtained. The quartz to be calibrated was then alternately placed between the grey filters and the pyrometer and then removed. The equation $1/T_o - 1/T_a = K(T)$ (Ref. 9) was used to least-squares fit the extensive data taken and obtain the best possible correction. $T_a$ = temperature measurement with no quartz in path; $T_o$ = temperature observed through the quartz; and $K(T)$ is a slight function of temperature, experimentally determined. All temperature data reported herein contain the appropriate correction.

Because the melting temperatures reported herein are higher than those reported by previous investigators (Refs. 4, 5, and 6), two types of tests were carried out to increase the credibility of these results. Firstly, three tests were made at 110 atm in which the AGKS specimens were heated in the normal manner to about 6500°R center temperature. The power was then increased rapidly by hand while the pyrometer made continuous readings. When extensive carbon vapor began to appear, the power was instantly reduced. Actual maximum pyrometer readings, including corrections for calibration and quartz window, of 7560, 7706, and 7606°R were obtained in this way and no evidence of melt was later observed on any of the three specimens. This method allowed high temperatures to be reached and measured before significant quantities of vapor could form and obstruct the readings.

These tests, based on actual readings involving no data extension, demonstrated that the melting temperature of graphite is above readings so obtained. Secondly, a test was run which involved measuring the power-temperature relation, taking the specimen almost to its melting point, cooling the specimen; and repeating the data. The data so obtained were very nearly the same. This demonstrated that no effects occur at higher temperatures which cause a significant deviation from the established and extended power-temperature relation.

VARIATION IN TEMPERATURE DATA

Several possible causes of the variation in the temperature data for the solidus-liquidus interface to 1000 atm are listed below with appropriate comments.

1. Errors in the temperature measurements due to instrument malfunction, operator error, etc., are considered to be very small because the technique involved taking several data points of temperature vs. power and performing a least-squares fit to that data. The effect of an error in a single temperature measurement would therefore be minimized. Fig. 4 represents a typical set of data and the corresponding least-squares fit. At no point does the measured temperature differ from the predicted temperature by more than about 100°R which would have little effect on the final predicted melting temperature.

2. Errors caused by carbon vapor were held to a small magnitude because the least-squares fit is firmly based on data at temperatures which are below the threshold of a significant vapor problem as shown in Fig. 5.

3. The central, hottest region could vary in composition from specimen to specimen because of inhomogenities in the graphite. Although this could account for some of the spread in data, no check was made on this item.

4. The impurities present could vary from specimen to specimen and could also vary from location to location in one specimen. No check was made on this item.

5. Variation in manufacturing procedure, baking temperature, and material is probably one of the major effects.

At this time it appears that the variation in the data is due to a combination of all items 2 through 5 listed above. Further research would be required to determine the effect of individual items.

## SUMMARY OF RESULTS

If any systemic errors such as excessive carbon vapor, changes in the power-temperature relation at high temperatures, gas absorption, misalignment, etc., are involved in these results, they would tend to cause the reported temperatures to be low.

A linear least-squares fit to the liquidus-solidus

interface data, Table 3, 200 to 1000 atm , for LANG grade graphite results in T = 0.1166 P + 7633, where P is absolute atm pressure and T is $^{o}$R. This yields the following results for LANG grade graphite: At 1000 atm the melting temperature is 7750$^{o}$R; at 500 atm, 7691$^{o}$R; at 103 atm, an extension of the least-squares equation predicts 7645$^{o}$R which is in excellent agreement with the 7606$^{o}$R mean triple-point temperature measured for LANG grade graphite as shown in Table 2. This agreement provides a verification of the triple-point temperature of LANG grade graphite. Actual measured values at the higher pressures, together with the appropriate mean values and standard deviations, are in Table 3.

Triple-point temperature results for the other grades of graphite are: AGKS, 7738 $\pm$ 45$^{o}$R; AGSR, 7629 $\pm$ 33$^{o}$R; AGOT, 7529 $\pm$ 19$^{o}$R. All triple-point temperature data expressed above are the mean value from the several experiments reported plus or minus one standard deviation of the mean. The standard deviation of the mean is the standard deviation divided by the square root of the number of data points used to compute the mean.

Triple-point pressure for the four grades of graphite is 103 atm absolute pressure.

## COMPARISON OF RESULTS WITH PREVIOUS INVESTIGATORS

The triple-point temperature was determined to be 400 to 500°R higher than that reported by Bassett (Ref. 4), Jones (Ref. 5), and Noda (Ref. 6). This difference appears to be justified by a comparison of the techniques and apparatus used by the investigators. Bundy (Ref. 7) did not report a triple-point measurement. Fateeva (Ref. 8) reported a triple-point temperature of 8370°R or about 750° higher than reported herein. The reasons for this difference are not apparent at this time. It may be noted, however, that Bundy (Ref. 7) reported a maximum temperature of 8280°R at about 65,000 atm which is 90° less than the 8370°R triple-point temperature reported by Fateeva.

The triple-point pressure was determined to be 103 atm absolute. Basset (Ref. 4) reported 102, Jones (Ref. 5) reported 100, and Noda (Ref. 6) reported 100 to 110 atm.

The solidus-liquidus interface, fitted straight line, shown on Figure 8 indicates an increase in melting temperature of 105°R from 103 to 1000 atm, or 12° per 100 atm. Basset (Ref. 4) reported an increase of 360°R from the triple-point to 4839 atm, or 8°R per 100 atm. Jones (Ref. 5) reported 60°R increase for an average pressure range up to about 1400 atm, or 4°R per 100 atm. Noda (Ref. 6) did not measure the solidus-liquidus interface temperature with increasing pressure. Bundy (Ref. 7) anchored the triple-

point per Basset's data, then contructed a phase diagram with the melting temperature rising with pressure up to 8280°R at about 65,000 atm, and then decreasing to 7200 - 7560°R at 125,000 atm. The average increase is only 2°R per 100 atm. However, the slope is not linear and he indicates a slope of 3°R per 100 atm at the triple point. Fateeva (Ref. 8) reported 180°R increase from triple point to 3000 atm, or 6°R per 100 atm.

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TABLE 1

Triple-Point Pressure Tests

| Test No. | Grade | Pressure psia | Comments | Resistivity Change | Specimen Melted |
|---|---|---|---|---|---|
| 79 | AGKS | 1515 | No spheres | No | No |
| 80 | AGKS | 1515 | No spheres | Yes | No |
| 84 | AGKS | 1515 | Filled spheres observed | Yes | Yes |
| 87 | AGKS | 1465 | No spheres | Yes | No |
| 88 | AGKS | 1465 | No spheres | Yes | No |
| 93 | AGKS | 1515 | Filled spheres observed | Yes | Yes |
| 94 | AGKS | 1535 | Filled spheres observed | Yes | Yes |
| 95 | AGKS | 1535 | Filled spheres observed | Yes | Yes |
| 97 | LANG | 1535 | Melted region near center | Yes | Yes |
| 98 | LANG | 1495 | No melt | Yes | No |
| 103 | ATJ | 1535 | Melted region, filled spheres observed | Yes | Yes |
| 104 | ATJ | 1495 | No spheres | Yes | No |
| 106 | ATJ | 1515 | Filled spheres observed | Yes | Yes |
| 107 | AGSR | 1515 | A few small filled spheres observed | Yes | Yes |
| 108 | AGSR | 1495 | No spheres | Yes | No |
| 109 | AGSR | 1535 | Filled spheres observed | Yes | Yes |

TABLE 2

Triple Point Temperature Tests

| AGKS | | | AGSR | | |
|---|---|---|---|---|---|
| Test No. | Pressure psia | $^{o}R$ | Test No. | Pressure psia | $^{o}R$ |
| 111 | 1515 | 7667 | 133 | 1555 | 7690 |
| 112 | 1515 | 7680 | 134 | 1555 | 7663 |
| 113 | 1515 | 7611 | 135 | 1555 | 7666 |
| 114 | 1515 | 7947 | 136 | 1535 | 7619 |
| 115 | 1515 | 7963 | 137 | 1535 | 7506 |
| 116 | 1535 | 7671 | | | |
| 118 | 1555 | 7634 | | | |
| 119 | 1475 | 7803 | | | |
| 125 | 1535 | 7665 | | | |
| Mean Values | | 7738 | | | 7629 |
| One $\sigma$ = | | 134 | | | 73 |
| $\sigma$ of mean = | | 45 | | | 33 |

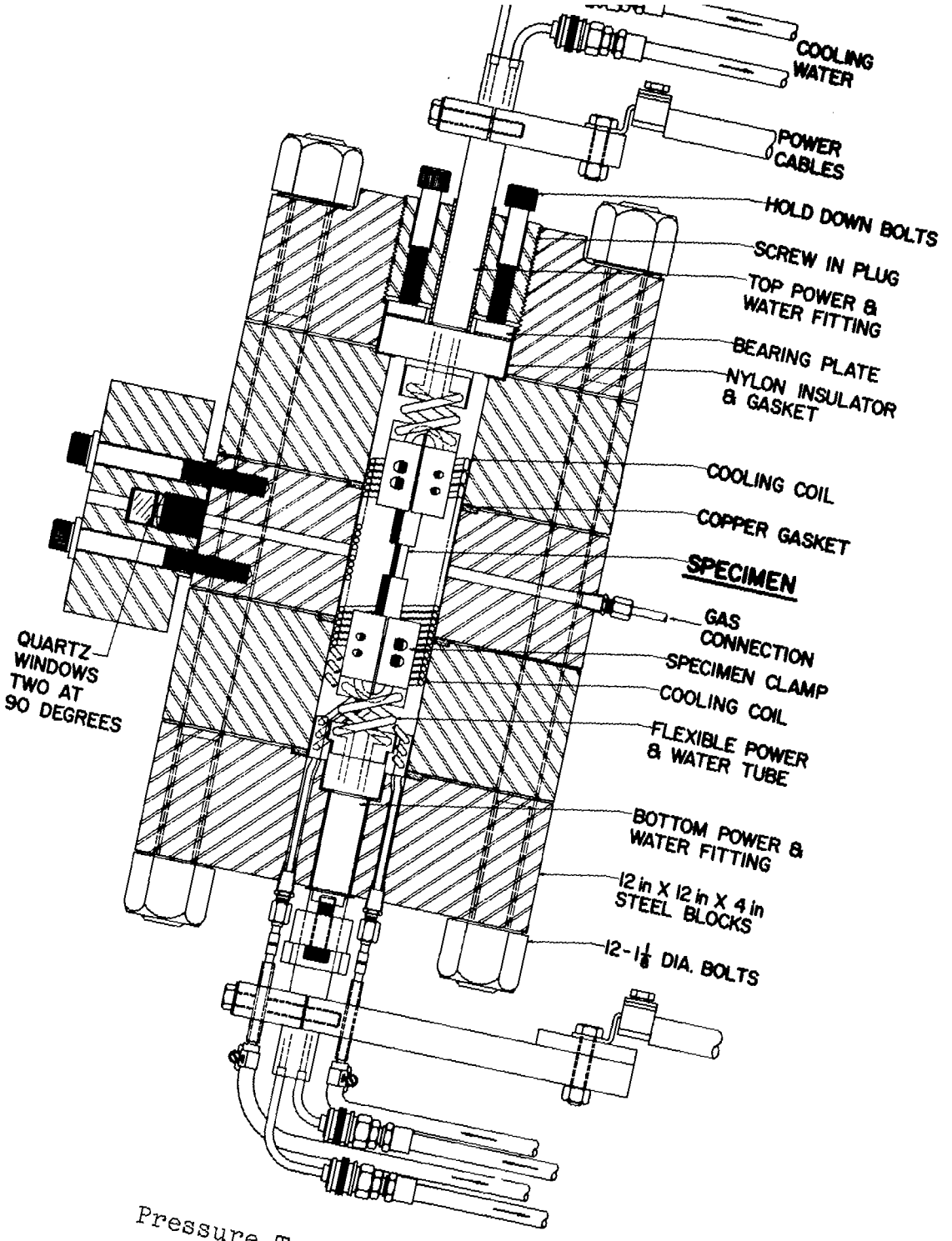
| LANG | | | AGOT | | |
|---|---|---|---|---|---|
| Test No. | Pressure psia | $^{o}R$ | Test No. | Pressure psia | $^{o}R$ |
| 120 | 1535 | 7710 | 138 | 1535 | 7585 |
| 121 | 1515 | 7711 | 139 | 1515 | 7497 |
| 122 | 1555 | 7389 | 140 | 1515 | 7545 |
| 123 | 1495 | 7447 | 141 | 1495 | 7539 |
| 124 | 1535 | 7675 | 142 | 1495 | 7580 |
| 126 | 1535 | 7701 | | | |
| Mean Values | | 7606 | | | 7529 |
| One $\sigma$ = | | 147 | | | 42 |
| $\sigma$ of mean = | | 60 | | | 19 |

TABLE 3

Solidus-Liquidus Interface Tests for
LANG Graphite

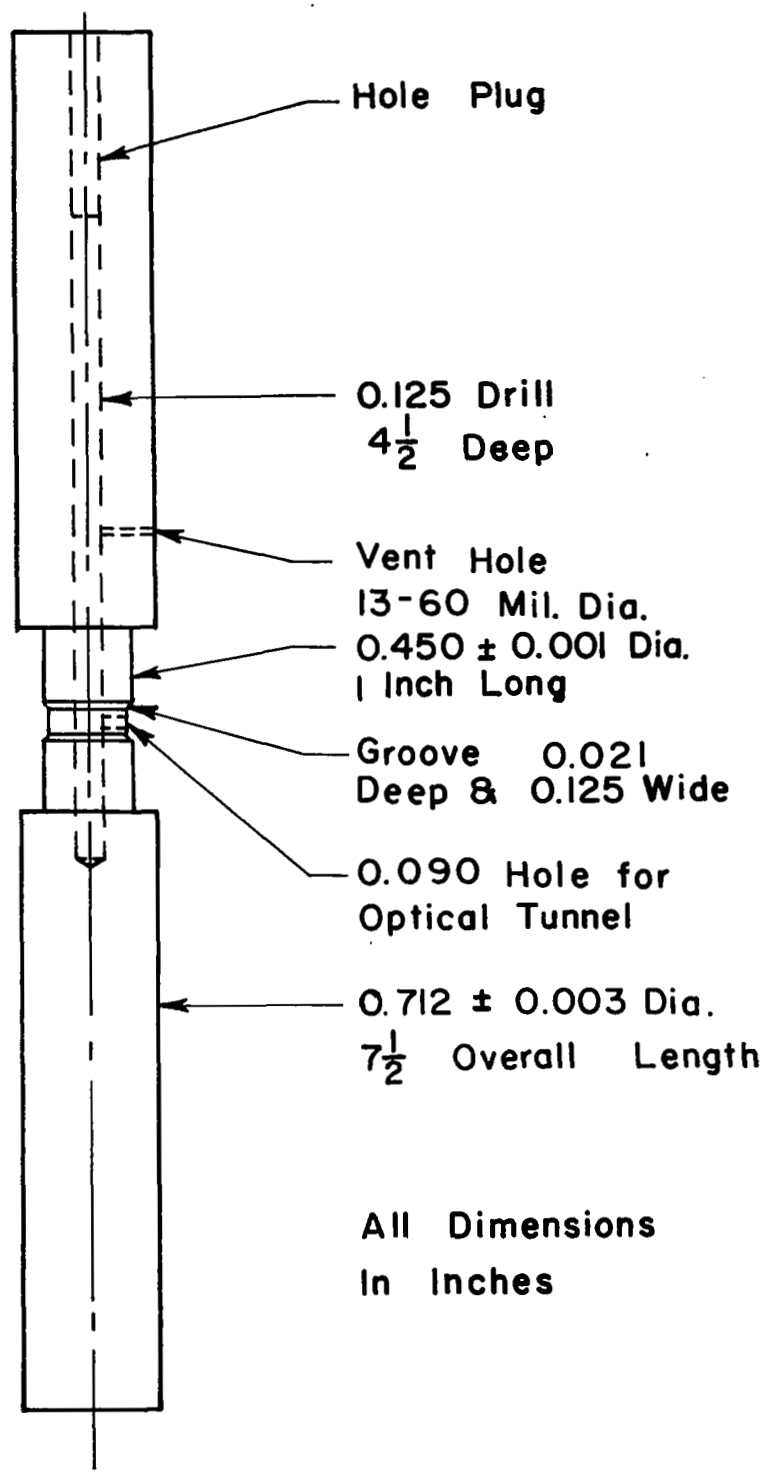
| 200 atm | | 300 atm | | 400 atm | | 500 atm | |
|---|---|---|---|---|---|---|---|
| Test No. | $^{o}R$ | Test No. | $^{o}R$ | Test No. | $^{o}R$ | Test No. | $^{o}R$ |
| 155 | 7727 | 147 | 7667 | 149 | 7910 | 164 | 7818 |
| 156 | 7730 | 148 | 7764 | 151 | 7546 | 166 | 7461 |
| 157 | 7652 | 159 | 7446 | 152 | 7695 | 168 | 7572 |
| | | 160 | 7566 | 153 | 7819 | 170 | 7751 |
| | | | | | | 172 | 7658 |
| Mean Values | 7703 | | 7611 | | 7743 | | 7652 |
| One $\sigma$ = | 44 | | 136 | | 158 | | 142 |
| $\sigma$ of mean = | 25 | | 68 | | 79 | | 64 |

| 600 atm | | 700 atm | | 800 atm | | 1000 atm | |
|---|---|---|---|---|---|---|---|
| Test No. | $^{o}R$ | Test No. | $^{o}R$ | Test No. | $^{o}R$ | Test No. | $^{o}R$ |
| 173 | 7705 | 183 | 7610 | 191 | 7736 | 199 | 7717 |
| 175 | 7643 | 185 | 7756 | 192 | 7593 | 200 | 7810 |
| 176 | 7644 | | | 193 | 7666 | 201 | 7935 |
| 182 | 7928 | | | 194 | 7605 | | |
| Mean Values | 7730 | | 7683 | | 7650 | | 7821 |
| One $\sigma$ = | 135 | | 103 | | 66 | | 109 |
| $\sigma$ of mean = | 68 | | 73 | | 33 | | 63 |



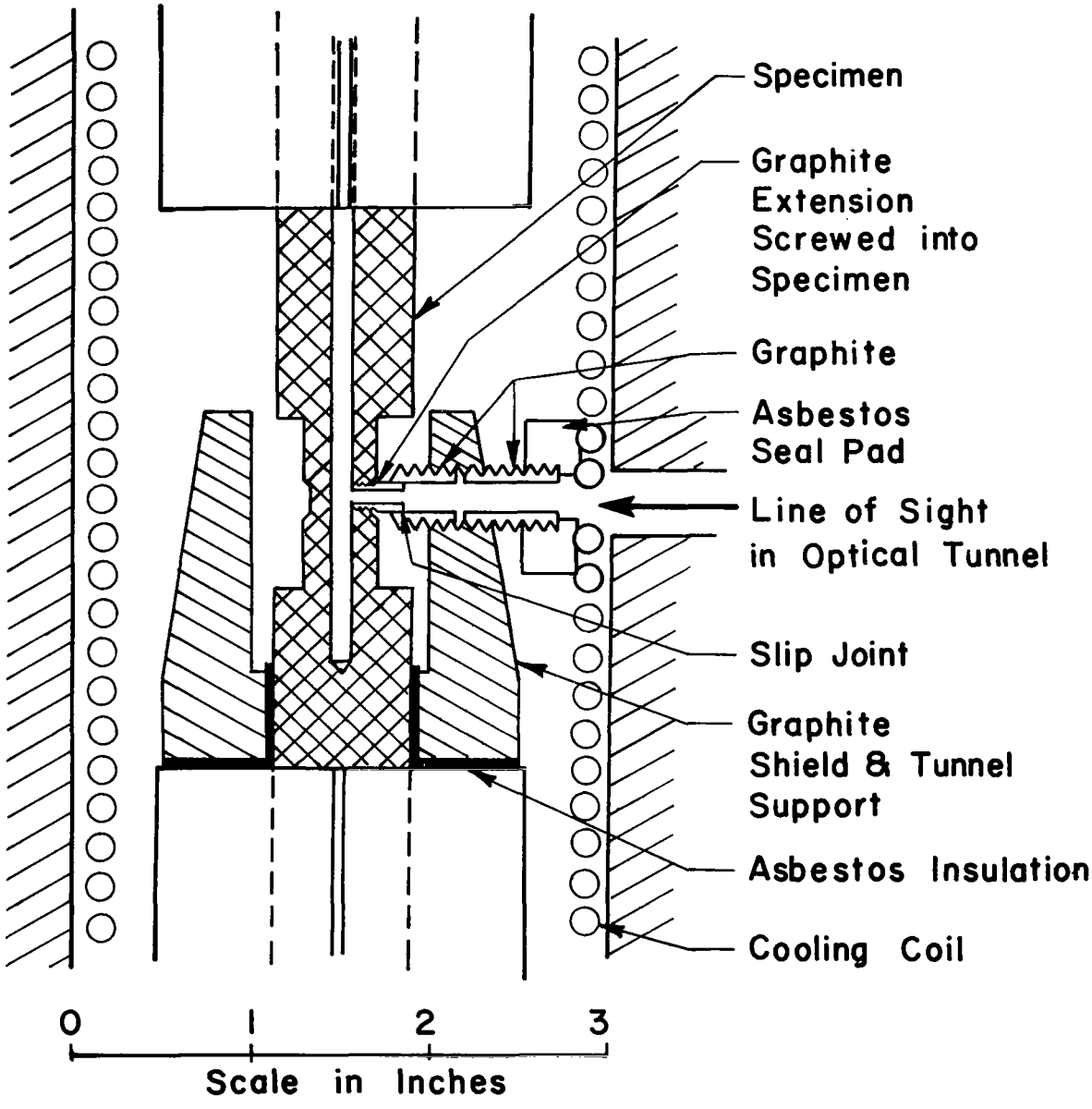

Pressure Temperature Cell

Figure 1



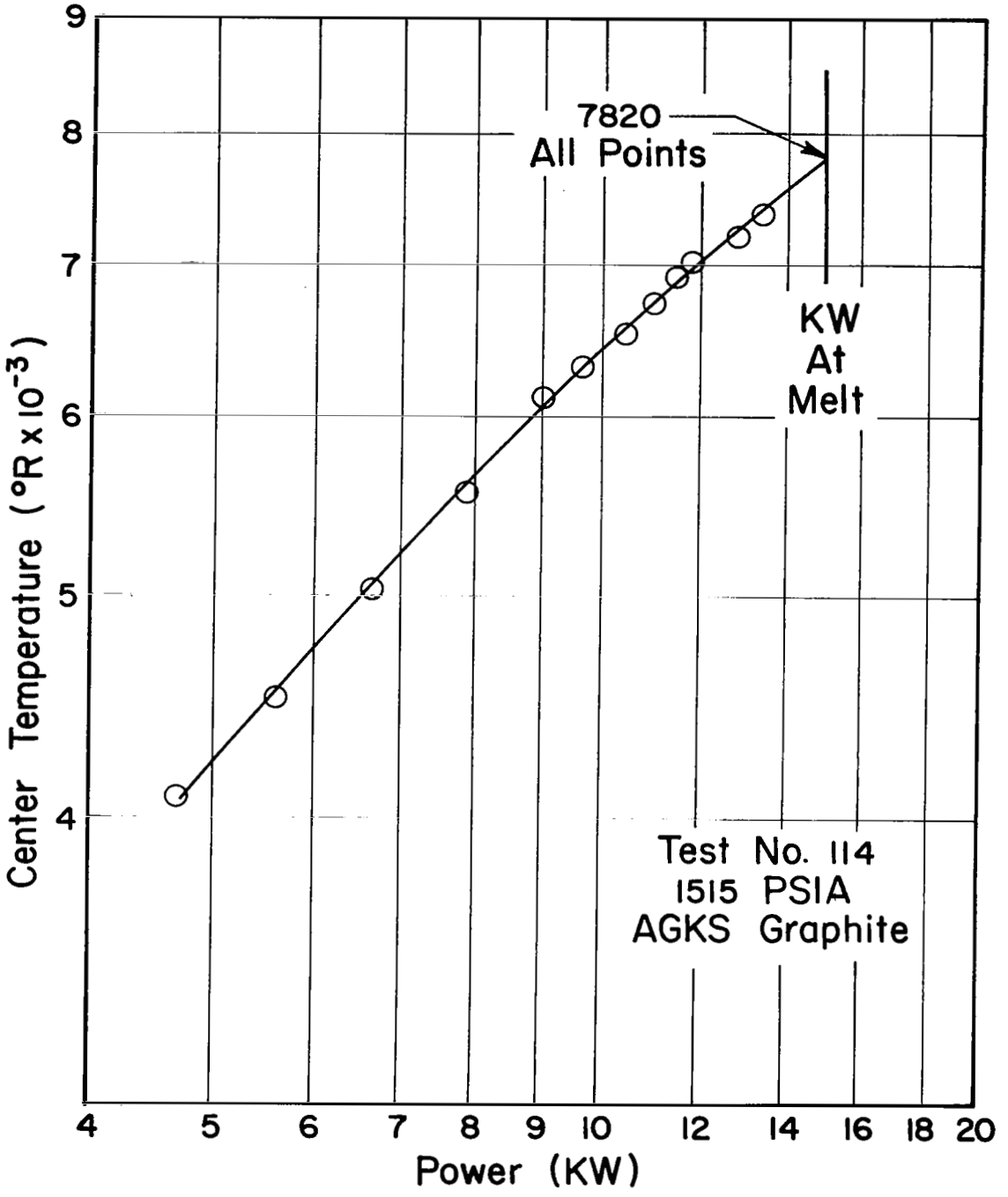

Graphite Specimen

Figure 2



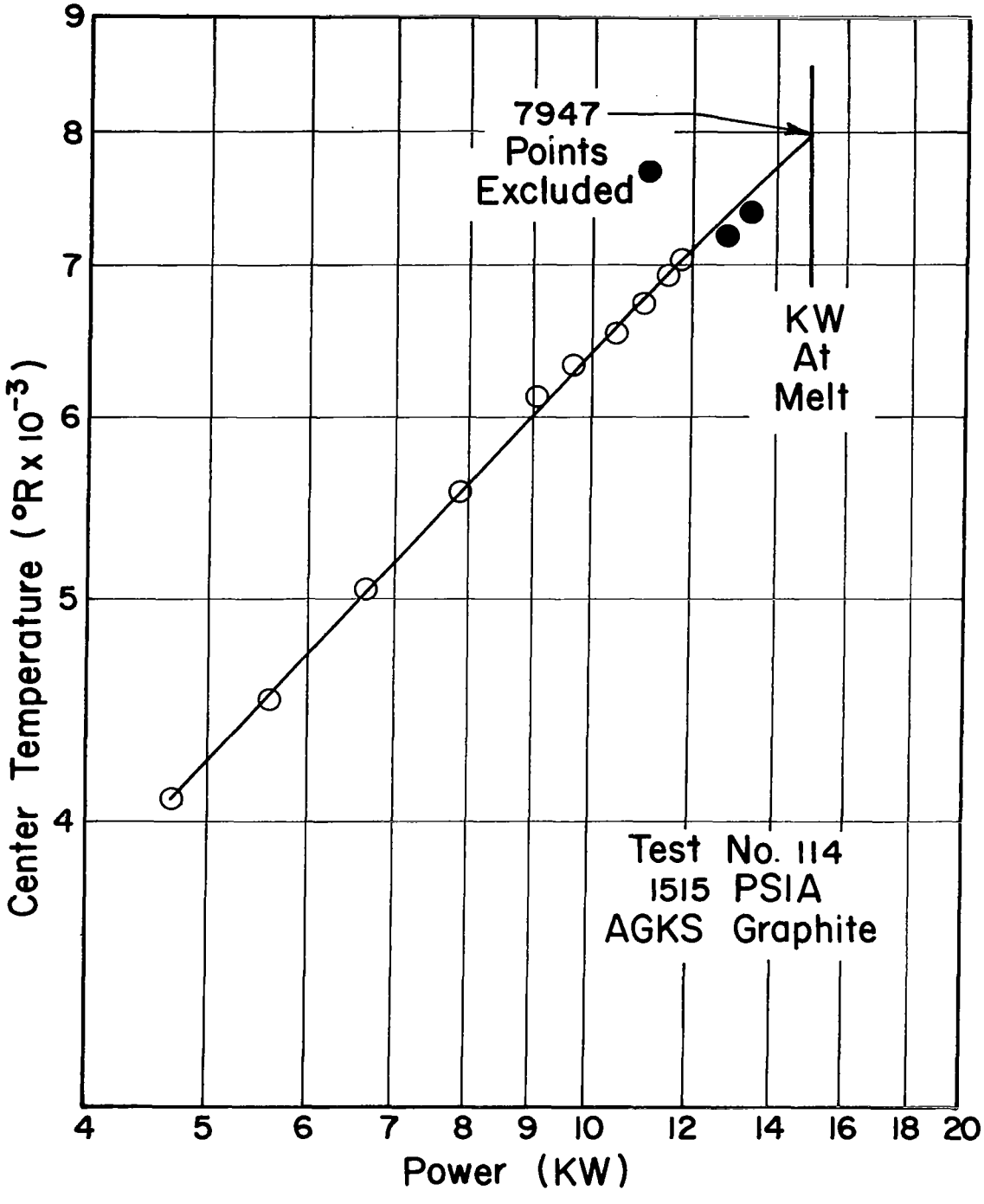

Optical Tunnel

Figure 3



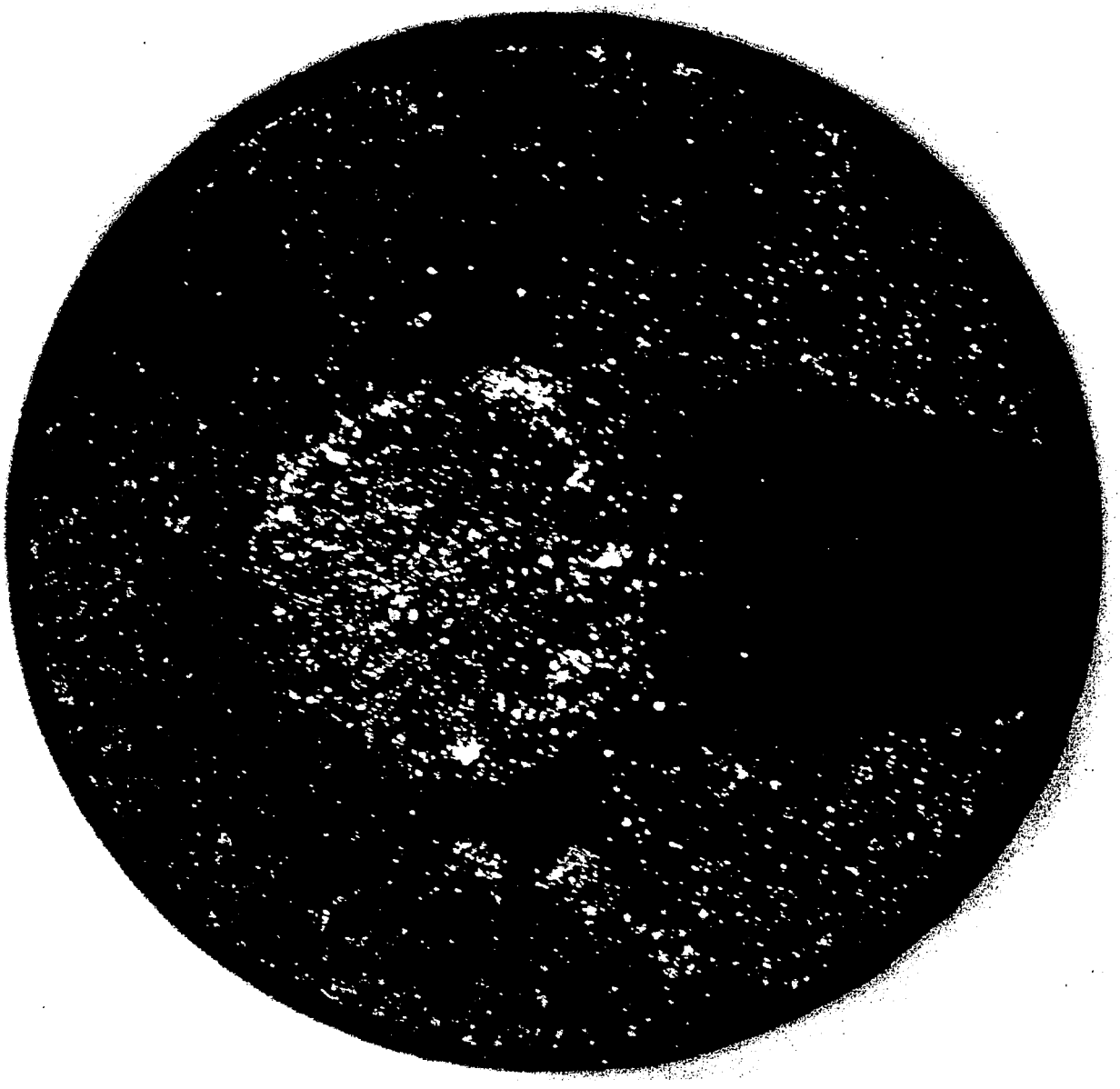

Center Temperature Using All Data Points

Figure 4




Center Temperature without Last Two Data Points

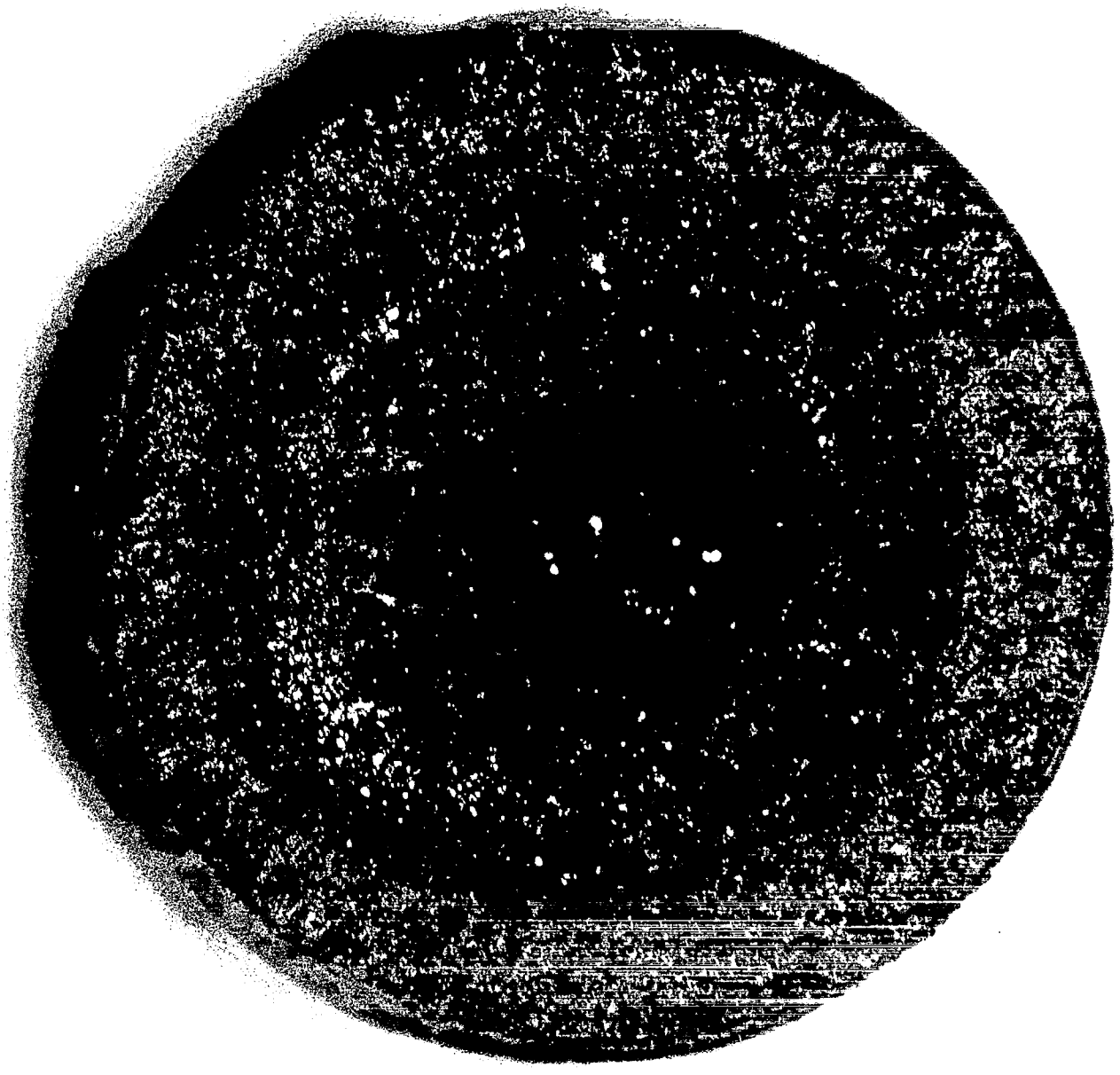
Figure 5



No. 152 - Table 3, 400 atm. 7695°R, LANG, melt (18x)
with resistance change i.e., VIP

1. Melt region shows clearly.
2. Three or four spheres show.
3. Vapor condensation and paritial plugging show in radial hole.
4. Highlights in balance of section are flat surfaces of grains.

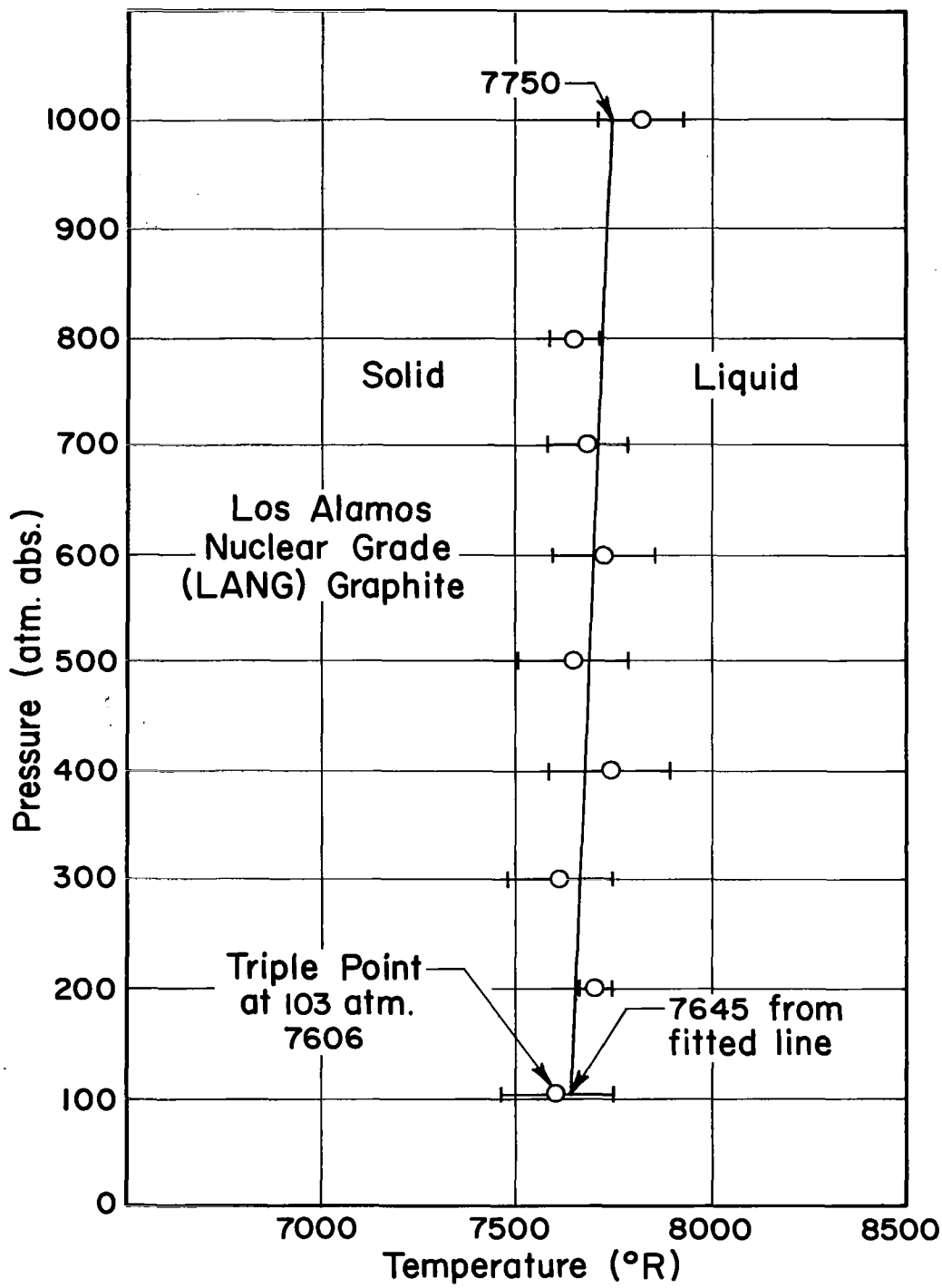
Figure 6



No. 98 - Table 1, 1495 psia, LANG, no melt (18x)
with resistance change i.e., VIP

1. Dark area surrounding hole is probably sublimation vapor deposit occuring at time of VIP.
2. Axial hole is full of "flakes" formed from gas.
3. Highlights are flat surfaces of grains.

Figure 7




Pressure-Temperature Phase Diagram for Los Alamos Nuclear Grade Graphite (LANG)

Figure 8

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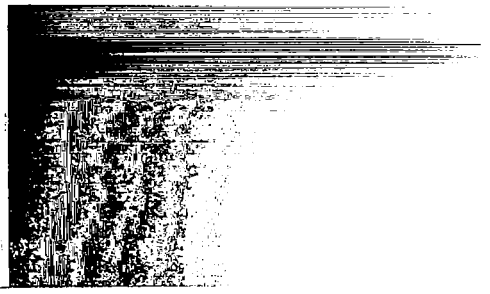

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