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Cesium Diffusion in Graphite

R. B. Evans III
W. Davis, Jr.
A. L. Sutton, Jr.

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Contract No. W-7405-eng-26

CHEMICAL TECHNOLOGY DIVISION

HTGR BASE TECHNOLOGY PROGRAM
HTGR Chemistry Studies (189a 01329)

CESIUM DIFFUSION IN GRAPHITE

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* Deceased.

Date Published: May 1980

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Oak Ridge, Tennessee 37830
operated by
UNION CARBIDE CORPORATION
for the
DEPARTMENT OF ENERGY

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FOREWORD

During the two years prior to his death in January 1972, R. B. Evans III had performed many experiments on the diffusion of cesium in various types of graphite. A. L. Sutton, Jr., and G. L. Towns were collaborators in some of this work, which was a continuation of that summarized on pp. 154-81 in *High Temperature Gas-Solids Reactor Base-Technology Program Progress Report, January 1, 1974-June 30, 1975* (ORNL-5108). In this report, Evans and Sutton presented a theory involving diffusional transport and first-order irreversible reaction of cesium in graphite. However, only a few experiments were available to test the theory.

At the time of his death, Evans had outlined or written about half of a report on the cesium-diffusion experiments. That material did not include a presentation or an analysis of the data, a task which he had apparently only just started. His writings did include a more detailed presentation of theory.

During the past year and a half, we have extracted the experimental data on cesium diffusion in graphite from various notebooks and have completed and corrected those sections of the report that Evans had started to write or had already written. This report presents these two items. It also presents an analysis of data on diffusion of ^{137}Cs in Hawker-Siddeley graphite but does not contain an analysis of the data on other graphites in terms of theory presented by Evans and Sutton. As the reader will note in the Appendix A, there are many experimental results, the analyses of which should add greatly to our knowledge of the processes that control the transport of cesium in at least five types of graphites. We are not presently performing analyses of these data; it is, however, part of the purpose of this report to make the data available to other workers so that they may be able to test their own theories.

Wallace Davis, Jr.
Alfred L. Sutton, Jr.

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CESIUM DIFFUSION IN GRAPHITE

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ABSTRACT

During the two years prior to his death in January 1977, R. B. Evans III, in part-time collaboration with A. L. Sutton, Jr., performed experiments on diffusion of ^{137}Cs in five types of graphite, namely, Hawker-Siddeley HS-1-1, Great Lakes Carbon Company H-327, H-451, HLM-85, and National Carbon Company ATJ. At the time of his death, Evans had written about half of a topical report; he had not yet analyzed or summarized the data. The present document provides a completion of the report that Evans had started and includes a presentation of all of the diffusion data, previously unpublished, obtained by Evans and Sutton.

Except for data on mass transfer of ^{137}Cs in the Hawker-Siddeley graphite, analyses of experimental results were initiated but not completed. The mass transfer process of cesium in HS-1-1 graphite at 600 to 1000°C in a helium atmosphere is essentially pure diffusion wherein values of $(D/\epsilon)_0$ and ΔE of the equation

$$D/\epsilon = (D/\epsilon)_0 \exp[-\Delta E/RT]$$

are about $4 \times 10^{-2} \text{ cm}^2/\text{s}$ and 30 kcal/mole, respectively.

Evans has presented a model of diffusion coupled with irreversible first-order fixation of cesium to explain data on mass transfer of ^{137}Cs in some of the other graphites. We tested this model by use of nonlinear least squares to estimate diffusion and reaction rate parameters of three sets of data, one on ATJ and two on H-327 graphites. Although the computer program did calculate these parameters, the fit of data to model was not good. The inadequacy of the model is its inability to account for a very steep ^{137}Cs concentration gradient at very small penetration distances. Further analyses of the data will clearly require the use of numerical techniques that permit a more complicated mechanism for cesium trapping than that given by irreversible first-order kinetics.

Three experiments were performed to test for vapor-phase transport of cesium in H-451 graphite at 635 to 680°C. In each case, such transport was less than the limits of measurement.

1. INTRODUCTION

It has been well established that most of the fission product gamma activity present in HTGR helium cooling circuits results from cesium that escaped fueled regions and then migrated into and through surrounding containment and support structures. A large portion of the available information concerning cesium migration in reactors has been generated through the auspices of the HTGR Surveillance Program,^{1,2} which directs and executes examination of Peach Bottom Experimental Reactor³ components. A schematic cutaway drawing of a Peach Bottom fuel-element assembly is shown in Fig. 1. All components of this assembly are composed of graphite, with the exception of the fuel kernels in the coated particles which are bonded together to form the fuel compacts. Considerable interest has been shown in the past in the nature of cesium release from fuel particles;⁴ however, current research attention has shifted to cesium migration in porous graphites as represented by the nonfueled components in Fig. 1. Thus, one facet of our program concerns the determination and interpretation of cesium distribution patterns as they exist in fuel elements retrieved from the Peach Bottom Reactor — particularly in spines and sleeves next to compacts. Another facet concerns an auxiliary investigation, which includes bench-scale experiments designed to support the surveillance effort. Reactor component data reflect long-term migration behavior; bench-scale results can readily be arranged to reflect short-term (transient) behavior. The primary purpose of this report is to present results of investigations of cesium diffusion performed since our previous studies.⁵⁻⁷

The basic relationships for the transport of volatile metals, such as cesium, through graphite matrices are predicated on the reasonable assumptions that the two primary paths for diffusion are: (1) the free volumes associated with open connection pores in the matrix; (2) the graphite surfaces that delineate the pore boundaries. The first are possible regions for gas or vapor phase transport; the second are regions for adsorbed phase transport. Vapor phase transport of cesium at the temperatures and bulk concentrations prevailing in a reactor core will be vanishingly small when vapor and adsorbed phases are in equilibrium. The

ALL DIMENSIONS IN INCHES

ASSEMBLY

FUEL ELEMENT OUTLINE

Fig. 1. Components of a Peach Bottom Reactor fuel element. The fuel compact assembly is of primary interest in this report.

only regions and conditions wherein cesium vapor transport appears to be important are the free volumes adjacent to highly saturated surfaces (forced flow down spine-compact-sleeve gaps) or volumes about bulk graphite surfaces prior to equilibration with respect to the adsorbed phase. Thus, it would seem that surface diffusion is the dominant cesium migration mechanism. However, we present the results of three experiments to test for the relative magnitudes of surface and vapor transport mechanisms. A combined mechanism of surface- and vapor-phase transport has been proposed by Özisik and Silverman⁸ for highly saturated systems at steady-state flow conditions and by Rodliffe⁹ for transient flow conditions. The latter treatment indicated that transverse flow effects are negligible at low cesium concentrations or loadings which correspond to the Henry's law region of the sorption isotherms.

In an effort to obtain some correspondence with results of other investigations and the U.S. development program, we chose to consider at least two impregnated graphites and two high-permeability graphites. Two of these are impregnated graphites, with questionable pedigree but pertaining to the Peach Bottom Reactor; one porous graphite was chosen for its relation to the Fort St. Vrain HTGR;¹⁰ and yet another porous graphite for the new "reference design" advanced HTGR.¹¹

The work plan for this investigation comprised two major parts or phases: (1) a series of cesium-diffusion experiments at uniform blanket helium pressure within the matrix, and (2) some cesium diffusion experiments with a helium pressure drop across the specimens to induce and superpose a forced carrier flow on the cesium diffusion system.

After preliminary experiments in phase 1, it became obvious that the best approach would be to employ the classical thin-layer technique wherein cesium is placed on adjacent faces of graphite specimens which are subsequently pressed together to form a diffusion couple. The source region can then be described as a spike or impulse function; diffusion times and specimen dimensions can be arranged so that the specimens represent semi-infinite media. With this technique, we could characterize the diffusion process by determining concentration profiles with the same grinding techniques and equipment used in earlier uranium-graphite diffusion studies.¹²

Except with Hawker-Siddeley graphite, cesium moved relatively rapidly out of the source region, then slowed down and assumed a quasi steady-state profile, similar to profiles for immobilized daughters of mobile precursors¹³ (e.g., Xe → Cs, or Kr → Sr). Something other than simple diffusion was apparently controlling the cesium migration in these other graphites.

2. PHYSICAL PROPERTIES OF TEST GRAPHITES

2.1 Description of Graphites

The graphites studied in this investigation fall into two rather broad categories: those which might be classified as structural materials, and those subjected to impregnation treatments for the express purpose of reducing the porosity and/or permeability of a given base stock. Initial conceptual designs of HTRs demanded impregnated graphite envelopes around fueled regions simply because the technology of coating individual fuel particles did not exist. Impregnated graphite envelopes coupled with a back purge of helium were the only fission-product-migration barrier available to minimize coolant contamination in the early days. As coating techniques improved, the need for impregnated graphites diminished. The emphasis then shifted toward the utilization of nearly isotropic materials with specific structural properties and resistance to radiation damage.

Nearly all artificial graphites are impregnated at least once in order to achieve an acceptable structure. Accordingly, we note that the term "impregnated graphite" in this report is used in reference to those materials that have undergone special and additional impregnation treatments primarily to attain permeability reduction. Typical impregnation treatments usually involve injection of pitch or an organic liquid, followed by a low-viscosity resin former, which are carbonized and finally partially graphitized to open pores; this is followed by a careful carbonization step to produce a char residue which serves to plug or reduce the diameters of the larger pores. Resins are composed of furfuryl alcohol or furfuraldehyde catalyzed to enhance polymer formation.

2.1.1 Impregnated graphites

Two impregnated graphites, HS-1-1 and HLM-85, were studied.

Hawker-Siddeley HS-1-1 is one of the remnants of the experimental batches produced by the now defunct Hawker-Siddeley Power Company in conjunction with an OEEC Dragon Project development program for the preparation of HTGR core components. The particular specimen used in our investigation was probably fabricated by subjecting a piece of Morgan EYX-50, a member of the EY9 family manufactured by Morganite Carbon Ltd., Battersea, London, to a single permeability-reduction treatment with condensation of furfuraldehyde with aniline. Use of this particular impregnant eliminates water-vapor evolution during bakeout. This material was received in the form of 7.6-cm-OD cylinders. Results of the single treatment were not very dramatic; the helium permeability at 1 atm was reduced from 1.0×10^{-2} to 4.4×10^{-3} cm²/s while the open porosity was reduced from 18 to 9 vol %. A pore-size distribution curve, obtained by mercury porosimetry, for the treated material is shown in Fig. 2. The pore structure is bimodal with a primary peak at $\langle r \rangle = 0.55$ μ m and a secondary peak at $\langle r \rangle = 0.01$ μ m. Treatment always attacks the primary peak. The impregnation must have involved a flow process of some type because flow tests indicated very uniform properties. It is interesting to note that pore radius is 0.4 μ m as seen by permeability alone. Perhaps the most significant feature of this material, insofar as cesium diffusion is concerned, relates to the EYX-60 base stock. The latter is manufactured from a finer-grained flour (not petroleum coke) mixed with ~30% carbon black. The small grain sizes are evident in the photomicrograph shown in Fig. 3. The presence of carbon black is reflected in the relatively high electrical resistivity of the treated product. The BET surface area of this material is unusually high, as noted in Table 1. This table includes data presented by Bromley and Large for comparison.¹⁴

Great Lakes Carbon Company HLM-85 came to use through General Atomics in the form of a full-size nonirradiated fuel element sleeve as used in the Peach Bottom HTGR (see Fig. 1). The base stock was fabricated from a moderate grain size "Y" coke (840- μ m maximum on edge), mixed with

Fig. 2. Porosity-pore size distribution curves for Hawker-Siddeley impregnated graphite. (Curves are based on mercury-porosimeter data.)

V-153997

As Received

ATJ - NATIONAL CARBON COMPANY

As Received

WS-1-1 - WALKER-SIDDELEY

As Received

HLN-85 - GREAT LAKES CARBON COMPANY

As Received

H-327 - GREAT LAKES CARBON COMPANY

Fig. 3. Photographs of sheared sections of various graphites of interest. (These photographs illustrate the relative sizes of the grains in the finished material, which, in turn, give some idea of the grain sizes in the original graphite flowers.)

Table 1. Characterization of the bulk properties of graphite related to cesium diffusion studies

Graphite	Bulk density (g/cm ³)	Open porosity (vol %)	Radius of primary peak, $\langle r \rangle^a$ (μ m)	BET surface area (m ² /g)	Permeability to helium at 1 atm (cm ² /s)
HS-1-1	1.80	7.8	0.55	21.5 ^b	4.4E-3 ^c
HLM-85	1.91	10.1	—	0.08	1.3E-3
Speer 711	1.84	12.0	—	—	1.0E-1
ATJ	1.77	13.4	3.0	0.14	1.0E+0
H-327	1.76	16.7	2.0 ^d	0.16	1.0E+0
H-451	1.73	17.2	4.0 ^d	0.63	3.4E-1
EY9 ^e	1.70	18.0	e	0.68	1.2E-1
HX12 ^e	1.83	10.0	e	0.63	2.2E-3

^aBased on mercury porosimetry; all graphites have a secondary peak at 0.01 μ m.

^bBased on nitrogen; all other areas are based on krypton adsorption.

^c4.0E-3 means 4.0×10^{-3} .

^dRough estimates; results uncertain due to the presence of isolated voids (0.20 mm diam).

^eData from J. Bromley and N. R. Large, ref. 14. Based on K_o and B_o , $\langle r \rangle$ values are 0.2 and 0.6 μ m for EY9 and HX12, respectively.

pitch, formed by extrusion, then graphitized and baked. Next, two or three impregnations with pitch were performed followed by additional furan-based resin impregnations and a final regraphitization at 2800°C. We speculate that at least some of these treatments were performed while the structure was in the form of a hollow cylinder to permit a radial flow impregnation. In any event, the helium permeability of the final material was reduced to $1.4 \times 10^{-3} \text{ cm}^2/\text{s}$. Mercury porosimetry results did not indicate a definitive primary peak pore radius. The average flow radius, $\langle r \rangle$, based on permeability measurements, was 0.5 μm . The effective diffusion coefficient was lower than the HS-1-1 values by a factor of about 50. One might expect this since HLM-85 was subjected to more impregnations than HS-1-1. The BET surface area of the treated material was relatively low,¹⁴ as shown in Table 1; thus we concluded that the resin char, after a final 2800°C treatment, will not enhance cesium absorptive capacity.

2.1.2 Structural graphites

National Carbon Company. ATJ graphite is employed as core support material in the Fort St. Vrain Reactor. This material contains fine-grain-size (150- μm) needle coke, and it appears to be fairly uniform in a given orientation direction (Fig. 3). We used this material in developing techniques for placing tracer on coupons and the development of grinding techniques. Although it had been impregnated with pitch more than once during fabrication, the nominal helium permeability is somewhat high, namely $1 \text{ cm}^2/\text{s}$ at 1 atm.

Great Lakes Carbon Company H-327 is the material used to fabricate the large hexagonal fuel blocks, as well as the reflector blocks, used in the Fort St. Vrain HTGR. The starting materials were petroleum-based needle coke mixed with a coal-tar binder. After extrusion and baking, the material was purified during graphitization. Somewhere in this scheme the bodies were subjected to coal tar pitch impregnations. Sheared specimens of the finished material reveal the presence of very large crystalline agglomerates; polished sections (see Fig. 3) reveal frequent large pores which we might classify as vugs. This very coarse structure might have resulted from the utilization of very large coke grains, up to 6.0 mm on

edge, in the starting materials. It is not surprising that this is one of the most permeable graphites we have encountered. As shown in Table 1, the helium permeability is $1.3 \text{ cm}^2/\text{s}$ at 24°C and 1 atm. Fortunately, the large vugs with radii up to $200 \text{ }\mu\text{m}$ do not overly influence the average bulk values. The average radius estimated from porosimetry data is $2.0 \text{ }\mu\text{m}$, while that based on permeability measurements is $3.0 \text{ }\mu\text{m}$.

Great Lakes Carbon Company H-451. The fabrication methods used for this graphite are quite similar to those used for H-327. The impregnant was petroleum rather than coal tar pitch. Of great significance is the fact that the starting coke grains were nearly isotropic, petroleum-derived particles as contrasted to the needle coke used in H-327. Inspection of sheared specimens suggests a finer grain structure than in H-327 (see Table 2). This material was to have been used in the advanced "large HTRs"; its most attractive feature is a relatively low degree of anisotropy (imparted by the improved coke), which enhances radiation stability.

2.2 Cesium Sorption by Graphites

Some typical results of cesium-graphite equilibrium studies are shown in Fig. 4 in the form of sorption isotherms; all these data were generated at the General Atomic Company.¹⁵⁻¹⁷ Included are data for several different graphites of current interest, namely: TS-688, which is National Carbon Company block AGOT; Great Lakes Carbon Company H-327 and H-451; and a high-surface-area coke, which is representative of a fuel bushing matrix binder and possibly of HS-1-1. The values sought from these isotherms are the α values as defined by Eq. (1) with $n = 1$,

$$C_g^n = \alpha C_s \quad (1)$$

These may be extracted, graphically or by use of Flowers' equation,¹⁸ from the lower left-hand segments of the isotherms that have a unit slope, namely the Henrian region of the isotherm. For example at 900°C [see Fig. 4(a)] graphite TS-688, which has a density of 1.7 g/cm^3 and a surface area of $0.585 \text{ m}^2/\text{g}$, loads to $10^{-0.35} \text{ }\mu\text{mole/m}^2$ at a vapor density of $10^{-10} \text{ }\mu\text{mole/cm}^3$ (i.e., $\sim 10^{-11} \text{ atm}$). Thus

Table 2. Characterization of the gas transport properties of graphites used in cesium-diffusion studies^a
(Nominal values based on measurements at 24°C)

Parameter	Graphite			
	HLH-85	HS-1-1	H-327	H-451
$LK/\Delta p$, cm ² /s·atm	5.36E-4 ^b	1.51E-3	1.05E-3	2.16E-1
B_0 , cm ²	1.04E-13	2.96E-13	2.06E-10	4.22E-11
D_{HeK} , cm ² /s	8.06E-4	2.84E-3	2.75E-1	1.2 E-1
K_O^d , cm	4.82E-9	1.70E-8	1.63E-6	7.18E-7
D_{He-air} , cm ² /s	1.98E-5	1.53E-3	2.72E-3	—
ϵ'/q (dimensionless)	2.76E-5	2.14E-3	2.05E-3	—

^aThe bases for the values in this table are as follows:

$\eta = 1.98 \times 10^{-4}$ g/(cm·s) and the conversion factor 1.013×10^6 g/cm·s·atm,

$v_{He} = 1.254 \times 10^5$ cm/s,

$D_{He-air} = 0.717$ cm²/s at experimental $t = 24^\circ\text{C}$ and $p = 0.941$ atm.

See Sect. 6 for Definitions and refs. 6 and 13 for detailed discussion of these parameters.

^b5.36E-4 means 5.36×10^{-4} .

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Fig. 4. Cesium sorption isotherms for various graphites and coke. [The flowers equation is Eq. (11) of ref. 18.]

$$\alpha = \frac{C_g}{C_s} = 2.2 \times 10^{-10} \frac{\text{bulk cm}^2}{\text{bulk cm}^3},$$

this value being constant along the 900°C isotherm (within the Henry's law region) up to a loading break point at $\sim 10^{-0.2} \mu\text{mole/m}^2$. Values of α for isotherms at other temperatures reveal that α varies with absolute temperature according to the equation

$$\alpha = \alpha_0 \exp(-\Delta E_{\text{sorb}}/RT),$$

where ΔE_{sorb} represents the internal sorption energy.

Regions to the right of the breakover loading are sometimes called Freundlich regions; above and to the right of these, the slopes of the isotherms tend to decrease again, suggesting the saturation of Henrian sites and sorption by other mechanisms. In any event, present interests focus on the Henrian region.

Cesium adsorption isotherms have been determined both by static and by dynamic methods.^{16,17} Each method involves a considerable number of measurements concerned with the rate of approach to equilibrium. For example, in the sorption studies of HLM-85 graphites, the times required to reach equilibrium in the Freundlich region at 1000°C ranged from 3 to 8 days. To keep equilibration times within this range, and also to enhance sorptivity of conveniently small specimens, most of the data (TS-688 and coke excepted) were obtained with specimens comprising small graphite particles produced by grinding and screening bulk specimens. The two-dimensional features of the coke allow direct conversion of loading values of H-327 and TS-688 charcoals from a surface-area basis [Figs. 4(a) and 4(b)] to bulk values [Fig. 4(d)]. Some questions arise concerning the conversion of particle data to bulk values for H-327 and H-451, and it has been recommended that a particle size correction (ratio of surface areas) of 1/2.5, from particle to bulk, be employed for both materials. However, the validity of this factor seems questionable in view of the different bulk surface areas cited in Table 1. Replots of the data, in units frequently used by the General Atomic Company [shown in Fig. 4(d)],

are based on use of the appropriate and/or recommended conversions from particle to bulk areas. Loci of points from an equation reported by Flowers,¹⁸ as fitted to TS-688 data, appear in Figs. 4(a) and 4(d). Cross plots of $\log(p_{Cs})$ vs $(T, ^\circ K)^{-1}$ at a fixed loading from plots such as Fig. 4(d) permit computation of ΔE_{sorb} from slopes corresponding to the Henrian region. In this region, the sorption energies for H-327, H-451, and the coke material all range about 67 k J/mole. Thus, from the standpoint of adsorption, the only distinguishing feature of these structures seems to be the different effective surface areas, with respect to cesium, and the different loading values at the breakover point.

2.3 Nature and State of the Diffusing Cesium

The cesium used in our laboratory experiments was a purified fraction extracted from a mixture of fission products. We received it as cesium chloride dissolved in hydrochloric acid. Since the extraction was performed 12 years before the present studies were initiated, the ^{134}Cs activity had effectively decayed, leaving only ^{137}Cs as the radioactive tracer. This considerably simplified counting procedures to assay cesium values in diffusion specimens referred to ^{137}Cs standards. Based on calculations with the ORIGEN code,¹⁹⁻²¹ it was estimated that ^{137}Cs constituted about 40% of the total cesium; stable barium, from cesium decay was present to the extent of about 12.5% of total cesium.

Before the cesium solution was placed on graphite to form source regions, aliquots of the chloride mixture were diluted with nitric acid and evaporated to dryness twice, then redissolved in distilled water. This was done to eliminate the chloride (as volatile nitrosylchloride and chlorine) and to convert cesium chloride to cesium nitrate. Appropriate volumes of the converted solution were then placed on graphite surfaces. After drying, the cesium-bearing graphite was placed in a quartz furnace tube which could be either evacuated or continuously purged with helium. The furnace temperature was raised to 450°C while the specimens were under a helium blanket. At this point, the system was evacuated to $\sim 2 \times 10^{-5}$ atm. Next, the helium blanket was reapplied and the

temperature was increased to 500°C; then the furnace was shut down to permit examination of the graphite specimen for uniformity of cesium placement. Tests with specimens before and after this heat treatment strongly suggested that the cesium had been converted from the nitrate to a strongly adsorbed state, probably through the intermediate oxide and metallic forms.

Under the assumption that trace amounts of cesium nitrate will decompose to cesium oxide at 450°C, we consider the equilibrium

which shifts as a function of temperature at various levels of oxygen chemical potential, as illustrated by the solid line on Fig. 5. The dot-dashed curves reflect the temperature dependence of the oxygen potential for the carbon - carbon monoxide - carbon dioxide - oxygen equilibrium and the dashed curves show the same sort of relationships for the equilibrium

The latter indicates that cesium vapor pressures over the oxides are relatively high.

Consider point A on Fig. 5, which is near the experimental conditions where we believe that the oxide decomposition takes place, namely 450°C at $\sim 2 \times 10^{-5}$ atm (we ignore the residual helium pressure). Since this point falls on the curve, elemental cesium and cesium oxide could coexist on or in the graphite specimens. We also find that carbon monoxide is the dominant oxide of carbon in the region of point A. Thus we invoke the auxiliary equilibrium

with

$$\Delta G = [-41.47 + 2 \times 4.37 - (-60.38)] - 3.203 \log_{10} (p_{\text{CO}})(p_{\text{Cs}})^2$$

at 700°K, using standard Gibbs' free energies.

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Fig. 5. Stability of cesium oxide and cesium carbonate as a function of oxygen potential and temperature. Isobars for cesium vapor and carbon oxides are also shown.

If the total pressure is 2×10^{-5} atm (pure carbon monoxide), the cesium vapor pressure at equilibrium is given by

$$\log_{10}(p_{\text{Cs}}) \leq 1/2 [-8.6 + 4.7] \sim -2;$$

furthermore, the equilibrium graphite loading, from H-327 sorption isotherms, for example, would be greater than 0.5 mg Cs/g graphite. Actual experimental loadings obtained with the subject procedures, conditions, and materials turned out to be ~ 0.1 mg/g, well within the uncertainties of theoretical calculations. We conclude that all the cesium placed on the graphite (as nitrate) reacted to form adsorbed cesium. Also, the adsorbed cesium at diffusion-anneal temperatures probably coexisted with small quantities of cesium in the vapor state.

3. EXPERIMENTAL PROCEDURES

Classical thin-layer techniques frequently employed for self-diffusion measurements in solid-state systems were used in this investigation. Briefly, a layer of cesium nitrate was placed on the inner faces of two identical cylindrical graphite specimens to form diffusion couples such that the diffusion would proceed axially from (normal to) a common plane source when subjected to diffusion anneal temperatures ranging from 600 to 1000°C. Annealing times and specimen lengths were adjusted to ensure that semi-infinite solutions to the diffusion equation would apply. After the diffusion anneals, the specimens were sectioned and analyzed to establish the nature and extent of cesium transport. Utilization of the thin-layer technique has special advantages: it allows some control of cesium concentrations; it obviates the need for complicated flux measurements; and all the characteristics of the cesium-transfer process are reflected by the shape of the section profiles.

The thin-layer technique was used in its simplest form in all experiments with ATJ and H-327 graphites and in four experiments with HS graphite. Static helium formed the atmosphere in each of these experiments. Experiments with H-451 graphite were all performed with helium flowing through the pair of samples to test the significance of vapor-phase transport of

cesium. Finally, experiments with HLM-85 graphite were performed with samples in the form of approximately 0.76-cm-diam cylinders pressed into annular cylinders of HS graphite of approximately 1.8-cm outside diameter.

3.1 Graphite Specimen Preparation

The diffusion specimens for experiments with ATJ, H-327, and H-451 graphites and for four experiments with HS graphite were fabricated by cutting and carefully machining stock material into individual cylindrical shapes with diameters ranging about 1.4 cm and lengths ranging between 1.0 and 1.4 cm. In the beginning, the circular surfaces normal to the axes were ground flat and parallel within very close tolerances after machining. This step was eliminated, however, when we became concerned about charging debris into surface pores, and it became obvious that the unexpectedly deep penetrations of cesium relaxed the need for close surface dimensions. To remove debris created by machining, finished specimens were heated to $\sim 250^{\circ}\text{C}$ on a hot plate and then dropped into cool isopropanol. A vigorous inhalation-ejection of alcohol was thus obtained; this expelled most of the debris present (ultrasonic cleaning appeared to us to be somewhat drastic). Finally, the specimens were outgassed under vacuum at 1000°C .

Specimens of HLM-85 graphite were obtained by cutting sections of approximately 1-cm width parallel to the axis of Peach Bottom sleeve No. 1010. This concentric-cylinder sleeve had an inner diameter of 5.7 cm and an outer diameter of 7.6 cm. The cut sections, of roughly trapezoidal cross section, were machined into 0.792-cm-diam cylinders. Each of four samples of HLM-85 graphite was pressed into a sleeve of HS graphite that had been bored to provide a tight fit of the two graphites. Outer diameters of the HS sleeves were about 1.8 cm.

3.2 Derivation and Production of Source Regions

One of the more difficult laboratory exercises was the development of an acceptable cesium source configuration for the diffusion experiments. Based on the initial assumption that the experiments might best be conducted

under constant source potential conditions, the possibility of using rather large (1.4-cm-diam x 0.5-cm) sources, in the form of cesium impregnated graphite, was explored. This approach was abandoned since it was nearly impossible to guarantee uniform cesium concentrations in such sources; furthermore, with most graphites, the sorbed cesium seemed to be permanently attached to the graphite grains after an initial movement through the matrix. This meant that a high level of activity would have to be carried throughout the diffusion anneals to obtain measurable levels in the specimens. The second approach involved attempts to generate a thin-layer boundary condition using thin impregnated wafers. Unfortunately, the problems mentioned above remained to some degree. Also the wafers were very fragile and difficult to handle.

Finally, we decided to plate the cesium tracer (in aqueous form) directly on one face of the diffusion specimens after treating the face with a temporary pore-blocking agent such as paraffin wax, mineral oil, kerosene, and/or naphthalene. The paraffin and oil were difficult to remove; kerosene had a tendency to evaporate before the aqueous solution dried up. Naphthalene gave the most satisfactory results, since judicious application of mild heat and vacuum to the treated specimens permitted solution drying before naphthalene removal via sublimation.

After tracer placement, specimen pairs were inserted in a quartz sleeve, which, in turn, was placed in a quartz furnace tube such that labeled ends of the specimens were in intimate contact. Then the entire assembly was heated to 500° for nitrate to adsorbed-state conversion. The total activity on each specimen was checked before and after the conversion step to make sure that the activities were nearly the same on each source surface.

During one source-region preparation, the two H-327 specimens drifted apart when the vacuum was applied at 450°C; thus the faces were not in contact during the conversion step (between 400 and 500°C). When the array was cooled down, we found that 25% of the tracer originally placed on the specimen surfaces had plated out all along the quartz sleeve. A similar test with two intentionally separated H-327 specimens exposed to 700°C

revealed that 50% of the cesium present deposited on the quartz sleeve. The results strongly suggested that elemental cesium vapor was present during conversions and initial stages of diffusion anneals. They also demonstrated how all of the specimens picked up various quantities of cesium during heat treatments even though the clearances between the sleeve walls and graphite surfaces were very small.

Exemplary integral concentration profiles for source regions (fraction activity remaining after grinding away an axial distance, x) are shown in Fig. 6. The curves resulted from procedure evaluation tests with H-327 (the most porous and nonuniform graphite); naphthalene was clearly the best pore-blocking agent, but some penetration via diffusion took place during conversion. Since the penetrations indicated on Fig. 6 at 500°C — plus those induced on the trip to higher diffusion anneals — must sometimes be accounted for through a dummy run, several specimen pairs were set aside for this purpose.

3.3 The Diffusion Anneals

Diffusion couples, except those involving HLM-85 cylinders in HS sleeves, with acceptable source regions were again placed in a tight-fitting quartz sleeve which, in turn, was slipped into a quartz furnace tube as shown in Fig. 7(a). Since we found that specimens could shift about, the couple was held in contact by graphite keys wedged into the inner sleeve at the ends of each specimen. Annealing was carried out in a small resistance furnace connected to a temperature controller with a sensor in contact with the outer quartz tube. Temperature measurements during annealing were made with a calibrated Chromel-Alumel thermocouple positioned as in Fig. 7(a). Separate temperature surveys [Fig. 7(b)] along the length of the inner tube were made with a thermocouple with the end attached to a close-fitting graphite plug to ensure that the diffusion couple resided in an isothermal region.

When an anneal was initiated, the system was held under vacuum until temperature-pressure conditions of 250°C and 20 μ m Hg were attained, at which point helium was admitted to a pressure of 20 torr above atmospheric.

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Fig. 6. Initial penetrations resulting from source region preparation and cesium vaporization during conversion between 400 and 500°C.

Fig. 7. Furnace tube assembly and temperature profiles for heat treatments (furnace not shown). The arrangement of *a* was used for both cesium conversion and diffusion anneal steps; the profiles at *b* relate to conditions for diffusion anneals.

The helium blanket, with a $1\text{-cm}^3/\text{s}$ sweep, was maintained during the rest of the operation which included (1) the remainder of the approach to annealing temperature, (2) the anneal at temperature, and (3) the cooling to room temperature. The helium inlet and outlet were arranged so that the region about the diffusion couple was at uniform pressure (no helium flow through the specimens except in experiments with H-451 graphite).

Blank or dummy runs were performed in exactly the same way as described above with the exception that the specimen was held at the annealing temperature for only 1 min. Times required to increase temperatures from 500°C to 600, 800, and 1000°C were 4, 18, and 70 min respectively.

As mentioned above, experiments with H-451 graphite were performed partly for evaluating the magnitude of vapor-phase transport of cesium. Equipment used in these experiments was more complicated than that used in all others because of the need to eliminate leakage flow of helium between graphite and its container and to measure helium flow rates. The apparatus used for annealing is shown in Fig. 8, while a line drawing of the flow-measuring apparatus is shown in Fig. 9. The setup for sample holding consisted of a hydraulic jack and associated compression sleeves and rings. In brief, axial pressure from the jack was transmitted radially through a silver sleeve to the internally copper-plated, thin-walled (0.015-cm or 0.006-in.) stainless-steel tube that contained the graphite specimens.

Extensive tests were made to ensure a good seal of the copper-plated stainless-steel tube to the graphite specimens. In fact, the metal tube had to be torn from the graphite specimens before we could measure radioactivity count rate versus penetration.

Figure 10 contains a more detailed drawing of the graphite sample and stainless-steel tube. Also shown in this drawing is a conceptual representation of the skewness in cesium penetration expected to be caused by flow of helium through the back face of the upstream graphite sample, through the region of contact of the two samples, and, finally through the back face of the downstream sample.

Fig. 8. Details of the system for sealing H-451 graphite to stainless steel tubing for helium-flow experiments.

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Fig. 9. Representation of equipment used in helium-flow experiments.

COUPLE ENCLOSED FOR DIFFUSION ANNEAL

CESIUM CONCENTRATION PROFILES ANTICIPATED
AT t_1 AND t_2 ($t_1 < t_2$)

Fig. 10. Details of graphite-stainless steel seal and of cesium concentration profiles if cesium is significantly transported in the vapor phase.

3.4 Determination of Integral Concentration Profiles

After removal from the annealing furnace, individual diffusion specimens were counted and then trimmed, in sections, over the sides and back using a small lathe enclosed in a glove box. This was done to eliminate spurious contributions of contamination over these surfaces and to obtain dimensions suitable for the lapping device; pieces were collected and counted to estimate the extent of contamination or to determine whether the specimen was acceptable for sectioning.

Trimmed specimens were cemented to a metal cylinder, and the latter was chucked into the lapping device shown in Fig. 11, using a height gage and surface plate (not shown). This was a critical step because accurate grinding required that the plane of the outer shoulder of the device be perfectly aligned with the specimen surface. The grinder is actually a modified micrometer, some details of which are shown in Fig. 12. In component I, a nylon collet surrounds the mounting cylinder and the back of the cylinder presses against a small-diameter anvil (not shown). Alignment or leveling was performed by tapping the cylinder at appropriate points while tightening the collet with a small spanner. The ground back surface of I served as a reference plane for leveling, lapping or grinding, and measurements of grinding distances. This surface was parallel with the shoulder on II and the plane of lapping.

During use, parts I and II were engaged as in Fig. 11, and the shoulder of II was fitted into the tracks of the base plate III (specimen down). For the first cut, considerable care was required for the height adjustment of the specimen surface with respect to the diamond lapping stone. Retraction of the shoulder advanced the specimen toward the stone. The calibrated markings and verniers were used to "set-up" a lapping distance of grinding interval. However, the final depth measurements were made with a direct-reading optical gage, as shown in Fig. 13, before and after each grinding operation. Grinding was carried out by moving the grinder back and forth over the grinding stone with a rotary motion.

All activity determinations, grinding operations, and height measurements were performed with the specimen chucked in the grinder; its

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Fig. 11. Illustration of the setup used to level the specimen prior to grinding. (All measurements are referred to the bottom of the grinding device, which is parallel to the upper shoulder.)

Fig. 12. Major component of the lapping device used for sectioning diffusion specimen. I is the anvil frame with specimen chucked into position; II is the thimble spindle with a polished retractable shoulder, which bears on the outer tracks of the base plate, III. A diamond lapping stone is positioned between the tracks.

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Fig. 13. Graphite sample mounted on optical gauge for measurement of relative length.

position was never altered. The sequence consisted of counting, grinding, measurement, and so on, until the gamma activity approached zero or a constant value (background contamination). The data involved measurements of the activity remaining in the specimen vs measurements of the total grinding distance. The stone was always saturated with kerosene, and the grindings were mopped up in the form of a slurry which was immediately discarded and never counted. Since this procedure eliminated dangers associated with handling dry contaminated dust, the operation could be performed in a hood. Count rate measurements were made with a single-channel analyzer system in conjunction with a 7.6-cm x 7.6-cm sodium iodide crystal. Whenever specimens were counted, a standard containing a known amount of cesium was also counted at the same distance from the crystal.

4. RESULTS AND DISCUSSIONS

As noted in the Foreword, a primary purpose of this report is to present essentially raw data from 36 experiments on the diffusion of cesium in five different graphites. These data are shown in tabular form in Appendix A; they are also presented in plots of relative concentration vs both penetration distance and the square of penetration distance in Appendix B. The relative concentration is the difference between two successive count rates, in counts/s, divided by the volume of graphite, in cm^3 , removed between the counting operations. The penetration distance in these plots is the distance to the midpoint between successive penetrations; it should be contrasted with several reduced (normalized) distances used in some figures of this section. Inserts in the data plots show the variation of specimen temperature, above 400°C , with time.

For the purpose of obtaining approximate concentration of cesium, in, for example, units of $\text{nanomoles}/\text{cm}^3$, in any graphite sample, the following product can be formed:

$$C (\text{nmole}/\text{cm}^3) = [f_1 f_2 / (f_3 f_4)] \times \text{CR} = 6.69 \times 10^{-4} \\ [\text{nmole Cs counts}^{-1} \text{ s} \times \text{CR (counts s}^{-1} \text{ cm}^{-3})].$$

Here

$$\begin{aligned} f_1 &= 2.275 \times 10^{-6} \text{ nmole } ^{137}\text{Cs}/(\text{dis/s}); \\ f_2 &= 2.5 \text{ nmole Cs/nmole } ^{137}\text{Cs}; \\ f_3 &= 0.85 \text{ photopeak photon}/^{137}\text{Cs disintegration}; \\ f_4 &= 0.01, \text{ the counting-system efficiency with the sample } 12 \text{ cm} \\ &\quad \text{above the 3-in. x 3-in. LiF(Ge) crystal, counts}/^{137}\text{Cs photon}; \\ \text{CR} &= \text{counting rate, counts s}^{-1} \text{ cm}^{-3}. \end{aligned}$$

Counting rates near the surface $x = 0$ in many of the experiments were in the range 10^4 to 10^5 counts $\text{s}^{-1} \text{ cm}^{-3}$; thus, concentrations of cesium were in the range 7 to 70 nmole/ cm^3 .

4.1 Data Treatment for Ideal Diffusion

Use of the term "ideal" for diffusion of sorbed cesium through graphite implies the absence of all rate terms other than the usual accumulation term in the diffusion equation. The equation that describes our transient experiments for these idealized conditions is

$$D \frac{\partial^2 C}{\partial x^2} = \epsilon \frac{\partial C}{\partial t} \quad (2)$$

for the impulse or thin-layer (combined initial-boundary) condition at $x = 0$, with $C(x, +0) = 0$ and $C(\infty, t) = 0$. C is the vapor-phase concentration of cesium in the pores, and D is an effective free-space diffusion coefficient. In reality, diffusion is primarily due to transport of cesium along pore surfaces. The solution is

$$\epsilon C_p(x, t) = C_b(x, t) = \left(Q_0 \sqrt{\frac{\epsilon}{\pi D t}} \right) \exp\left(\frac{-\epsilon x^2}{4 D t}\right). \quad (3)$$

The pre-exponential term is the surface concentration, $C_b(0, t)$. Subscripts p and b refer to pore and bulk, respectively. The symbol Q_0 represents the amount of cesium placed, at $x = 0$, on a single specimen per square centimeter of geometrical area normal to the linear diffusion path x , whereas the total amount per couple is $2 Q_0$; and t is the diffusion time.

Of the five graphites considered in this report, only Hawker-Siddeley graphite exhibits the characteristics of simple diffusion. This is shown by the plot in Appendix B, wherein logarithms of relative concentrations of ^{137}Cs , in counts $\text{s}^{-1} \text{cm}^{-3}$, appear to be linear functions of the square of the penetration distance, as required by Eq. (3). Extraction of diffusion coefficients from four of the experiments with Hawker-Siddeley graphite (HS-J-1 samples 11, 14, 3, and 5) by the graphical comparison method have been presented previously,⁶ as shown in Fig. 14. This figure is based on the reduced-penetration $u = (x/2) \sqrt{\epsilon/Dt}$; Fig. 15 shows data from two of the experiments based on the penetration distance, x .

All of the data pertaining to the diffusion of ^{137}Cs in Hawker-Siddeley graphite have now been analyzed in terms of Eq. (3) by use of the Marquardt nonlinear least-squares program.^{22,23} Results of this analysis of seven experiments are summarized in Table 3. It should be noted that there are small differences between reaction times and diffusion coefficients, D/ϵ , of Table 3 and Fig. 14 for experiments 11, 14, 3, and 5. Differences in diffusion coefficients reflect differences between graphical and numerical methods of analysis.

An examination of the plots of ^{137}Cs concentration vs penetration distance in Hawker-Siddeley graphite (Appendix B) reveals that the first point is higher than expected by a factor of about 2 in experiment 11 and by a factor of about 5 in experiment 12; in all other experiments, the first point appears to be lower than expected. Parameters listed in Table 3 are based on analysis of all of the data; however, an analysis based on the exclusion of the first point in all experiments produces results that are not much different. Figure 16 displays data from Table 3 in a reciprocal-temperature plot. Numerical analyses of the data in terms of the equation

$$D/\epsilon = (D/\epsilon)_0 \exp[-\Delta E/RT] \quad (4)$$

are summarized as follows:

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Fig. 14. Generalized integral profiles for cesium diffusion in Hawker-Siddeley graphite.

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Fig. 15. Plots of two integral concentration profiles for Hawker-Siddeley graphite showing the logarithm of the fraction of cesium activity remaining as a function of the grinding or penetration distance, x .

Table 3. Analysis of data on ^{137}Cs diffusion in Hawker-Siddeley graphite

Run No. ^a	Approximate run		No. of points	$\hat{S}(\text{fit})$ %	Parameters and standard deviations ^b					
	Temp. (°C)	Time (min)			B(1) (counts s ⁻¹ cm ⁻³)	$\hat{S}[B(1)]$ (cm ⁻¹)	B(2) (cm ⁻¹)	$\hat{S}[B(2)]$ (cm ⁻¹)	D/ε (cm ² s ⁻¹)	$\hat{S}(D/\epsilon)$ (cm ² s ⁻¹)
I1	800	1250	21	17.5	1.536E+4	0.078E+4	10.238	0.162	1.27E-07	0.04E-07
I2	800	1250	24	28.6	1.240E+4	0.108E+4	9.840	0.219	1.38E-07	0.06E-07
I3	600	4150	15	15.6	1.143E+5	0.062E+5	63.538	0.664	9.95E-10	0.21E-10
I4	600	4150	10	32.1	1.144E+5	0.165E+5	59.451	1.363	1.14E-09	0.05E-09
1	>900 ^c	47	13	25.0	1.513E+5	0.132E+5	48.572	1.081	1.50E-07	0.07E-07
3	700	2090	33	43.0	5.879E+4	0.677E+4	34.678	0.786	6.63E-09	0.30E-09
5	1000	175	24	18.4	3.558E+4	0.186E+4	19.443	0.205	2.52E-07	0.05E-07

^aRuns I1, I2, I3, and I4 involved the HS graphite as sleeves around HLM 85 graphite. Runs 1, 3, and 5 involved separate HS cylinders.

^bB(1) corresponds to the term $Q_0 \sqrt{\epsilon/\pi Dt}$ of Eq. (20); B(2) corresponds to $\sqrt{\epsilon/Dt}$. $\hat{S}(\text{fit})$ is the estimated standard deviation of fit of the data to Eq. (20). $\hat{S}[B(1)]$, $\hat{S}[B(2)]$, and $\hat{S}(D/\epsilon)$ similarly are estimated standard deviations of B(1), B(2), and D/ε. We calculate $D/\epsilon = 1./[t * B(2) * B(2)]$ and $\hat{S}(D/\epsilon) = 2. * \hat{S}[B(2)] * (D/\epsilon)/B(2)$.

^cThe time-averaged temperature of this experiment was assumed to be 950°C for the purpose of plotting in Fig. 16 and of calculating the activation energy of diffusion.

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Fig. 16. Arrhenius plot of coefficients for cesium diffusion in Hawker-Siddeley graphite.

	$[(D/\epsilon)_0 \text{ and } \hat{S}(D/\epsilon)_0] \times 10^2,$ cm^2/s		$[\Delta E \text{ and } \hat{S}(\Delta E)],$ $\text{kcal/mole } ^{137}\text{Cs}$		$\hat{S}(\text{fit}),$ $\%$
All data used	4.44	5.87	30.4	2.7	50
Exclude runs I1 and I2	3.71	0.64	30.1	0.35	6.1

It is apparent from this summary, or from Fig. 16, that most of the uncertainty in $(D/\epsilon)_0$ is due to runs I1 and I2; the calculated energy of activation is not changed much by their exclusion.

4.2 Diffusion Coupled With Nonequilibrium Sorption

When the same type of thin-layer cesium experiments are performed with materials such as H-327 graphites, which contain three-dimensional grains, one obtains penetration profiles that are significantly different from those indicative of an idealized diffusion process. The first deviation from ideal behavior became apparent in the early stages of the grinding procedures; the cesium activity that remained dropped sharply in the first series of sections, then more slowly with distance as the grinding depths increased. It appeared that only part of the cesium initially placed on the surfaces diffused into the specimens. A residual amount seemed to be immobilized near the source, which resulted in abnormally high cesium concentrations at the $x = 0$ surface.

Illustrations of this effect, in terms of typical fraction remaining versus grinding distance plots, are shown on Fig. 17. This same characteristic applies to all data on ATJ, H-327, and HLM-85 graphites, as may be seen in the plots of Appendix B. We should also stress the point that such results were never obtained with Hawker-Siddeley graphite.

Surface effects of this kind were first reported for cesium-graphite (and barium-graphite) systems in 1961 by Bromley and Large¹⁴ with the following explanation:

The high activities (in out-of-pile diffusion specimens) near the source disc have been attributed to the fact that creation of a (machined) surface has opened up a number of otherwise blind pores which do not provide a through path for the diffusion process. The surfaces of these blind pores become covered

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Fig. 17. Initial appearance of integral profile data for cesium diffusion in large-grain H-327 graphite. (Appreciable amounts of cesium are "held-up" near the source region; an adjusted base for FR and Q_0 calculations was based on extrapolations of the bulk of the data back to $x = 0$, e.g., $FR = 0.79$ and 0.82 .)

with an adsorbed layer of diffusing species which is unable to migrate further into the graphite.

If this explanation had included exposure of an unusually high number of active chemisorption sites, its plausibility would have been considerably enhanced. Nonetheless, Bromley's ideas do seem to form a nucleus for an extended explanation of holdup effects (over superficial surfaces) when possible sorption mechanisms are also included.

We envision that the nature of machined surfaces truncating three-dimensional grains in structural graphite is quite complex, and, in theory, one must consider all forms of sorbed states as they relate to a cesium vapor pressure: a physically adsorbed state (P), an intermediate chemisorbed state (A), and a final chemisorbed state (B). A representation given by Hayward and Trapnell²⁴ is

$$[Cs]_v \rightleftharpoons [Cs]_P \rightleftharpoons [Cs]_A \rightleftharpoons [Cs]_B \quad (5)$$

For the special case of Hawker-Siddeley graphite, wherein holdup was never detected, the above seems to reduce to

$$[Cs]_v \xrightleftharpoons{K_e} [Cs]_A,$$

where contributions of the (P) and (B) states are negligible. The system appears always to be in equilibrium even though the concentrations shift about markedly as a result of diffusion. Perhaps the absence of (P) and (B) state contributions results from the dominance of the two-dimensional materials (coke and lamp black) in this particular structure.

For more typical graphites, like H-327, with three-dimensional grains, we suspect that Eq. (5) reduces to

$$[Cs]_v \xrightleftharpoons{K_e} [Cs]_A \xrightleftharpoons[k_4]{k_3} [Cs]_B \quad (6)$$

since (P) coverage is small compared with (A) coverage and (v-P-A) equilibrium is rapidly attained. Consider now a possible sequence of events which might occur near the source surface of a structure wherein

the (B) state can exist [and Eq. (6) applies] under conditions related to a thin-layer experiment. Initially, the cesium coverage suddenly becomes relatively high, which ensures significant B coverage if the rate constant k_3 is high. Immediately after this buildup, the cesium concentration starts to fall as the mobile cesium, under the K regime, drains or diffuses out into the specimen. If $k_4[\text{Cs}]_B$ is smaller than $k_3[\text{Cs}]_A$, considerable cesium can be left behind in an immobilized (B) state. A disproportionate amount of this immobilization or holdup occurs at the surface since this is a region of unusually high concentration of (B)-state sites (artificially formed) and high initial cesium concentrations.

The model is predicated on the assumption that the surface (coupled with gas) phase diffusion rates are equal to, or greater than, the equilibration rates associated with ultimate sorption saturation; thus, the mobile fraction will not "wait around" for a slow sorption process to achieve equilibrium. With this in mind, we invoke a system similar to that described by Eqs. (5) and (6). Whether a P-A system without B, or and A-B system without P, controls the sorption rate is of little concern since we do not know enough about the sorption kinetics of cesium in "H-327-like" graphites to make such a distinction.

A new hypothetical system, namely a coupled mobile gas-surface phase which tends to become trapped or occupy strongly chemisorbed sites and to diffuse as well, is visualized at this point. The subscript $\underline{\text{T}}$ refers to immobilization sites or states; subscript $\underline{\text{M}}$ means mobile. For example, $C_{\underline{\text{M}}}(\text{x},\text{t})$ and $C_{\underline{\text{T}}}(\text{x},\text{t})$ designate mobile and immobile concentrations, respectively, which are referred to the pore volume; if a prime appears, the concentrations are referred to the bulk volume. We seek a reaction or sorption-rate expression, r_{ads} , which is to be added to the $\partial C/\partial t$ term of the transient diffusion relationship Eq. (2). In the absence of diffusion, the rate refers to the following system:

where

$$r_{ads} = \frac{dC_T(t)}{dt},$$

$$r_{ads} = k_1 C_M(t) [C_T(\infty) - C_T(t)] - k_2 C_T(t),$$

or

$$r_{ads} = k_1 C_M(t) \cdot C_T(\infty) - [k_1 C_M(t) + k_2] C_T(t). \quad (8)$$

The symbol $C_T(\infty)$ means the maximum number of available chemisorption sites, and the last term on the right within the brackets represents a combined fill-up and reversing back-pressure term. In view of the nature of the thin-layer experiments, we anticipate that the fraction of available τ -sites actually occupied will be small; furthermore, the k_2 desorption rate will be negligible. Thus, $r_{ads} = k C_M(t)$, where $k = k_1 C_T(\infty)$ with units s^{-1} .

The equation for transient diffusion of the mobile cesium species subject to an irreversible first-order sorption rate is

$$\frac{\partial^2 C_M}{\partial x^2} = \frac{\varepsilon}{D} \frac{\partial C_M}{\partial t} + \frac{\varepsilon k}{D} C_M, \quad (x > 0, t > 0), \quad (9a)$$

where C_M is the mobile concentration as a function of position and time, $C_M(x, t)$. The initial and boundary conditions corresponding to the experimental procedures are:

$$C_M(x, 0) = 0, \quad (x > 0), \quad (10a)$$

and

$$\lim_{x \rightarrow \infty} C_M(x, t) = 0, \quad (t > 0), \quad (10b)$$

when

$$J(t) = J_0, \quad (x = 0, t < t_0)$$

$$J(t) = 0, \quad (x = 0, t > t_0),$$

where we hold $J_0 t_0 = Q_0$, while $t_0 \rightarrow 0$ and $J_0 \rightarrow \infty$. This flux condition at the $x = 0$ boundary may be written as

$$J_{x=0} = Q_0 \delta(t), \quad (10c)$$

where $\delta(t)$ is the Dirac delta function whose Laplace transform is unity.

The Laplace transformed version of this system is

$$\frac{d^2}{dx^2} \left[\bar{c}_M(x,s) \right] = \frac{\epsilon}{D} \left[s \bar{c}_M(x,s) - c_M(x,0) \right] + \frac{\epsilon}{D} k \bar{c}_M(x,s), \quad (9a,s)$$

where transformation of Eq. (10a) is not required. Also

$$\bar{c}_M(\infty, s) = 0, \quad (10b,s)$$

with

$$(\bar{j})_{x=0} = Q_0 = -D \frac{d}{dx} [\bar{c}_M(0,s)]. \quad (10c,s)$$

The solution in transformed coordinates under the conditions stated is:

$$\bar{c}_M(x,s) = Q_0 [(\epsilon/D)(k+s)]^{-1/2} \exp\{-x[(\epsilon/D)(k+s)]^{1/2}\}. \quad (11s)$$

With the aid of the translation properties and Eq. (7), Appendix V of Carslaw and Jaeger,²⁵ the inverted form of Eq. (11s) and the solution for the mobile fraction based on the bulk volume is:

$$\epsilon C_M(x,t) = C'_M(x,t) = Q_0 \sqrt{\frac{\epsilon}{\pi D t}} \exp\left[-\left(\frac{\epsilon x^2}{4 D t} + k t\right)\right]. \quad (12)$$

The prime here means the same as subscript b in Eq. (3).

The same form would have been obtained if we had derived a solution for a species undergoing radioactive decay during the diffusion anneal; the result here is merely Eq. (3) multiplied by an attenuation factor e^{-kt} , with k representing the decay factor. However, ^{137}Cs does not decay significantly during an anneal; it is simply immobilized, yet continues to be counted. This means that the total count rate in an experiment is the cumulative value for both mobile and immobile fractions. In other words, if we integrate Eq. (12) from zero to infinity with respect to x , we find that the total amount of mobile cesium in the graphite specimen is $Q_0 e^{-kt} \text{erfc}(u=1)$ or $Q_0 e^{-kt}$; the immobilized fraction (still counting) is $Q_0 (1 - e^{-kt})$.

Thus we must compute the distribution of the immobilized cesium along x and add this to the amount given by Eq. (12) in order to obtain the total concentration detected by grinding. We seek an expression such as

$$C'_T(x,t) = C'_M(x,t) + C'_I(x,t), \quad (13)$$

where the subscript T means total. In particular, we seek the term

$$C'_I(x,t) = \int_0^t \epsilon k C'_M(x,t) dt. \quad (14)$$

In view of Eqs. (14) and (11s), we write the transform of $C'_T(x,t)$ as follows:

$$\bar{C}'_T(x,s) = (\epsilon k/s) \bar{C}'_M(x,s) = Q_0 s \sqrt{\frac{\epsilon(k+s)}{D}} \exp\{-x[(\epsilon/D)(k+s)]^{1/2}\}. \quad (14s)$$

Inversion of Eq. (14s) to obtain $C'_T(x,t)$ is accomplished by using the translation property of Eq. (31), Appendix VI of Carslaw and Jaeger.²⁵ The result, in terms of the new independent variables, $u = (x/2)(\sqrt{\epsilon/Dt})$ and $\tau = \sqrt{kt}$, is:

$$C'_T(x,t) = Q_0 \frac{u\tau}{x} [e^{-2u\tau} \operatorname{erfc}(u-\tau) - e^{+2u\tau} \operatorname{erfc}(u+\tau)]. \quad (15)$$

Next, we add Eq. (15) to Eq. (12), in terms of u and τ , to obtain $C'_T(x,t)$, evaluate this at $x = 0$, and form the ratio

$$\frac{C'_T(x,t)}{C'_T(0,t)} = \frac{e^{-(u^2 + \tau^2)} + (\tau\sqrt{\pi}/2)[e^{-2u\tau} \operatorname{erfc}(u-\tau) - e^{+2u\tau} \operatorname{erfc}(u+\tau)]}{e^{-\tau^2} + (\tau\sqrt{\pi})[1 - \operatorname{erfc}(\tau)]}. \quad (16)$$

This is the expression for the reduced concentration profile that is applicable to many studies wherein concentrations, as opposed to FR, are determined.

By using the dimensionless groups u and τ , one can readily compute appropriate values for generalized curves which, in turn, can be used to evaluate D from experimental concentration profiles after the applicable τ value has been established. We mention here that another choice of a

dimensionless penetration can be used in lieu of u , namely z , where $z = 2u\tau$ or $u = z/2\tau$. Plots of the concentration ratio as a function of either u or z , for fixed τ values, are presented in the top half of Fig. 18; tabulations of the values used to prepare these plots appear in Appendix C. While the "u plots" are more useful for correlation purposes, the "z plots" give a more realistic interpretation of the way the profiles move out as the diffusion anneal times increase. Inspection of the curves on Fig. 18 show that the logarithm of the concentration ratios become linear with u or z as τ approaches 2.2 or greater. This can be verified through Eq. (16) by using the identity $\text{erfc}(-u) = 2 - \text{erfc}(+u)$. In other words, the slope of a plot of the natural logarithm of concentration ratio vs x approaches the group $\sqrt{ek/D}$ as τ approaches 2.2 or greater. Finally, we note that the ideal relationship, Eq. (3), is recovered when $\tau \rightarrow 0$.

As mentioned before, the nature of our grinding and ^{137}Cs counting procedures makes it convenient to use an integral form of the concentration function that will give a "working equation" in terms of u and τ so that values of D/ϵ and k can be extracted from experimental FR-vs- x plots. Integration of the penetration distance is from x to infinity, which corresponds to measuring the ^{137}Cs remaining in the cylinder, not that which has been removed by grinding. The result of integrating the first term in Eq. (16), which represents the mobile fraction, is $e^{-\tau} \text{erfc}(u)$. Integration of the last two terms, which represent the immobile fraction, can be readily accomplished using the parts formula. The complete result for the total fraction of cesium remaining (FR) in the cylinder, at the penetration distance x and in terms of u and τ , is:

$$\text{FR} = (1/2)[e^{+2u\tau} \text{erfc}(u+\tau) + e^{-2u\tau} \text{erfc}(u-\tau)]. \quad (17)$$

For the experimental plots of FR vs x , we note that the fraction remaining will be an adjusted value based on an extrapolation to $x = 0$ as indicated in Fig. 17.

Plots of the FR function according to Eq. (17) as a function of u and z at fixed τ values are presented in the bottom half of Fig. 18; corresponding tabulations appear in Appendix C. When $\tau \rightarrow 2.2$ or greater,

Fig. 18. Methods of plotting generalized results of the equations accounting for internal trapping in a matrix subjected to the usual thin-layer boundary conditions. (Solutions of the diffusion equations with an impulse source function.)

the FR function approaches $\exp(-x \sqrt{\epsilon k/D})$, as in the case of Eq. (16). When $\tau \rightarrow 0$, the "ideal" relationship Eq. (3) is recovered. Comparison of the concentration and FR functions in Fig. 18 reveals a striking degree of similarity; the only difference is that the spread of the curves with respect to fixed τ values is less pronounced on the FR plots.

It seems reasonable to expect that the same phenomenon giving rise to holdup at external graphite surfaces should also take place along the internal surfaces of the same graphite, albeit with fewer dramatic effects. This seems to be the case in an experiment with H-327 graphite, the results of which are shown in a comparative plot with a Hawker-Siddeley graphite in Fig. 19. When $\tau = 0$ on this plot, ideal behavior is indicated since the solid curve represents computed values based on an integrated form of Eq. (3), namely,

$$FR = (2\sqrt{\pi}) \operatorname{erfc}(u).$$

When $\tau \neq 0$, the diffusion equations (and H-327 data) reflect the effects of an irreversible sorption rate, which acts in competition with the transient diffusion rates. The data and the computed values at $\tau = 1.25$ show fair agreement. The assumptions and resultant diffusion equations appear to have some merit.

Use of the theoretical FR-vs- u plots to extract k from τ , and D/ϵ from u , by comparisons with experimental FR-vs- x plots can result in laborious iterative procedures unless one can arrange to isolate or separate τ or u so that one does not depend directly on the other. We have found that a satisfactory way of doing this is to divide all penetrations beyond the $FR = 0.5$ value by the penetration for this particular value. Then all u , z , and x ratios are equivalent for a fixed τ since groups such as $\sqrt{\epsilon/Dt}$ or $\sqrt{\epsilon k/D}$ will cancel out. We selected the $FR = 0.5$ penetration as the base since most of the diffusion-vs-trapping rate action occurs beyond this point. This produced a set of theoretical curves, based on Eq. (16), on which experimental data could be placed directly without modification other than forming the x ratio on the basis of an interpolated $FR = 0.5$ penetration. Plots of theoretical ratio curves, with data superposed as described, are shown in Fig. 20. By treating the

Fig. 19. Comparison of generalized diffusion results obtained with an "ideal" graphite (Hawker-Siddeley curve I) and a large-grain graphite (H-327 curve II) in which internal trapping or "plate-out" occurs. (All Hawker-Siddeley data tend to fall on the $\tau = 0$ curve, regardless of the diffusion anneal times or temperature. Data for H-327 fall on different τ curves as a function of time at a fixed temperature.)

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Fig. 20. Modified plots of fraction remaining used to obtain an initial value of τ for correlation purposes.

data in this fashion, good initial estimates of the applicable τ could be obtained without prior knowledge of the D/ϵ .

Some efforts have been devoted to checking the model of cesium transport as presented in Eq. (16) by use of nonlinear least-squares methods. An examination of the data plots in Appendix B shows that the following experiments would appear susceptible to such testing: (1) experiment 7U with ATJ graphite; (2) experiments 1U, 2, 2U, 5C, 7UU, 8, 8UU, and 9UU with H-327 graphite. We have fitted data of the first three of these experiments to Eq. (16); unfortunately, the fitting procedure does not lead to acceptable agreement between calculated and observed concentrations of ^{137}Cs in the regions of small penetration, where the concentrations are changing rapidly. From these tests of the model of Eq. (16), it would appear that better agreement between observed and calculated concentrations near the initial surface will require some explanation other than that of first-order irreversible reaction. At present, no effort has been devoted to devising or testing other models.

4.3 Concerning Vapor-Phase Transport of Cesium

Three tests for a helium-sweep effect were performed with H-451 graphite; temperatures of two of the tests were about 635°C, while the third was performed at about 680°C. In addition to the usual data on count rate vs penetration and sample temperature vs time (Appendixes A and B), Appendix D lists the flow-related data of flow rate, fore pressure, back pressure, the more accurately measured pressure differential, temperature, and time for these experiments. Relative concentration data from these experiments are plotted in Figs. 21-23 vs the square root of penetration distance from the sample-pair interface. The square root was used to expand the data at small values of penetration. It is apparent, from symmetry about the 0-penetration distance, that there is no effect of the helium sweep used in these experiments.

The magnitudes of the helium flow rates are presented in Appendix D in terms of volumetric flow in 10 s. In experiment 1 (samples 1D and 1U, where D represents the downstream sample and U the upstream sample), the

Fig. 21. Cesium-137 concentration profiles in experiment 1 with H-451 graphite. (Maximum temperature = 636°C.)

Fig. 22. Cesium-137 concentration profiles in experiment 1A with H-451 graphite. (Maximum temperature = 681°C .)

Fig. 23. Cesium-137 concentration profiles in experiment 3A with H-451 graphite. (Maximum temperature = 644°C .)

flow-rate range of 34 to 92 cm³/10 s corresponds to 3.4 to 9.2 cm³/s or a superficial velocity of 2.86 to 7.75 cm/s on the basis of the graphite area. Similarly, helium flow rates in the second experiment at 635°C (experiment 3A with samples 3AD and 3AU) were 2.4 to 10 cm³/s (2.06 to 8.46 cm/s); those of experiment 1A (samples 1AD and 1AU) at 680°C ranged from 0.82 to 8.0 cm³/s (0.69 to 6.74 cm/s). Also, as may be seen in the tabulations of Appendix D, pressure differentials across a pair of H-451 samples were in the range 500 to 800 torr.

5. SUMMARY AND CONCLUSIONS

Data are presented on the transport of ¹³⁷Cs into and in five different graphites, namely ATJ, H-327, H-451, HLM-85, and HS-1-1. Temperatures of the experiments ranged from 600 to >1000°C. Also presented are flow measurements made in an effort to determine whether vapor-phase transport could be observed. Transport of ¹³⁷Cs appears to follow a simple diffusion process only in the case of the Hawker-Siddeley graphite HS-1-1. For this graphite, the following equation and parameters apply:

$$D/c = (D/c)_0 \exp[-\Delta E/RT],$$

$$(D/c)_0 \approx 4 \times 10^{-2} \text{ cm}^2/\text{s},$$

$$\Delta E \approx 30 \text{ kcal/mole}.$$

Transport of ¹³⁷Cs into the other four graphites cannot be described by a simple diffusion process since, after the diffusion anneal, there is a very steep concentration gradient near the surface to which the cesium was applied. Only at penetration distances beyond the end of this steep gradient does simple diffusion appear, from some of the data plots, to become the dominant mass transport mechanism. We used nonlinear least-squares techniques and some of the data appearing most susceptible to analysis to test a mechanism involving diffusion plus an irreversible, first-order removal of cesium. The three parameters of this model, namely surface concentration, diffusion coefficient, and chemical (or physical) reaction rate constant, were evaluated. However, the fit of data to the model was very poor, indicating that the model is unlikely to be correct.

Measurements made to test for transport of ^{137}Cs via the vapor phase within H-451 graphite showed such transport to be undetectable under the experimental conditions. These conditions involved flowing helium through a pair of samples whose contacting surfaces had been coated with ^{137}Cs . Fore pressures on the upstream sample ranged up to 100 psi, while pressure drops across the two samples were about 1 atm.

6. NOMENCLATURE

- b = subscript referring to bulk phase
- B_o = viscous flow parameter for a porous medium, cm^2
- C_i = concentration of component i in free space
- D = effective free-space diffusion coefficient (as of ^{137}Cs in graphite), cm^2/s
- D_{ij} = normal diffusion coefficient for an i-j binary mixture in free space, cm^2/s
- D_{iK} = Knudsen diffusion coefficient for component i, cm^2/s
- FR = fraction remaining
- g = subscript referring to gas
- k = reaction rate constant, usually used with a subscript
- K = combined Knudsen-viscous flow permeability coefficient for porous medium, cm^2/s
- K_o = Knudsen flow coefficient
- n = an exponent in the Freundlich sorption equation
- p = total gas pressure, dynes/cm^2 or atm
- $\langle p \rangle$ = arithmetic mean pressure, dynes/cm^2 or atm
- Δp = pressure drop across specimen, dynes/cm^2 or atm
- q = effective tortuosity factor for porous medium
- Q_o = the amount of cesium placed on the end of one cylinder of graphite
- r_o = average flow radius, cm
- $\langle r \rangle$ = mean pore radius, cm
- r_{ads} = rate of adsorption
- s = subscript referring to a surface
- t = time coordinate
- T = absolute temperature, $^\circ\text{K}$
- u = reduced dimension = $(x/2)\sqrt{\epsilon/Dt}$

- v = subscript indicating viscous flow component
- $\bar{v}$ = total mass-average velocity, cm/s
- $\bar{v}_i$ = average linear velocity (same as u_i), J_i/n_i , cm/s
- x = linear flow coordinate, cm
- z = reduced dimension = $2\pi r$
- α = coefficient of adsorption, concentration in gas/concentration on surface
- α_0 = value of α at infinite temperature
- δ_i = Knudsen fraction of total admittance for i diffusion
- ϵ = fraction of bulk volume comprised of open pores, namely the void fraction; porosity "seen" by equilibrium gas (no flow)
- ϵ' = fraction of open porosity engaged in linear steady-state flow
- ϵ'/q = effective porosity-tortuosity ratio
- η = coefficient of viscosity, poise, dynes cm s⁻²
- λ_i = decay constant of nuclide i , s⁻¹
- τ = reduced reaction rate constant = $\sqrt{kt}$

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8. APPENDIXES

8.1 Appendix A: Experimental Data

Some details of experimental procedures were given in Sect. 3. In Appendix A, we present the data from each experiment. Data are entered in a standard format in anticipation of computer-assisted manipulation and analysis. The successive entries are as follows:

1. Listing of the particular graphite (such as ATJ), the sample number (such as 4C), and the date on which the experiment was performed.
2. Two row entries listing nominal run temperatures and heating times. Two rows are used because Evans listed two half-temperatures in some experiments (such as run 7U with ATJ graphite). However, actual experimental temperature-time data are listed in entry 12, below.
3. Original length of graphite sample.
4. Original diameter of graphite sample.
5. The length to which the graphite sample was reduced after the diffusion anneal. The sample length was reduced by machining some of the graphite from the end of the cylinder opposite that to which ^{137}Cs had been applied. No ^{137}Cs was associated with the graphite removed in this manner.
6. The diameter to which the graphite sample was reduced after the diffusion anneal. The sample diameter was reduced by machining the full length. In this case, ^{137}Cs was removed.
7. The distance from sample to crystal detector and the duration of counting.
8. The count rate of the specimen mounted as shown in Fig. 13. This is, of course, the count rate corresponding to zero penetration distance.
9. The count rate of a standard ^{137}Cs source, such as 145,200 counts/min due to standard 3 used with sample 4C of the ATJ graphite.

10. The next entries are variable in number and each row consists of 1, 2, or 3 sets of the following three data:
 - (1) the count rate of the sample,
 - (2) the count rate of the standard,
 - (3) the length by which the sample length had been shortened by abrasion, in inches.
11. A commentary (\$ in column 1) indicating the start of temperature-time data.
12. The next entries are variable in number, and each row consists of one to eight sets of temperature-vs-time data. The computer-programming READ format is 8(F5.0,I3,I2). The first number in each field of 10 is temperature, in °C; the next two digits are hours of the day; and the last two digits are minutes of the hour. For example, the first complete entry for sample 4C, ATJ graphite, lists a temperature of 195°C at 8:17 AM on 8-25-74. If an experiment extended overnight, the hours of the day exceeded 24. Thus, sample 3 of graphite HS-1-1 (Hawker-Siddeley) has an entry of 700°C at 5019 h on 4-30-75. The entry of 5019 h corresponds to 0219 h (2:19 AM) on 5-2-75.

Tabulations of data from experiments with HS graphite contain additional entries, as follows: in four experiments, namely HS-1-1.I1, -I2, -I3, and -I4, the HS graphite was used as a sleeve around HLM85 graphite samples HLM85-I1, -I2, -I3, and -I4. After separating the two types of graphite, the HS sleeves were machined to increase the inner diameters as well as to reduce outer diameters. Thus, in addition to the original (outer) diameter listed according to 4, above, there is listed an original inner diameter, which is the same (0.312 in.) in all cases as the original (outer) diameter of the HLM85 samples that were pressed into the HS sleeves. Finally, the diameter to which this inner diameter was increased is listed on the same line as item 6, above.

GRAPHITE SPECIMEN : ATJ SAMPLE NO. 4C DATE : 8-23-74
 HEATING TEMP. 700 C: HEATING TIME 26 MIN.
 HEATING TEMP. C: HEATING TIME MIN.
 ORIGINAL LENGTH .239 IN.
 ORIGINAL DIAMETER .521 IN.
 REDUCED LENGTH TO .250 IN.
 REDUCED DIAMETER TO .293 IN.
 COUNTS AT 12 CM HP FOR 2 MIN.

SPECIMEN MOUNTED ON GRINDER = 121015
 STANDARD NO. 3 = 145200
 121015 145200 .00000 107870 145200 .00040 104775 147300 .00090
 103035 146000 .00172 100190 146200 .00338 96300 147000 .00645
 90345 147300 .01140 78840 147800 .02135 81010 148300 .02135
 71110 148500 .03122 59495 147800 .04642 43805 148000 .06621
 11325 148100 .08590 15300 147800 .10590 6135 147800 .12590
 1640 148600 .15590

TEMPERATURE-TIME DATA

105 0817 181 0820 272 0822 405 0825 493 0827 590 0832 659 0835 680 0836
 709 0837 718 0840 716 0842 715 0848 718 0855 717 0900 717 0906 695 0907
 678 0909 555 0910 447 0914 343 0915 091 0927

GRAPHITE SPECIMEN : ATJ SAMPLE NO. 4CU DATE : 10-29-74
 HEATING TEMP. 700 C: HEATING TIME 26 MIN.
 HEATING TEMP. C: HEATING TIME MIN.
 ORIGINAL LENGTH .225 IN.
 ORIGINAL DIAMETER .540 IN.
 REDUCED LENGTH TO .144 IN.
 REDUCED DIAMETER TO .254 IN.
 COUNTS AT 12 CM HP FOR 2 MIN.

SPECIMEN MOUNTED ON GRINDER = 253148
 STANDARD NO. 3 = 346482
 263715 346900 .00000 236626 347000 .00025 227459 346700 .00065
 220290 347800 .00175 212685 345800 .00397 204670 346800 .00741
 197370 346800 .01050 190695 346800 .01345 173967 347500 .01810
 160938 346600 .02803 136287 347900 .04295 116173 347100 .05613
 93624 347200 .07120 75602 348100 .08322 55370 346800 .09689
 47094 347900 .10250 31521 347500 .11295 25642 347700 .11755
 17360 347100 .12403 11650 346800 .12893 7098 347500 .13365
 3968 347900 .13700

TEMPERATURE-TIME DATA

105 0817 181 0820 272 0822 405 0825 493 0827 590 0832 659 0835 680 0836
 709 0837 718 0840 716 0842 715 0848 718 0855 717 0900 717 0906 695 0907
 678 0909 555 0910 447 0914 343 0915 091 0927

GRAPHITE SPECIMEN : ATJ SAMPLE NO. 5P DATE : 9-30-74
 HEATING TEMP. 717 C: HEATING TIME 1 MIN.
 HEATING TEMP. C: HEATING TIME MIN.
 ORIGINAL LENGTH .280 IN.
 ORIGINAL DIAMETER .258 IN.
 REDUCED LENGTH TO .280 IN.
 REDUCED DIAMETER TO .298 IN.
 COMPTS AT 12 CM HP FOR 2 MIN.

SPECIMEN MOUNTED ON GRINDER = 208270
 STANDARD NO. 3 = 187310
 217965 187500 .00005 188180 187000 .00005 179330 188300 .00070
 173640 183300 .00105 168835 183500 .00185 164140 188100 .00280
 158105 188000 .00480 153105 188100 .00670 144380 188100 .00978
 137950 187900 .01270 129490 188000 .01665 117285 187700 .02278
 102200 188600 .03088 85580 188500 .04086 71850 188000 .04875
 53535 188600 .05210 36065 188200 .06890 23960 187800 .07620
 15975 188400 .08280 8995 188200 .08860 1781 189100 .09610
 1250 187800 .10320 390 187000 .11070

TEMPERATURE-TIME DATA

247 1335 382 1339 457 1340 532 1343 595 1346 629 1348 641 1350 673 1352
 702 1354 716 1355 718 1356 702 1357 689 1359 668 1359 602 1400 533 1401
 480 1402 381 1403

GRAPHITE SPECIMEN : ATJ SAMPLE NO. 5B DATE : 11-13-74
 HEATING TEMP. 700 C: HEATING TIME 1 MIN.
 HEATING TEMP. 830 C: HEATING TIME 1 MIN.
 ORIGINAL LENGTH .225 IN.
 ORIGINAL DIAMETER .549 IN.
 REDUCED LENGTH TO .194 IN.
 REDUCED DIAMETER TO .288 IN.
 COMPTS AT 12 CM HP FOR 2 MIN.

SPECIMEN MOUNTED ON GRINDER = 80721
 STANDARD NO. 3 = 347778
 84869 347800 .00000 71848 346400 .00040 67435 347100 .00104
 64069 378300 .00240 61875 346000 .00439 58902 347600 .00738
 56363 347700 .01060 54229 345500 .01360 51062 346600 .01835
 46383 347200 .02730 41423 347700 .03730 35290 347800 .05214
 29755 347200 .06718 24970 347000 .08180 20611 347200 .09693
 16890 348400 .11208 13183 347700 .12508 7572 347400 .14212
 7123 349100 .15246 6002 349500 .15692 5009 349100 .16138
 4313 348500 .16197

TEMPERATURE-TIME DATA

118 1224 183 1225 204 1226 240 1227 304 1229 343 1230 372 1231 410 1232
 452 1234 516 1235 520 1237 602 1240 649 1242 679 1244 706 1245 745 1247
 760 1250 783 1252 798 1254 804 1255 818 1257 927 1258 830 1259 830 1300
 770 1301 773 1305 708 1306 642 1309 429 1315 341 1320 256 1320

GRAPHITE SPECIMEN : ATJ SAMPLE NO. 500 DATE : 11-18-74
 HEATING TEMP. 930 C: HEATING TIME 1 MIN.
 HEATING TEMP. C: HEATING TIME MIN.
 ORIGINAL LENGTH .253 IN.
 ORIGINAL DIAMETER .546 IN.
 REDUCED LENGTH TO .250 IN.
 REDUCED DIAMETER TO .545 IN.
 COUNTS AT 12 CM HP FOR 2 MIN.

SPECIMEN MOUNTED ON GRINDER = 287825
 STANDARD NO. 3 = 388100
 287229 388100 .00000 213683 388100 .00057 129889 387800 .00050
 93059 387800 .00211 75112 388400 .00337 52791 389200 .00603
 78981 388600 .00886 29182 387200 .01178 22485 388400 .01546
 12637 388700 .02015 16063 389200 .02639 13599 388000 .03322
 11938 385700 .04119 10500 388600 .05030 9558 387700 .05993
 8988 388800 .06950 8331 388100 .07915 7595 388300 .09380
 6987 387800 .10965 6095 388100 .12347 5095 388000 .14347
 3895 388600 .16135 3816 388100 .17345 2827 387900 .18397
 2889 387600 .19115 2837 387600 .19369

§ TEMPERATURE-TIME DATA

118 1228 188 1225 208 1225 280 1227 308 1229 383 1230 372 1231 410 1232
 452 1234 510 1235 520 1237 602 1240 649 1242 679 1244 706 1245 745 1247
 760 1250 783 1252 798 1254 808 1255 818 1257 927 1258 830 1259 830 1300
 770 1301 771 1305 798 1306 642 1309 829 1315 341 1320 256 1320

GRAPHITE SPECIMEN : ATJ SAMPLE NO. 500 DATE : 9-8-74
 HEATING TEMP. 717 C: HEATING TIME 7 MIN.
 HEATING TEMP. C: HEATING TIME MIN.
 ORIGINAL LENGTH .234 IN.
 ORIGINAL DIAMETER .549 IN.
 REDUCED LENGTH TO .233 IN.
 REDUCED DIAMETER TO .547 IN.
 COUNTS AT 12 CM HP FOR 2 MIN.

SPECIMEN MOUNTED ON GRINDER = 98860
 STANDARD NO. 1 = 186890
 96075 186900 .00000 72670 186500 .00030 65700 186200 .00118
 63855 186400 .00277 61585 187100 .00480 59470 187000 .00685
 58710 186200 .00990 58030 186600 .00890 57265 187300 .01091
 55733 186800 .01103 53725 188100 .01610 50370 188000 .02100
 45746 187900 .02955 39903 188200 .03947 31590 187800 .05340
 20825 187600 .07387 9270 185600 .09335 1015 185000 .10795
 877 186900 .11434 360 186900 .12816

§ TEMPERATURE-TIME DATA

125 1308 160 1350 192 1352 272 1354 388 1355 410 1358 462 1400 500 1401
 515 1404 562 1406 595 1407 643 1409 655 1410 681 1411 710 1415 716 1416
 717 1417 717 1419 716 1422 660 1425 610 1425 567 1430

GRAPHITE SPECIMEN : A-J SAMPLE NO. 70 DATE : 12-2-70
 HEATING TEMP. 500 C: HEATING TIME 1 MIN.
 HEATING TEMP. 710 C: HEATING TIME 2.5 MIN.
 ORIGINAL LENGTH .372 IN.
 ORIGINAL DIAMETER .541 IN.
 REDUCED LENGTH TO .268 IN.
 REDUCED DIAMETER TO .345 IN.
 COUNTS AT 17 CM HP FOR 2 MIN.

SPECIMEN MOUNTED ON GRINDER = 159864
 STANDARD NO. 3 = 345608
 159375 345600 .00000 156609 835860 .00055 129891 346900 .00134
 108038 345800 .00234 98054 346000 .00371 90595 345600 .00637
 92203 346900 .00932 74071 346900 .01304 65934 346800 .01782
 56961 346900 .02304 48916 345900 .02864 37164 345800 .03864
 27526 345300 .04784 17493 346000 .05770 12885 349600 .06739
 8395 346800 .07894 5541 346200 .09490 4739 345800 .10586
 4745 346800 .11584 4536 345500 .12637 4394 346800 .14000
 4155 346800 .15854 1261 347000 .16823 1243 347900 .18131
 1276 347300 .19201

8 TEMPERATURE-TIME DATA

272 1231 309 1232 347 1233 379 1234 440 1236 477 1237 486 1239 538 1240
 575 1242 590 1243 605 1244 641 1245 645 1246 673 1247 698 1248 718 1251
 717 1252 718 1253 718 1254 531 1203 518 1304 499 1306

GRAPHITE SPECIMEN : H-127 SAMPLE NO. 10 DATE : 10-9-70
 HEATING TEMP. 609 C: HEATING TIME 70 MIN.
 HEATING TEMP. C: HEATING TIME MIN.
 ORIGINAL LENGTH .532 IN.
 ORIGINAL DIAMETER .543 IN.
 REDUCED LENGTH TO .269 IN.
 REDUCED DIAMETER TO .374 IN.
 COUNTS AT 3 CM HP FOR 2 MIN.

SPECIMEN MOUNTED ON GRINDER = 262816
 STANDARD NO. 3 = 818206
 759716 818800 .00000 223769 818300 .00020 207499 818300 .00068
 198805 818500 .00164 193943 818400 .00231 189525 818400 .00330
 174450 818400 .00704 166576 818500 .00971 166576 818500 .00971
 154444 818600 .01341 143964 819900 .01741 134476 818600 .02140
 124012 819400 .02542 113575 819400 .03050 103955 818500 .03540
 93125 820400 .04142 79798 819500 .05142 71142 820800 .05737
 62287 820300 .06442 51119 819500 .07424 42513 820600 .09413
 73520 820100 .10407 24225 819200 .11432 20131 819700 .12410
 16303 819800 .13432 14490 819000 .13912 11244 820200 .14953
 8171 819800 .15452 8035 820900 .16440 5181 820300 .17920
 3430 818300 .19412 1357 820700 .21127

8 TEMPERATURE-TIME DATA

215 1227 335 1232 445 1236 567 1239 592 1241 631 1243 599 1259 597 1320
 598 1320 608 1304 608 1309 295 1357 206 1358 162 1400 110 1407

GRAPHITE SPECIMEN : H-327 SAMPLE NO. 2 DATE : 10-22-74
 HEATING TEMP. 609 C: HEATING TIME 1 MIN.
 HEATING TEMP. C: HEATING TIME MIN.
 ORIGINAL LENGTH .511 IN.
 ORIGINAL DIAMETER .504 IN.
 REDUCED LENGTH TO .250 IN.
 REDUCED DIAMETER TO .250 IN.
 COUNTS AT 12 CM HP FOR 2 MIN.

SPECIMEN MOUNTED ON GRINDER = 93858
 STAGE NO. 3 = 819514
 9365 217184 .00000 67548 817462 .00047 44479 818509 .00104
 13552 217710 .00150 30732 818726 .00360 25317 817261 .00466
 21354 216213 .00756 18129 817954 .01057 16443 817942 .01316
 14551 217327 .01645 17415 815560 .02006 12525 817212 .02327
 11770 219300 .02675 11333 820467 .03077

TEMPERATURE-TIME DATA

230 0929 205 0931 359 0933 424 0934 470 0935 494 0936 520 0937 543 0938
 567 0939 607 0940 609 0941 610 0942 567 0944 537 0945 509 0946 485 0948
 396 0949

GRAPHITE SPECIMEN : H-327 SAMPLE NO. 20 DATE : 10-16-74
 HEATING TEMP. 609 C: HEATING TIME 1 MIN.
 HEATING TEMP. C: HEATING TIME MIN.
 ORIGINAL LENGTH .375 IN.
 ORIGINAL DIAMETER .558 IN.
 REDUCED LENGTH TO .256 IN.
 REDUCED DIAMETER TO .308 IN.
 COUNTS AT 12 CM HP FOR 2 MIN.

SPECIMEN MOUNTED ON GRINDER = 313371
 STAGE NO. 3 = 817467
 308910 818100 .00000 223122 818500 .00054 146366 819000 .00065
 127219 217200 .00133 108519 817500 .00168 94463 819700 .00231
 82914 217260 .00301 69243 817700 .00390 59374 818800 .00522
 47617 217500 .00700 37521 819300 .00869 29116 818300 .01022
 24171 819100 .01170 17954 819360 .01373 12242 819500 .01576
 2513 219200 .01742 6296 820000 .01978 4126 817400 .02190
 3137 218300 .02172 2796 816700 .02477 2177 814900 .02572
 1906 218100 .02664 6610 817200 .02750 1452 817000 .02992
 1141 215800 .03094 1011 819000 .03197 929 817900 .03320
 476 218100 .03427 739 818200 .03600 716 815700 .03752
 636 818100 .03962

TEMPERATURE-TIME DATA

230 0929 205 0931 359 0933 424 0934 470 0935 494 0936 520 0937 543 0938
 567 0939 607 0940 609 0941 610 0942 567 0944 537 0945 509 0946 485 0948
 396 0949

GRAPHITE SPECIMEN : H-327 SAMPLE NO. 8 DATE : 8-28-78
 HEATING TEMP. 700 C: HEATING TIME 26 MIN.
 HEATING TEMP. C: HEATING TIME MIN.
 ORIGINAL LENGTH .256 IN.
 ORIGINAL DIAMETER .541 IN.
 REDUCED LENGTH TO .250 IN.
 REDUCED DIAMETER TO .290 IN.
 COMSIS AT 12 CM HP PCB 2 MIN.

SPECIMEN MOUNTED ON GRINDER =
 STANDARD NO. 3 =
 51767 147600 .00028 49303 146900 .00050 48579 147100 .00118
 46390 147300 .00199 46803 147500 .00188 44521 147800 .00309
 43150 147510 .00509 39186 147700 .00913 37325 147700 .01197
 15751 147200 .01105 31076 147600 .02525 27498 147700 .03561
 23195 147800 .05041 20443 147600 .06761 17313 147800 .08959
 14605 146800 .11459 12149 147700 .13595 9136 146900 .16419
 6830 146900 .18880

TEMPERATURE-TIME DATA
 120 1450 197 1452 224 1454 348 1455 385 1457 473 1458 530 1500 573 1502
 610 1504 643 1505 666 1506 679 1507 697 1508 717 1510 716 1512 717 1515
 715 1520 718 1525 716 1530 717 1536 704 1537 691 1538 661 1539

GRAPHITE SPECIMEN : H-327 SAMPLE NO. 40 DATE : 10-25-78
 HEATING TEMP. 700 C: HEATING TIME 26 MIN.
 HEATING TEMP. C: HEATING TIME MIN.
 ORIGINAL LENGTH .284 IN.
 ORIGINAL DIAMETER .542 IN.
 REDUCED LENGTH TO .179 IN.
 REDUCED DIAMETER TO .256 IN.
 COMSIS AT 9 CM HP PCB 2 MIN.

SPECIMEN MOUNTED ON GRINDER = 157482
 STANDARD NO. 3 = 602953
 164502 603900 .00000 144213 604600 .00029 135093 603000 .00079
 118164 600700 .00179 109236 602600 .00298 103230 604000 .00419
 94875 604400 .00744 95414 600900 .01264 77497 604700 .01761
 69808 603600 .02274 61387 605600 .02897 51103 605100 .03934
 42407 602100 .04951 32405 606100 .06439 24604 604400 .07959
 19059 604600 .09444 12783 603500 .10933 5133 602700 .12417
 2644 604500 .12943 1025 603700 .13372 502 605900 .13563
 384 604500 .13586

TEMPERATURE-TIME DATA
 120 1450 197 1452 224 1454 348 1455 385 1457 473 1458 530 1500 573 1502
 610 1504 643 1505 666 1506 679 1507 697 1508 717 1510 716 1512 717 1515
 715 1520 718 1525 716 1530 717 1536 704 1537 691 1538 661 1539

GRAPHITE SPECIMEN : B-327 SAMPLE NO. 5C DATE : 9-3-74

HEATING TEMP. 500 C: HEATING TIME 1 MIN.

HEATING TEMP. 716 C: HEATING TIME 1 MIN.

ORIGINAL LENGTH .274 IN.

ORIGINAL DIAMETER .549 IN.

REDUCED LENGTH TO .250 IN.

REDUCED DIAMETER TO .295 IN.

COUNTS AT 12 CM HP FOR 2 MIN.

SPECIMEN MOUNTED ON GRINDER		= 156360	
STANDARD NO. 1		= 147070	
157237	145906 .00000	132980	146306 .00026
121465	147200 .00110	116105	146206 .00230
104770	144966 .00472	101140	146000 .00629
89945	144900 .01079	81860	147400 .01310
68245	149200 .01932	57165	147506 .02515
31014	143000 .04440	22105	147200 .05510
16586	146606 .07532	7610	147600 .08536
2410	147200 .10529	1385	147606 .11520
365	147206 .13500		

§ TEMPERATURE-TIME DATA

247 1335	382 1338	457 1340	532 1343	595 1346	629 1348	641 1350	673 1352
702 1354	716 1355	718 1356	702 1357	689 1358	668 1359	602 1400	533 1401
420 1402	391 1403						

GRAPHITE SPECIMEN : B-327 SAMPLE NO. 5C DATE : 11-11-74

HEATING TEMP. 700 C: HEATING TIME 1 MIN.

HEATING TEMP. 490 C: HEATING TIME 1 MIN.

ORIGINAL LENGTH .215 IN.

ORIGINAL DIAMETER .549 IN.

REDUCED LENGTH TO .194 IN.

REDUCED DIAMETER TO .255 IN.

COUNTS AT 12 CM HP FOR 2 MIN.

SPECIMEN MOUNTED ON GRINDER		= 94119	
STANDARD NO. 3		= 346656	
94119	346923 .00000	92279	347451 .00049
84376	347243 .00310	92161	348329 .00610
77394	347232 .01182	75374	346740 .01470
63574	347923 .03406	57779	346013 .04420
44153	347212 .07704	35435	346260 .09731
17400	346391 .13227	14850	348097 .14195
10555	348094 .16144	4228	347378 .17121

§ TEMPERATURE-TIME DATA

247 1335	382 1338	457 1340	532 1343	595 1346	629 1348	641 1350	673 1352
702 1354	716 1355	718 1356	702 1357	689 1358	668 1359	602 1400	533 1401
490 1402	381 1403						

GRAPHITE SPECIMEN : H-127 SAMPLE NO. 6CU DATE : 11-6-74
 HEATING TEMP. 700 C: HEATING TIME 7.25 MIN.
 HEATING TEMP. 750 C: HEATING TIME 7.25 MIN.
 ORIGINAL LENGTH .299 IN.
 ORIGINAL DIAMETER .529 IN.
 REDUCED LENGTH TO .250 IN.
 REDUCED DIAMETER TO .407 IN.
 COUNTS AT 12 CM HP FOR 2 MIN.

SPECIMEN MOUNTED ON GRIPPER = 96516
 STANDARD NO. 1 = 346378

96516	346378	.00000	93096	346225	.00024	88174	345091	.00104
94746	345827	.00271	82214	345325	.00522	79836	346647	.00867
77922	345893	.01173	78832	345196	.01642	69704	346370	.02247
62522	346629	.03217	55796	346226	.04271	50428	346619	.05271
34698	345268	.06271	37706	346369	.07730	32285	345847	.09192
78915	346059	.10177	27184	345407	.10749	24176	345917	.11772
21958	346049	.12642	18265	346235	.14187	14796	346188	.15704
12916	347207	.16742	10993	347146	.17742	9031	346498	.18712
7611	346530	.19712	6227	347562	.20712	5049	347098	.21972
4132	347875	.22411						

TEMPERATURE-TIME DATA

125 1349	160 1350	192 1352	272 1354	384 1355	410 1358	462 1400	500 1401
535 1404	562 1406	595 1407	643 1409	655 1410	681 1411	710 1415	716 1416
717 1417	717 1419	716 1422	660 1425	630 1426	567 1430		

GRAPHITE SPECIMEN : H-127 SAMPLE NO. 700 DATE : 1-14-75
 HEATING TEMP. 500 C: HEATING TIME 1 MIN.
 HEATING TEMP. 609 C: HEATING TIME 9 MIN.
 ORIGINAL LENGTH .375 IN.
 ORIGINAL DIAMETER .558 IN.
 REDUCED LENGTH TO .269 IN.
 REDUCED DIAMETER TO .271 IN.
 COUNTS AT 12 CM HP FOR 2 MIN.

SPECIMEN MOUNTED ON GRIPPER = 136064
 STANDARD NO. 3 = 320724

139691	320700	.00000	123329	320200	.00079	101326	321600	.00158
98925	320200	.00234	80372	320600	.00336	76055	320700	.00470
70721	321700	.00671	65809	321700	.00914	60066	320400	.01280
51613	320500	.01775	41591	320000	.02429	34473	321200	.02882
26259	320500	.03503	21714	321400	.03872	17650	317000	.04347
14342	320200	.04745	11801	319400	.05137	9161	321900	.05582
6249	320400	.06037	5323	319200	.06492	3872	320400	.06980
2943	320600	.07512	2349	320030	.08132	1849	322100	.08802
1396	319500	.09642	1024	319700	.10717	942	320700	.11732
259	320100	.12712	786	319100	.13702			

TEMPERATURE-TIME DATA

320 1323	386 1325	440 1327	460 1329	485 1329	534 1330	559 1331	591 1333
609 1334	609 1335	610 1336	609 1339	608 1339	608 1340	609 1342	608 1343
578 1345	450 1353						

GRAPHITE SPECIMEN : H-327 SAMPLE NO. 8 DATE : 12-2-78
 HEATING TEMP. 500 C: HEATING TIME 1 MIN.
 HEATING TEMP. 608 C: HEATING TIME 19 MIN.
 ORIGINAL LENGTH .371 IN.
 ORIGINAL DIAMETER .562 IN.
 REDUCED LENGTH TO .250 IN.
 REDUCED DIAMETER TO .250 IN.
 COUNTS AT 12 CM HP FOR ? MIN.

SPECIMEN MOUNTED ON GRINDER = 78824
 STANDARD NO. 3 = 386792

77609	346700	.00000	69786	347000	.00045	61862	348300	.00105
59556	347200	.00129	56992	347700	.00164	55415	348800	.00214
51622	348000	.00284	52417	348000	.00343	51321	347900	.00406
50472	347300	.00477	49906	347300	.00564	48474	347400	.00678
46451	347100	.00896	43446	346500	.01345	39497	347600	.01932
35571	346900	.02506	31809	347500	.03278	27474	347400	.04252
25589	346900	.04676	23706	346000	.05065	23301	346900	.05353
21305	346500	.05824	19950	347100	.06322	18320	347700	.06764
17054	347600	.07276	15459	347500	.07798	14154	346800	.08312
13194	347000	.08906	11719	346800	.09409	9905	346400	.10122
8742	347600	.10862	7569	347600	.11564	6412	347000	.12299
5305	346800	.13032	4692	346400	.13735	3852	345800	.14564
3066	345300	.15301	2517	347000	.16036	1929	346000	.16809
1549	345500	.17504						

TEMPERATURE-TIME DATA

140 1413	302 1416	137 1425	390 1426	413 1427	426 1428	540 1430	553 1431
574 1422	591 1433	661 1434	609 1435	610 1437	608 1439	609 1442	609 1445
609 1450	609 1454	560 1457	496 1501	450 1505			

GRAPHITE SPECIMEN : H-327 SAMPLE NO. 890 DATE : 1-14-75
 HEATING TEMP. 500 C: HEATING TIME 1 MIN.
 HEATING TEMP. 609 C: HEATING TIME 9 MIN.
 ORIGINAL LENGTH .375 IN.
 ORIGINAL DIAMETER .558 IN.
 REDUCED LENGTH TO .390 IN.
 REDUCED DIAMETER TO .250 IN.
 COUNTS AT 12 CM HP FOR 2 MIN.

SPECIMEN MOUNTED ON GRINDER = 96796
 STANDARD NO. 3 = 320306

47769	319300	.00000	74129	319000	.00107	62674	319300	.00254
53933	318600	.00393	47258	313800	.00393	43701	319500	.00796
39926	319500	.01055	36460	318000	.01349	32645	318700	.01695
27524	318400	.02257	24178	319500	.02745	18792	320900	.03515
15153	319700	.04165	11916	318200	.04817	8994	318800	.05481
6844	319900	.06134	5269	319000	.06797	1941	320400	.07450
3074	319300	.08074	2230	319100	.08775	1427	318700	.09616
1104	318900	.10474	909	317800	.11166	720	318500	.12046
584	318400	.12944	456	319200	.14018	440	319000	.15041
413	319000	.16041						

TEMPERATURE-TIME DATA

370 1323	186 1325	440 1327	460 1328	485 1329	514 1330	559 1331	591 1332
609 1334	609 1335	610 1336	609 1338	608 1339	604 1340	609 1342	609 1343
576 1345	450 1353						

GRAPHITE SPECIMEN : H-327 SAMPLE NO. 907 DATE : 1-23-75
 HEATING TEMP. 500 C: HEATING TIME 1 MIN.
 HEATING TEMP. 609 C: HEATING TIME 36 MIN.
 ORIGINAL LENGTH .775 IN.
 ORIGINAL DIAMETER .558 IN.
 REDUCED LENGTH TO .214 IN.
 REDUCED DIAMETER TO .250 IN.

COUNTS AT 12 CM HP FOR 2 MIN.

SPECIMEN MOUNTED ON GRINDER = 92788

STANDARD NO. 3 = 320021

90499	320000	.00000	82210	319400	.00058	72913	320400	.00147
64352	319900	.00258	56601	320600	.00542	54996	320200	.00730
51221	319700	.01106	47732	319700	.01499	44690	319700	.01931
42014	320800	.02349	34796	319800	.02752	37695	319200	.03151
36059	319200	.03535	33965	320400	.03958	32139	320000	.04355
30239	320100	.04821	28713	320300	.05220	27041	319700	.05646
25466	319900	.06070	23530	320500	.06576	21432	319100	.07071
20176	320300	.07554	18728	319700	.08070	17120	320300	.08560
15722	320600	.09056	14612	319400	.09668	13863	319300	.10050
12470	320000	.10630	11298	320700	.11155	10166	319800	.11725
9076	319500	.12258	8548	320500	.12781	7566	320800	.13320
6784	320100	.13932	5944	320500	.14445	5283	320100	.14964
4680	320800	.15487	4224	320300	.16078	3681	319800	.16569
3220	320300	.17060	2807	320500	.17479	2322	320800	.18195
1884	319900	.18708	1611	319500	.19210	1325	320300	.19712
1120	320000	.20242						

TEMPERATURE-TIME DATA

321 1202 167 1204 432 1208 506 1210 567 1212 580 1214 609 1216 609 1216
 609 1223 609 1226 608 1230 608 1235 609 1239 609 1244

GRAPHITE SPECIMEN : H-451 SAMPLE NO. 10 DATE : 5-26-76
 HEATING TEMP. 616 C: HEATING TIME 12 MIN.
 HEATING TEMP. C: HEATING TIME MIN.
 ORIGINAL LENGTH .624 IN.
 ORIGINAL DIAMETER .444 IN.
 REDUCED LENGTH TO .480 IN.
 REDUCED DIAMETER TO .297 IN.

COUNTS AT 12 CM HP FOR 2 MIN.

SPECIMEN MOUNTED ON GRINDER = 125292

STANDARD NO. 3 = 329950

125252	313081	.00000	94575	334284	.00017	62617	334024	.00031
59934	313698	.00031	57121	333280	.00054	55730	332087	.00251
30165	334079	.00330	27280	334172	.00441	24746	333341	.00542
22902	313755	.00654	20625	313930	.00892	19388	334449	.01064
17162	334198	.01334	16813	334053	.01571	15728	333379	.01790
14625	334477	.02062	14112	334277	.02280	13337	332760	.02692
12390	313320	.02982	11988	335085	.03213	11037	332330	.03658
9948	333239	.04237	9139	315178	.04755	8463	332821	.05302
7477	334209	.06090	6462	333829	.07071	5437	334009	.08610
4415	313874	.10592	3473	313359	.13130	2883	334667	.16068
2409	334664	.20044	2155	334059	.24140			

TEMPERATURE-TIME DATA

238 0943 292 0946 300 0949 332 0959 409 1028 442 1030 500 1033 587 1039
 611 1041 634 1044 620 1046 636 1049 540 1101 477 1102 400 1105 320 1107
 249 1112

GRAPHITE SPECIMEN : R-451 SAMPLE NO. 10 DATE : 5-26-76
 HEATING TEMP. 536 C: HEATING TIME 12 MIN. RELIUM PLGW
 HEATING TEMP. C: HEATING TIME MIN.
 ORIGINAL LENGTH .500 IN.
 ORIGINAL DIAMETER .483 IN.
 REDUCED LENGTH TO .437 IN.
 REDUCED DIAMETER TO .361 IN.

COUNTS AT 12 CM HP FOR 2 MIN.

SPECIMEN MOUNTED ON GRINDER	=	302712						
STANDARD NO. 3	=	334230						
302712 334230	.00000	277139	313460	.00069	95566	334248	.00133	
42125 332775	.00185	32503	333464	.00290	27879	333566	.00360	
26357 332836	.00665	20949	312949	.00966	18488	333440	.01291	
16866 334994	.01629	15023	312656	.02175	11231	332570	.02783	
11922 334245	.03305	10562	312571	.03885	8728	334484	.04903	
7369 333231	.05863	5683	334343	.07375	4653	333261	.08687	
3964 334617	.09687	3143	333263	.11377	2713	332936	.12847	
2432 333607	.14284	2182	334405	.15975	2035	334234	.17585	
1992 332343	.18816	1940	332586	.20425				

TEMPERATURE-TIME DATA

238 0943	292 0946	300 0949	332 0959	409 1028	442 1030	500 1033	587 1039
611 1041	634 1044	620 1046	636 1049	540 1101	477 1102	400 1105	320 1107
259 1112							

GRAPHITE SPECIMEN : R-451 SAMPLE NO. 1A0 DATE : 6-9-76
 HEATING TEMP. 680 C: HEATING TIME 72 MIN.
 HEATING TEMP. C: HEATING TIME MIN.
 ORIGINAL LENGTH .674 IN.
 ORIGINAL DIAMETER .444 IN.
 REDUCED LENGTH TO .474 IN.
 REDUCED DIAMETER TO .208 IN.

COUNTS AT 0 CM HP FOR 2 MIN.

SPECIMEN MOUNTED ON GRINDER	=	35411					
STANDARD NO. 5	=	73648					
34895 73648	.00000	31108	73440	.00099	21696	73440	.00152
20929 73636	.00231	20454	73491	.00378	20384	73718	.00528
19731 74146	.00792	19322	73644	.01066	19861	73237	.01323
18659 73866	.01569	14546	73600	.02087	18477	73654	.02349
17984 73535	.02895	14184	73706	.02954	17774	73437	.03703
18018 73479	.04465	17188	73554	.05491	17158	73597	.06751
17119 73363	.08028	16552	73658	.10011	16404	73956	.12567
16244 73175	.16377	15559	74001	.20165	15346	73574	.25242
15046 73557	.30261	14207	74055	.37746	13591	73572	.45362
13071 73712	.53909	12207	73403	.60556	11277	73875	.68232
10450 73586	.74846	10448	72684	.76672	9990	73305	.94325
8976 73065	.91917						

TEMPERATURE-TIME DATA

271 1351	296 1352	235 1355	402 1359	422 1412	444 1407	667 1412	671 1418
663 1425	676 1430	690 1440	691 1444	680 1500	477 1545	292 1615	

GRAPHITE SPECIMEN : H-451 SAMPLE NO. 1A9 DATE : 6-9-76
 HEATING TEMP. 680 C: HEATING TIME 72 MIN.
 HEATING TEMP. C: HEATING TIME MIN.
 ORIGINAL LENGTH .499 IN.
 ORIGINAL DIAMETER .484 IN.
 REDUCED LENGTH TO .422 IN.
 REDUCED DIAMETER TO .196 IN.

COUNTS AT 0 CM HP FOR 2 MIN.

SPECIMEN MOUNTED ON GRINDER				=	66630				
STANDARD NO. 5				=	73587				
56630	73587	.00000	63244	73205	.00012	53204	73363	.00044	
35333	73363	.00099	29472	73569	.00150	24569	73098	.00220	
22253	73812	.00274	20375	73198	.00380	19124	73496	.00432	
18100	72882	.00512	16857	73541	.00655	16021	73716	.00760	
15636	73942	.00462	14840	73610	.01010	14074	73583	.01135	
12719	73037	.01293	13222	73069	.01473	13112	73402	.01635	
13058	73638	.01976	12851	73566	.02241				

\$ TEMPERATURE-TIME DATA

271	1351	296	1352	235	1355	402	1359	422	1402	444	1407	667	1412	671	1419
663	1425	676	1430	690	1440	681	1444	680	1500	477	1545	292	1615		

GRAPHITE SPECIMEN : H-451 SAMPLE NO. 3AD DATE : 7-13-76
 HEATING TEMP. 636 C: HEATING TIME 45 MIN.
 HEATING TEMP. C: HEATING TIME MIN.
 ORIGINAL LENGTH .624 IN.
 ORIGINAL DIAMETER .493 IN.
 REDUCED LENGTH TO .475 IN.
 REDUCED DIAMETER TO .302 IN.

COUNTS AT 12 CM HP FOR 2 MIN.

SPECIMEN MOUNTED ON GRINDER				=	75031				
STANDARD NO. 3				=	322577				
85680	322188	.00000	63510	322217	.00010	52205	321988	.00028	
15919	323057	.00064	25139	321770	.00115	20049	322245	.00168	
17746	322391	.00201	16232	318530	.00260	15269	318829	.00350	
13645	319822	.00449	12536	318225	.00594	11467	321184	.00793	
10878	320851	.00990	10378	321330	.01216	10082	321436	.01418	
9803	320289	.01908	9549	318260	.02407	9285	318390	.03408	
9023	319735	.04875							

\$ TEMPERATURE-TIME DATA

218	1002	367	1010	425	1020	427	1025	427	1107	421	1155	425	1250	438	1311
445	1315	528	1317	555	1318	575	1320	598	1322	622	1326	629	1327	633	1329
637	1329	644	1340	617	1345	633	1350	632	1356	634	1400	635	1409		

GRAPHITE SPECIMEN : H-451 SAMPLE NO. 3A0 DATE : 7-13-76
 HEATING TEMP. 636 C: HEATING TIME 45 MIN.
 HEATING TEMP. C: HEATING TIME MIN.
 ORIGINAL LENGTH .479 IN.
 ORIGINAL DIAMETER .483 IN.
 REDUCED LENGTH TO .375 IN.
 REDUCED DIAMETER TO .252 IN.
 COUNTS AT 12 CM HP FOR 2 MIN.

SPECIMEN MOUNTED ON GRINDER = 128051
 STANDARD NO. 3 = 322843
 121818 322843 .00000 57498 321688 .00059 34192 321289 .00110
 23222 322327 .00162 18204 322000 .00204 13263 323672 .00268
 9684 322508 .00374 6413 321223 .00526 4463 320755 .00690
 3689 321826 .00824 3102 320586 .01018 2690 322904 .01320
 2422 320499 .01723 2227 321140 .02133 2037 321621 .02520
 1902 321574 .02520 1794 322755 .03012

TEMPERATURE-TIME DATA

218 1002 367 1010 425 1020 427 1025 427 1107 421 1155 425 1250 438 1311
 485 1315 528 1317 555 1318 575 1320 598 1322 622 1326 625 1327 633 1328
 637 1329 644 1340 677 1345 633 1350 632 1358 674 1400 635 1409

GRAPHITE SPECIMEN : H1485 SAMPLE NO. T1 DATE : 4-1-75
 HEATING TEMP. 800 C: HEATING TIME 1260 MIN.
 HEATING TEMP. C: HEATING TIME MIN.
 ORIGINAL LENGTH .590 IN.
 ORIGINAL DIAMETER .312 IN.
 REDUCED LENGTH TO .375 IN.
 REDUCED DIAMETER TO .214 IN.
 COUNTS AT 12 CM HP FOR 2 MIN.

SPECIMEN MOUNTED ON GRINDER = 14010
 STANDARD NO. 5 = 19291
 14010 19291 .00000 13450 19275 .00035 13060 19411 .00092
 12994 19432 .00230 12360 19364 .00530 12136 19459 .00800
 11907 19525 .01320 11868 19525 .02510 11575 19525 .03095
 11476 19525 .04324 11123 19525 .06270 10774 19525 .08750
 10474 19525 .10750 10360 19525 .12720 10074 19525 .15260
 9975 19525 .17700 9926 19525 .19700 9835 19525 .20270
 9780 19525 .22580 9608 19525 .26060

TEMPERATURE-TIME DATA

248 1202 447 1205 572 1215 620 1218 685 1221 721 1225 754 1227 778 1228
 797 1328 805 1438 805 1515 805 1605 804 1735 804 1815

GRAPHITE SPECIMEN : HLM85 SAMPLE NO. 12 DATE : 3-27-75
 HEATING TEMP. 800 C: HEATING TIME 1260 MIN.
 HEATING TEMP. C: HEATING TIME MIN.
 ORIGINAL LENGTH .500 IN.
 ORIGINAL DIAMETER .312 IN.
 REDUCED LENGTH TO .375 IN.
 REDUCED DIAMETER TO .205 IN.

COUNTS AT 12 CM HP FOR 2 MIN.

SPECIMEN MOUNTED ON GRINDER				=	17103				
STANDARD NO. 5				=	19549				
17103	19549	.00000	15684	19604	.00030	15049	19537	.00055	
18256	19609	.00113	14078	19377	.00170	13812	19394	.00530	
13635	19119	.00920	13557	19291	.01430	13257	19593	.01910	
13312	19355	.02420	13303	19362	.03100	12965	19060	.03920	
12918	19285	.04880	12832	19319	.05880	12256	19404	.07120	
12303	19537	.10110	11959	19444	.11850	11916	19206	.13620	
11767	19393	.17690							

\$ TEMPERATURE-TIME DATA

218	1202	447	1205	572	1215	620	1218	685	1221	721	1225	754	1227	778	1228
797	1238	805	1438	805	1515	805	1605	804	1235	804	3315				

GRAPHITE SPECIMEN : HLM85 SAMPLE NO. 13 DATE : 3-25-75
 HEATING TEMP. 600 C: HEATING TIME 4140 MIN.
 HEATING TEMP. C: HEATING TIME MIN.
 ORIGINAL LENGTH .500 IN.
 ORIGINAL DIAMETER .312 IN.
 REDUCED LENGTH TO .312 IN.
 REDUCED DIAMETER TO .198 IN.

COUNTS AT 0 CM HP FOR 2 MIN.

SPECIMEN MOUNTED ON GRINDER				=	77450				
STANDARD NO. 5				=	71326				
77450	71326	.00000	63366	71747	.00034	61008	72248	.00051	
57285	72480	.00073	53767	72333	.00110	49994	72480	.00240	
48499	71539	.00328	46982	71998	.00450	45884	72755	.00645	
44656	72011	.00870	43874	72048	.01180	42963	71724	.01590	
42644	71957	.02210	41469	72268	.02710	40041	71935	.03410	
38972	71801	.04190	37578	71845	.04860	36226	71898	.05590	
34340	72308	.06520	32842	71821	.07630	30470	72161	.08900	
28141	71773	.10110	26377	72262	.11280	25694	72150	.12440	
22142	71875	.13990	20310	72374	.15950	17897	71617	.17950	
16406	71731	.19930	14924	72162	.21990	13952	72036	.23980	
13264	71448	.25970	12510	72222	.27960				

\$ TEMPERATURE-TIME DATA

206	1047	271	1048	346	1050	390	1051	416	1052	451	1053	499	1055	532	1056
556	1057	500	1058	592	1059	600	1100	602	1105	598	1135	592	1145	599	1150
600	1157	601	7947	600	8006	574	8008	549	8010	510	8011	509	8012	500	8013
460	8017	425	8021	393	8025										

GRAPHITE SPECIMEN : H1885 SAMPLE NO. 14 DATE : 3-21-75
 HEATING TEMP. 600 C: HEATING TIME 4180 MIN.
 HEATING TEMP. C: HEATING TIME MIN.
 ORIGINAL LENGTH .500 IN.
 ORIGINAL DIAMETER .312 IN.
 REDUCED LENGTH TO .375 IN.
 REDUCED DIAMETER TO .287 IN.
 COUNTS AT 0 CM HP FOR 2 MIN.

SPECIMEN MOUNTED ON GRINDER = 93062
 STANDARD NO. 5 = 72129
 93062 72129 .00000 78833 71998 .00075 75063 72770 .00205
 55785 72044 .00255 51878 72315 .00345 50716 72415 .00445
 89258 72397 .00545 88135 72021 .00995 86480 72132 .01395
 85807 71783 .01775 83755 71950 .02285 82777 72065 .02625
 80635 71908 .03245 78819 71960 .03865 35538 72240 .04525
 73275 72423 .05345 29692 72084 .06375 26978 72674 .07345
 24139 71937 .08585 20300 72538 .10015 19668 72453 .11135
 17692 72233 .11555 16189 72083 .13051 14988 72044 .14571
 13947 72082 .16071 13455 72386 .20070

TEMPERATURE-TIME DATA
 206 1047 271 1048 346 1050 390 1051 416 1052 451 1053 499 1055 532 1056
 556 1057 580 1058 592 1059 600 1100 602 1105 598 1135 598 1145 599 1150
 600 1157 601 7947 600 8006 574 8008 549 8010 510 8011 509 8012 500 8013
 460 8017 425 8021 393 8025

GRAPHITE SPECIMEN : H5-1-1 SAMPLE NO. 11 DATE : 3-18-75
 HEATING TEMP. 800 C: HEATING TIME 1260 MIN.
 HEATING TEMP. C: HEATING TIME MIN.
 ORIGINAL LENGTH .627 IN.
 ORIGINAL DIAMETER .714 IN. ORIGINAL INNER DIAMETER .312 IN.
 REDUCED LENGTH TO .500 IN.
 REDUCED DIAMETER TO .556 IN. INCREASED INNER DIAMETER TO .365 IN.
 COUNTS AT 12 CM HP FOR 2 MIN.

SPECIMEN MOUNTED ON GRINDER = 292027
 STANDARD NO. 3 = 313695
 292027 313695 .00000 287437 312713 .00034 276654 312986 .00292
 267577 311721 .00554 256009 312327 .00834 239394 312144 .01234
 219220 312302 .01744 199494 312921 .02224 177886 311295 .02784
 151655 311968 .03444 124666 311991 .04264 98767 311879 .05144
 78350 312594 .06144 55185 311974 .07174 39672 312352 .08174
 28809 311765 .09154 19759 311393 .10264 14069 311896 .11244
 9804 312401 .12474 7051 313293 .13764 5092 311758 .15154
 4223 312087 .16484

TEMPERATURE-TIME DATA
 298 1202 447 1205 572 1215 620 1218 685 1221 721 1225 754 1227 778 1228
 777 1338 805 1438 805 1515 805 1605 804 3235 804 3315

GRAPHITE SPECIMEN : HS-1-1 SAMPLE NO. 12 DATE : 3-11-75
 HEATING TEMP. 800 C: HEATING TIME 1260 MIN.
 HEATING TEMP. C: HEATING TIME MIN.
 ORIGINAL LENGTH .625 IN.
 ORIGINAL DIAMETER .714 IN. ORIGINAL INNER DIAMETER .312 IN.
 REDUCED LENGTH TO .500 IN.
 REDUCED DIAMETER TO .576 IN. INCREASED INNER DIAMETER TO .328 IN.
 COUNTS AT 12 CM TP PCB 2 MIN.

SPECIMEN MOUNTED ON GRINDER = 318643
 STANDARD NO. 3 = 318839
 318971 318643 .00000 316703 320240 .00007 311541 318468 .00137
 297629 318915 .00456 279175 318639 .00902 254302 318804 .01320
 209591 318641 .02466 181342 318641 .03156 151032 318291 .03954
 125709 317084 .04744 103921 317637 .05475 82452 318908 .06373
 64063 318334 .07178 50141 317416 .07955 38769 318116 .08735
 28200 318155 .09744 20845 317125 .10783 18645 317703 .11790
 10946 319100 .12813 8243 317535 .13860 6859 318400 .14923
 6061 317565 .15897 5124 317740 .16980 5124 317740 .16980
 4341 317201 .17843

§ TEMPERATURE-TIME DATA

238 120 447 1205 572 1215 620 1218 685 1221 721 1225 754 1227 778 1228
 797 1231 805 1238 805 1245 805 1250 804 1255 804 1258

GRAPHITE SPECIMEN : HS-1-1 SAMPLE NO. 13 DATE : 3-20-75
 HEATING TEMP. 600 C: HEATING TIME 4140 MIN.
 HEATING TEMP. C: HEATING TIME MIN.
 ORIGINAL LENGTH .625 IN.
 ORIGINAL DIAMETER .714 IN. ORIGINAL INNER DIAMETER .312 IN.
 REDUCED LENGTH TO .500 IN.
 REDUCED DIAMETER TO .549 IN. INCREASED INNER DIAMETER TO .365 IN.
 COUNTS AT 12 CM TP PCB 2 MIN.

SPECIMEN MOUNTED ON GRINDER = 334618
 STANDARD NO. 3 = 311867
 334618 311867 .00000 324421 312525 .00031 290455 312019 .00180
 239366 311585 .00174 191235 313168 .00530 156246 313420 .00700
 123415 313578 .00830 89031 311862 .01020 61004 312697 .01220
 14972 312602 .01460 20551 312432 .01700 10743 312824 .01930
 5494 311823 .02260 3525 313220 .02540 2799 312536 .02930
 7556 312213 .03460

§ TEMPERATURE-TIME DATA

206 1047 271 1048 346 1050 390 1051 416 1052 451 1053 499 1055 532 1056
 554 1057 580 1058 592 1059 600 1100 602 1105 598 1135 592 1145 599 1150
 600 1157 601 7947 600 8006 574 8008 549 8010 510 8011 509 8012 500 8013
 460 8017 425 8021 393 8025

GRAPHITE SPECIMEN : HS-1-1 SAMPLE NO. 10 DATE : 3-17-75
 HEATING TEMP. 600 C: HEATING TIME 2140 MIN.
 HEATING TEMP. C: HEATING TIME MIN.
 ORIGINAL LENGTH .625 IN.
 ORIGINAL DIAMETER .714 IN. ORIGINAL INNER DIAMETER .312 IN.
 REDUCED LENGTH TO .375 IN.
 REDUCED DIAMETER TO .558 IN. INCREASED INNER DIAMETER TO .365 IN.
 COUNTS AT 12 CM HP FOR 2 MIN.
 SPECIMEN MOUNTED ON GRINDER = 483608
 STANDARD NO. 3 = 317579
 483884 317576 .0000% 472752 317786 .00053 399363 317808 .00309
 220129 317264 .00767 151867 317C19 .00985 85623 316982 .01287
 70438 317887 .01721 8378 317199 .02267 48846 316923 .02725
 4017 316914 .03275 3888 317CP7 .03737

§ TEMPERATURE-TIME DATA

206 1047	271 1088	346 1050	390 1051	416 1052	451 1053	499 1055	532 1056
556 1057	580 1058	592 1059	500 1100	602 1105	598 1135	598 1145	599 1150
600 1157	601 7987	600 8606	574 8009	549 8010	510 8011	509 8012	500 8013
440 4017	425 4021	393 4025					

GRAPHITE SPECIMEN : HS-1-1 SAMPLE NO. 1 DATE : 4-7-75
 HEATING TEMP. 1000 C: HEATING TIME 2 MIN.
 HEATING TEMP. C: HEATING TIME MIN.
 ORIGINAL LENGTH .187 IN.
 ORIGINAL DIAMETER .714 IN.
 REDUCED LENGTH TO .181 IN.
 REDUCED DIAMETER TO .241 IN.
 COUNTS AT 12 CM HP FOR 2 MIN.
 SPECIMEN MOUNTED ON GRINDER = 232025
 STANDARD NO. 3 = 314808
 232025 314819 .00000 220928 313519 .00120 207434 313966 .00220
 191542 313594 .00386 169319 313491 .00472 140808 311961 .00652
 111225 313310 .00867 79844 313210 .01076 56228 313210 .01322
 70587 313633 .01790 11572 311481 .02102 4221 312562 .02840
 2237 311813 .03250 1607 311613 .03718

§ TEMPERATURE-TIME DATA

290 1356	473 1400	592 1406	691 1410	840 1421	879 1430	895 1434	921 1440
942 1447	950 1450	963 1455	987 1497	1000 1518	1001 1519	1000 1520	959 1521
827 1525	680 1533	460 1554					

GRAPHITE SPECIMEN : HS-1-1 SAMPLE NO. 3 DATE : 4-30-75
 HEATING TEMP. 700 C: HEATING TIME 2400 MIN.
 HEATING TEMP. C: HEATING TIME MIN.
 ORIGINAL LENGTH .625 IN.
 ORIGINAL DIAMETER .714 IN.
 REDUCED LENGTH TO .320 IN.
 REDUCED DIAMETER TO .312 IN.
 COUNTS AT 12 CM HP FOR 2 MIN.

SPECIMEN MOUNTED ON GRINDER = 250419
 STANDARD NO. 3 = 313428
 249922 313428 .00006 246931 312278 .00070 235746 312995 .00180
 221249 314434 .00320 204108 311751 .00488 187214 312378 .00652
 172216 311264 .00746 155402 312724 .00940 134641 313470 .01157
 123564 313343 .01284 111664 312173 .01430 97491 312189 .01585
 84114 310172 .01742 72444 309169 .01900 61611 311055 .02043
 52374 310238 .02160 43384 309057 .02320 35869 308998 .02442
 30044 309839 .02604 27701 310044 .02834 16881 309208 .02980
 13004 308241 .03151 10289 309067 .03298 7521 308480 .03498
 5809 309773 .03651 4360 307962 .03820 3415 306396 .03985
 2622 307221 .04163 2112 307854 .04350 1644 309471 .04546
 1482 308074 .04747 1200 309319 .04982 1084 309275 .05200
 1023 309084 .05398

8 TEMPERATURE-TIME DATA
 334 1549 355 1551 426 1553 481 1555 544 1557 600 1559 647 1601 688 1603
 697 1615 700 1606 698 1615 674 2529 700 2609 699 2819 699 3019 700 3234
 700 3441 700 5019 702 5049 669 5051 609 5056 551 5101 467 5108 348 5124
 259 5126

GRAPHITE SPECIMEN : HS-1-1 SAMPLE NO. 5 DATE : 4-23-75
 HEATING TEMP. 1000 C: HEATING TIME 150 MIN.
 HEATING TEMP. C: HEATING TIME MIN.
 ORIGINAL LENGTH .625 IN.
 ORIGINAL DIAMETER .714 IN.
 REDUCED LENGTH TO .312 IN.
 REDUCED DIAMETER TO .302 IN.
 COUNTS AT 12 CM HP FOR 2 MIN.

SPECIMEN MOUNTED ON GRINDER = 198196
 STANDARD NO. 3 = 313978
 198196 313798 .00000 186498 311715 .00025 183109 314211 .00122
 175904 313726 .00295 167641 314526 .00465 152333 314206 .00832
 129374 312284 .01362 106664 312839 .01922 82197 313041 .02518
 61894 311002 .03054 49688 311757 .03450 37783 312435 .03900
 27721 312764 .04354 20358 312783 .04798 15200 312862 .05166
 10787 311342 .05628 7615 315504 .06008 5540 311977 .06400
 3923 311471 .06840 2600 310633 .07345 1951 311569 .07790
 1534 311247 .08199 1225 310880 .08701 1102 313190 .09107
 966 311624 .09650

8 TEMPERATURE-TIME DATA
 240 0851 398 0855 558 0903 630 0911 754 0915 810 0919 834 0923 864 0925
 892 0929 916 0933 933 0938 949 0943 962 0947 973 0951 983 0955 988 0959
 996 1004 998 1006 1000 1007 1002 1009 1004 1020 1004 1030 1003 1143 1001 1224
 1001 1237 936 1239 807 1242 686 1250 466 1310

8.2 Appendix B: Plots of Diffusion Data

The 36 plots included in this appendix display all of the ^{137}Cs -diffusion data in terms of relative concentration vs both penetration distance (the upper points $\circ$) and the square of penetration distance (the lower points Δ). The temperature-time data for temperatures greater than 400°C are also displayed. Each plot is labeled according to the type of graphite (ATJ, H-327, H-451, HLM-85, and HS-1-1) and experiment number. It is apparent that some features of these plots will be difficult to explain. Experiments with the HS graphite are obviously the simplest in appearance; analysis of these data is described in Sect. 4.1. Some of the other experimental data appear to conform to Eq. (16). Included in this category are the following: experiment 7U with the ATJ graphite; experiments 1U, 2, 2U, 5C, 7UU, 8, 8UU, and 9UU with H-327; and experiments 1D and 1U with H-451. Some of the other experiments were performed for times either too short or too long, at the selected temperature, for the data to be conveniently analyzed.

8.3 Appendix C: Evaluation of Eqs. (16) and (17)

Graphical methods can be used to evaluate the dimensionless variables u and τ of Eqs. (16) and (17). Table C.1 lists values of the concentration ratio expressed in Eq. (16), while Table C.2 lists values of the fraction remaining of Eq. (17) for the appropriate ranges of u and τ .

Table C.1. Computed values of the concentration-ratio function^a for diffusion with first-order plate-out in an infinitely long medium with an instantaneous-plane (impulse) source

u	CR ^b , concentration ratios at τ values of:									
	0.00	0.25	0.50	0.75	1.00	1.25	1.50	1.75	2.00	2.20
0	1.0000	1.0000	1.0000	1.0000	1.0000	1.0000	1.0000	1.0000	1.0000	1.0000
0.05	0.9975	0.9877	0.9639	0.9360	0.9091	0.8844	0.8615	0.8397	0.8188	0.8026
0.10	0.9900	0.9716	0.9272	0.8754	0.8264	0.7822	0.7422	0.7052	0.6705	0.6441
0.20	0.9608	0.9285	0.8515	0.7633	0.6821	0.6118	0.5509	0.4973	0.4495	0.4149
0.30	0.9139	0.8722	0.7732	0.6618	0.5617	0.4781	0.4088	0.3507	0.3014	0.2672
0.40	0.8521	0.8048	0.6934	0.5695	0.4609	0.3731	0.3032	0.2473	0.2021	0.1721
0.50	0.7788	0.7292	0.6132	0.4858	0.3763	0.2905	0.2247	0.1743	0.1355	0.1109
0.60	0.6977	0.6486	0.5342	0.4100	0.3053	0.2253	0.1662	0.1228	0.0908	0.0714
0.70	0.6126	0.5661	0.4581	0.3420	0.2458	0.1740	0.1226	0.0864	0.0608	0.0460
0.80	0.5273	0.4847	0.3865	0.2816	0.1960	0.1336	0.0902	0.0607	0.0407	0.0296
1.00	0.3679	0.3355	0.2611	0.1830	0.1207	0.0769	0.0481	0.0297	0.0182	0.0122
1.20	0.2369	0.2148	0.1642	0.1117	0.0706	0.0426	0.0249	0.0143	0.0080	0.0050
1.40	0.1409	0.1271	0.0959	0.0637	0.0390	0.0225	0.0125	0.0067	0.0035	0.0020
1.60	0.0773	0.0695	0.0519	0.0339	0.0202	0.0113	0.0060	0.0030	0.0015	0.0008
1.80	0.0392	0.0351	0.0260	0.0168	0.0098	0.0053	0.0027	0.0013	0.0006	0.0003
2.00	0.0183	0.0164	0.0121	0.0077	0.0044	0.0023	0.0012	0.0005	0.0002	0.0001
2.20	0.0079	0.0071	0.0052	0.0033	0.0019	0.0010	0.0005	0.0002	0.0001	0.0000

^aEq. (16) in text; values plotted in Fig. 15; $\tau = \sqrt{kt}$; $u = \frac{x}{2} \sqrt{\frac{c}{Dt}}$.

^bCR = $C_T(x,t)/C_T(0,t)$; includes mobile and immobile fractions.

Table C.2. Computed values of the fraction-remaining function^a for diffusion with first-order plate-out in an infinitely long medium with an instantaneous-plane (impulse) source

u	FR ^b , fraction remaining ^b at τ values of:									
	0.00	0.25	0.50	0.75	1.00	1.25	1.50	1.75	2.00	2.20
0.00	1.0000	1.0000	1.0000	1.0000	1.0000	1.0000	1.0000	1.0000	1.0000	1.0000
0.05	0.9436	0.9404	0.9313	0.9173	0.8998	0.8803	0.8598	0.8392	0.8186	0.8025
0.10	0.8875	0.8817	0.8651	0.8399	0.8088	0.7745	0.7391	0.7041	0.6701	0.6440
0.20	0.7773	0.7673	0.7407	0.7000	0.6503	0.5980	0.5455	0.4954	0.4490	0.4146
0.30	0.6714	0.6598	0.6270	0.5783	0.5205	0.4601	0.4013	0.3482	0.3007	0.2669
0.40	0.5716	0.5592	0.5243	0.4731	0.4134	0.3524	0.2951	0.2445	0.2012	0.1718
0.50	0.4795	0.4672	0.4329	0.3831	0.3257	0.2684	0.2161	0.1713	0.1345	0.1105
0.60	0.3961	0.3847	0.3527	0.3066	0.2544	0.2031	0.1575	0.1197	0.0899	0.0710
0.70	0.3222	0.3119	0.2833	0.2424	0.1967	0.1526	0.1143	0.0834	0.0599	0.0456
0.80	0.2579	0.2490	0.2243	0.1892	0.1504	0.1137	0.0824	0.0579	0.0399	0.0293
1.00	0.1573	0.1512	0.1343	0.1106	0.0849	0.0613	0.0419	0.0275	0.0175	0.0120
1.20	0.0897	0.0859	0.0754	0.0610	0.0455	0.0316	0.0206	0.0127	0.0075	0.0048
1.40	0.0477	0.0456	0.0397	0.0316	0.0230	0.0155	0.0097	0.0057	0.0032	0.0019
1.60	0.0237	0.0225	0.0195	0.0153	0.0110	0.0072	0.0044	0.0025	0.0013	0.0007
1.80	0.0109	0.0104	0.0089	0.0069	0.0049	0.0031	0.0019	0.0010	0.0005	0.0003
2.00	0.0047	0.0044	0.0038	0.0029	0.0020	0.0013	0.0007	0.0004	0.0002	0.0001
2.20	0.0019	0.0018	0.0015	0.0012	0.0008	0.0005	0.0003	0.0001	0.0001	-

^aEq. (17) in text; values plotted in Fig. 15; $\tau = \sqrt{kt}$; $u = \frac{x}{\tau} \sqrt{\frac{D}{k}}$.

^bFR values result from integrating concentration function which includes mobile and immobile fractions.

8.4 Appendix D: Helium Flow During Experiments with H-451 Graphite

As stated in Sect. 5.3, helium flow through the graphite was maintained during experiments with H-451 for the purpose of testing for vapor-phase transport of cesium. Figures 21-23 show that no such transport occurred under conditions of these experiments. Helium flow data are listed in the three tables of this appendix. The first five lines in each table are self-explanatory; data, of which there are two sets per line, are as follows:

FTIME = the time required for 10 cm^3 of helium (at nearly standard conditions) to flow through the two abutting samples of graphite, s;

PFORE = the fore pressure on the graphite, psig;

PBACK = the back pressure on the graphite, psig;

DELP = the pressure differential across the graphite samples (more accurate than PFORE-PBACK), torr;

PTEMP = temperature of the graphite at the time of measurement, °C;

THRS = the clock hour at the time of measurement;

TMNS = the clock minute at the time of measurement.

These data are read with format (2(F6.,2F5.,2F6.,I3,I2)).

Permeabilities calculated from the tabular data are plotted in Figs. D-1, D-2, and D-3 to provide a qualitative view of the tightness with which the graphite was sealed to the 0.006-in.-wall stainless steel tubing (Figs. 8 and 10).

Thermal cycling in experiment H-451.1 (samples 1D and 1U of Appendixes A and B) was as follows: (1) heat to about 635°C for ^{137}Cs diffusion experiment; (2) cool to room temperature; (3) reheat to 400°C for permeability measurements; (4) cool to 300°C, then successively to 200, 100, and 22°C for further permeability measurements, as shown in Fig. D-1. It is apparent that the graphite--stainless steel seal was relatively tight at 400 and 300°C but leaked as the system cooled to room temperature. The leak was reduced, or eliminated, before the

GRAPHITE SPECIMEN : H-451 SAMPLE NO. 1 DATE : 5-26-76
 ORIGINAL LENGTH 1.124 IN. BAROMETRIC PRESSURE = 736.8 TORR
 ORIGINAL DIAMETER .483 IN.
 RELIUM FLOW TIME (SEC/10 CC), FCRF PRESSURE (PSIG), BACK PRESSURE (PSIG), PRESSURE DIFFERENTIAL (TORR), TEMPERATURE (DEG C), TIME

79.	21.5	7.	765.	409.	1254	76.5	21.5	7.	762.	420.	1301
79.	21.5	7.	770.	425.	1310	64.	21.4	6.5	773.	399.	1329
67.5	21.4	6.2	821.	399.	1329	75.6	21.4	5.7	829.	397.	1330
75.	21.4	5.6	837.	396.	1340	75.2	21.4	5.6	837.	396.	1344
70.5	21.3	5.9	822.	309.	1348	69.8	21.3	6.1	913.	394.	1410
68.2	21.3	6.1	803.	299.	1414	67.2	21.3	6.2	875.	392.	1418
67.8	21.3	6.2	873.	305.	1426	67.6	21.3	6.2	873.	304.	1432
67.2	21.3	6.2	802.	304.	1438	61.2	21.2	6.6	782.	219.	1448
59.3	21.2	6.9	762.	219.	1507	58.2	21.2	6.9	762.	219.	1518
58.2	21.2	6.9	762.	219.	1528	58.2	21.2	6.9	762.	220.	1531
58.1	21.2	8.	791.	109.	1536	49.6	21.2	8.	698.	98.	1559
49.3	21.2	8.4	682.	99.	1603	46.5	21.2	8.4	678.	100.	1611
46.2	21.2	8.4	690.	99.	1616	46.2	21.2	8.4	589.	22.	1612
38.5	21.0	9.8	599.	22.	1615	38.5	21.	9.8	589.	22.	1623
38.0	21.	9.7	596.	22.	1634	38.2	21.	9.7	616.	22.	1642
41.0	21.	9.3	616.	22.	1659	34.	52.2	77.	763.	21.5	1339
37.2	46.7	71.8	793.	21.5	1437	16.5	79.5	64.2	759.	21.5	1452
44.5	71.7	57.5	772.	21.	1520	47.	60.7	45.9	746.	21.	1538
55.5	52.	17.5	760.	22.	1600	62.5	41.3	27.2	740.	22.	1642
75.	22.3	18.1	719.	22.	1625	92.	22.6	7.4	796.	22.	1005

GRAPHITE SPECIMEN : H-451 SAMPLE NO. 1A DATE : 6-9-76
 ORIGINAL LENGTH 1.123 IN. BAROMETRIC PRESSURE = 743.1 TORR
 ORIGINAL DIAMETER .484 IN.
 RELIUM FLOW TIME (SEC/10 CC), FCRF PRESSURE (PSIG), BACK PRESSURE (PSIG), PRESSURE DIFFERENTIAL (TORR), TEMPERATURE (DEG C), TIME

8.2	37.0	21.1	879.	463.	1425	8.5	37.0	21.1	813.	676.	1430
8.5	37.0	21.1	916.	680.	1440	8.2	37.0	21.1	818.	681.	1448
9.5	37.0	21.1	920.	680.	1500	14.	37.0	21.1	796.	477.	1545
20.	96.8	92.0	766.	22.5	1094	28.5	84.8	70.6	712.	22.5	1065
26.	77.0	62.2	794.	22.5	1015	29.	69.6	53.8	798.	22.5	1025
33.	56.1	47.9	758.	22.5	1048	38.5	49.8	37.2	741.	22.5	1103
41.8	19.0	23.7	748.	22.5	1158	58.2	27.8	13.9	722.	22.5	1210
80.0	14.8	1.4	713.	22.5	1221						

GRAPHITE SPECIMEN : H-451 SAMPLE NO. 2A DATE : 7-13-76
 ORIGINAL LENGTH 1.124 IN. BAROMETRIC PRESSURE = 738.8 TORR
 ORIGINAL DIAMETER .483 IN.
 RELIUM FLOW TIME (SEC/10 CC), FCRF PRESSURE (PSIG), BACK PRESSURE (PSIG), PRESSURE DIFFERENTIAL (TORR), TEMPERATURE (DEG C), TIME

51.	36.8	20.0	520.	422.	1040	52.	36.8	22.8	712.	417.	1050
100.	36.6	22.5	720.	424.	1104	102.	36.6	22.5	720.	426.	1112
100.	36.6	22.5	725.	421.	1155	100.	36.6	22.0	724.	424.	1240
95.	36.6	22.5	679.	444.	1340	95.	36.6	23.	679.	637.	1345
95.	36.6	23.	665.	472.	1358	95.	36.6	23.5	642.	635.	1404
24.3	96.7	91.6	753.	21.5	1042	27.8	77.2	62.2	775.	21.5	1047
31.3	70.	55.2	752.	21.5	1005	34.3	59.3	42.6	778.	21.5	1048
47.6	46.8	35.5	713.	21.5	1133	48.1	38.2	23.8	739.	21.5	1203
59.3	28.4	13.6	738.	21.5	1240	81.7	14.9	0.5	746.	21.2	1310

ORNL DWG 79-1316

Fig. D-1. Helium permeabilities measured with H-451 graphite samples 1U/1D.

ORNL DWG 79-1315

Fig. D-2. Helium permeabilities measured with H-451 graphite samples 1AU/1AD.

ORNL DWG 79-1317

Fig. D-3. Helium permeabilities measured with H-451 graphite samples 3AU/3AD.

pressure was increased to 7.2 atm by increasing pressure on the jack (Fig. 8); calculated permeability parameters of the linear least-squares fit are given below. In Fig. D-1, numbers beside points are the orders in which data were taken.

Thermal cycling in experiment H-451.1A (samples 1AD and 1AU of Appendixes A and B) was as follows: (1) heat samples to about 675°C for ¹³⁷Cs diffusion experiment, taking permeability data during the time at this temperature; (2) cool to 477°C and take one set of permeability data; (3) cool to room temperature and take additional data.

Thermal cycling in experiment H-451.3A (samples 3AD and 3AU of Appendixes A and B) was as follows: (1) heat samples to 400°C and start taking permeability data; (2) continue heating to about 640°C for ¹³⁷Cs diffusion and simultaneously take permeability data; (3) cool to room temperature to take additional data.

It is not possible to state that no helium bypassed the two graphite samples of each experiment during the times that these were at anneal temperature. The data of Figs. D-1, D-2, and D-3, do, however, suggest that a significant fraction of the helium did pass through the graphite. This is particularly apparent in Fig. D-3.

Permeability parameters for 21-23°C obtained by linear least-squares analysis (the straight lines of Figs. D-1, D-2, and D-3), and referred to Eq. (D-1), are tabulated below. In each heading, the term $\hat{S}(\)$ is the standard deviation of the quantity in parentheses; $\hat{S}(\text{fit})$ is the standard deviation of fit of data to the model, namely to the equation

$$K = D_{2K} + (B_o/\eta) <p> . \quad (\text{D-1})$$

<u>Expt.</u>	$\hat{S}(\text{fit})$ <u>(cm²/s)</u>	$D_{2K} \pm \hat{S}(D_{2K})$ <u>(cm²/s)</u>	$B_o/\eta \pm \hat{S}(B_o/\eta)$ <u>(cm² s⁻¹ atm⁻¹)</u>
1	0.0286	0.0813 $\pm$ 0.0292	0.0853 $\pm$ 0.0061
1A	0.0518	0.0848 $\pm$ 0.0472	0.1378 $\pm$ 0.0099
3A	0.0096	0.1123 $\pm$ 0.0089	0.1196 $\pm$ 0.0020