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EVALUATION OF (Th,U)C₂, CARBON-COATED (Th,U)C₂
PARTICLES, AND CARBON COATINGS

by

G. B. Engle, C. S. Luby, and J. C. Bokros

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April 26, 1962

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EVALUATION OF $(Th,U)C_2$, CARBON-COATED $(Th,U)C_2$ PARTICLES, AND CARBON COATINGS*

by

G. B. Engle, C. S. Luby, and J. C. Bokros

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ABSTRACT

Thorium and uranium carbide nuclear fuel particles were evaluated by metallographic and X-ray diffraction techniques. Techniques were developed to etch the polished surface of thorium-uranium carbide to reveal the grain structure. In addition, techniques to determine particle density and coating thickness were developed. Comparison of the data indicates that the use of spherical particles allowed for more precise determination of the coating thickness, density, and strength of coatings. Strength of individual particles was about 700 to 1300 grams per particle. A large scatter in crushing-strength values was observed when individual particles were crushed. Consequently, a relative crushing-strength test was developed for comparing coating strength: loads were applied to a column of particles in stepped increments, and broken coatings were detected by observing a weight gain in moist air from hydrolysis of the thorium-uranium carbides. A correlation of relative crushing strength and coating thickness was obtained.

The crystal structure of the carbon coatings was found to depend on the temperature of deposition in the range from 1400° to 2400°C; the two-dimensional structure became more defined with increasing deposition temperatures. Subsequent annealing at 2400°C of the coatings deposited at 1400°C and at 2200° to 2400°C produced graphitization, whereas those deposited at 1800° to 2000°C retained their two-dimensional character.

Macrostructure of the coatings as revealed by metallographic techniques showed a concentric shell-like structure in the 1400°C deposits and mottled or conical structure in the 1800°C deposits. Annealing to higher temperatures did not alter the general appearance of either macrostructure.

I. INTRODUCTION

One of the primary requisites of graphite-base fuel elements for use in high-temperature, gas-cooled reactors is associated with the ability of the elements to delay the escape of fission products from the fuel body into the coolant stream. This may be accomplished by coating each fuel particle with a barrier material. A suitable material for this purpose is a pyrolytic carbon coating. If the fuel particles contain thorium carbide, the coating serves the additional purpose of protecting the particles from attack by moisture in the air, and thereby greatly facilitates the handling of the fuel during manufacture of the fuel elements.

A considerable amount of experimental work has been carried out correlating the structure of massive pyrocarbon as deposited on commercial graphite substrates with different fabrication techniques.^{1, 2, 3} This material has been called "pyrolytic graphite."⁴ The bulk of the experimental work on the evaluation of pyrolytic carbons has been carried out on materials deposited from a variety of hydrocarbon gases in the pressure range from less than 1 to 150 mm Hg and in the temperature range from 1200° to 2300°C.

The portion of this paper dealing with the carbon structure of the coatings differs from much of the published work in that the carbon coatings discussed here were deposited from a hydrocarbon gas onto small carbide particles (100- to 400- μ diameter) in a fluidized bed. The hydrocarbon gases were decomposed in the presence of a helium carrier gas at relatively high partial pressures ($\sim$ 100 to 160 mm Hg) and at a relatively high velocity.

This report discusses metallographic techniques for handling coated and uncoated thorium-uranium carbide particles, as well as several methods of evaluating the coated particles and coatings.

II. EXPERIMENTAL PROCEDURES AND RESULTS

Preparation of Coated Particles

The coating experiments were conducted on $(\text{Th}, \text{U})\text{C}_2$ particles (100- to 400- μ) coated from a hydrocarbon gas in the presence of a helium carrier. The hydrocarbon-helium mixture flowed through the furnace at a velocity sufficiently high to fluidize the particles at an absolute pressure of about 1 atm. The partial pressure of the hydrocarbon was varied by varying the hydrocarbon to helium ratio.

Furnace temperatures at 1400°C were measured by placing a Pt versus Pt + Rd thermocouple in contact with the outside of the reaction tube during the preparation of samples. For the coatings prepared above 1400°C, the furnace was brought to equilibrium, and the temperature was determined optically with the helium flowing, prior to insertion of the particles. The particles were then inserted and the deposition commenced. During the coating process, no temperature readings could be determined, because of dense fumes generated in the reaction tube. The furnace was controlled during these experiments by maintaining a constant power supply.

Subsequent to coating of the particles, annealing experiments were carried out in helium in a small graphite crucible.

Metallographic Method

Two methods were developed to retard the reaction of ThC_2 , $(\text{Th}, \text{U})\text{C}_2$, and coated carbide particles during polishing, etching, and photographing; namely, the dry-box method, where the entire metallographic process was carried out in a dry-argon atmosphere, or an oil-immersion method, where the samples were polished in air and were immediately covered with a protective coating of oil.

In order to carry out the metallography by the first method, four dry boxes were arranged in series. A schematic diagram of the equipment is shown in Fig. 1. The samples were transferred in an argon-filled vial from a separate dry-argon storage box to an argon-filled bag. The

Fig. 1.--Dry-argon box system for polishing and photographing thorium carbides

samples were mounted in Kold Weld, a dental plastic that sets in less than 5 min. The mounted samples were then transferred to the polishing glove box, where they were rough-polished by grinding, first on 320-grit and then on 600-grit silicon ~~carbon~~^{carbide} paper. The samples were next moved to the second dry box, where they were final-polished on a cloth charged with 1- μ -diameter diamond dust. The entire grinding operation was carried out dry. Finally, the polished surfaces were examined and photographed in a third box within the dry-argon atmosphere. The bare ThC_2 samples were etched by exposing a freshly polished surface to the dry-box atmosphere for 10 to 30 min, etching of the surface occurring by reaction with the 30 to 60 ppm residual water vapor present in the box atmosphere. The photomicrographs shown in Fig. 2 illustrate the results of the etching technique.

The method for polishing samples in air was similar to that used with dry boxes. The same type of paper and cloth polishing apparatus was used, except that during polishing kerosene was used as a lubricant and dust deterrent. After the final grinding pass, the surface of the sample was wiped clean and quickly swabbed with immersion oil to prevent the occurrence of hydrolysis during subsequent examination.

The samples polished in air were examined and photographed through the oil film. The photomicrographs in Fig. 3 illustrate etching in laboratory air through the oil film. Because of its convenience and the relatively good results attained, the oil-immersion technique was used extensively to examine and view the coated particles. Also, the coating imparted some degree of protection to the particle during the mounting and polishing steps.

The dry-box technique is recommended for polishing uncoated thorium carbide particles.

The photomicrographs in Fig. 4, made with polarized light, show the general appearance of pyrolytic carbon deposited on a small spherical particle from methane at 1400°C. For comparison, a photomicrograph of a commercial pyrocarbon body is shown in Fig. 5. These photomicrographs

(a)

(350x)

(b)

(350x)

(c)

(350x)

Fig. 2--(Th, U) C_2 particle: (a) as polished; (b) etched 8 min in dry box (50 ppm H_2O); and (c) etched 10 min in dry box (50 ppm H_2O)

(480x)

(a)

(480x)

(b)

Fig. 3 --(Th, U)C₂ particle etched by moisture in laboratory air through an oil coating: (a) for 5 min; (b) for 20 min

(480x)

(c)

(1000x)

(d)

Fig. 3 (cont.)--(Th,U)C₂ particle etched by moisture in laboratory air through an oil coating: (c) and (d) for 40 min

(200x)

(615x)

Fig. 4--Photomicrographs of pyrocarbon deposited on small particle at 1400°C and at ~160 mm Hg of CH₄

(500×)

Fig. 5 -- Photomicrograph of commercial
pyrocarbon body

illustrate the results of the oil-immersion method when applied to examination of only the coatings of the particles.

Measurements of Coating Thickness

Two methods were used for determining the coating thickness: (1) metallography (by polishing to the equator and making direct measurement), and (2) calculation (from density and mean particle diameter as determined by screen analysis).

In the metallographic method, the coated-carbide sample was mounted and polished using the oil-immersion technique already described. The sample was placed on a metallograph, and the apparent coating thicknesses of the sectioned particles were measured. At least 20 to 30 particles from each batch were measured; the average of these values was taken as the coating thickness for a batch. Only particles having a reasonably uniform coating thickness were measured, and any particles that did not appear to have been polished to their equator were rejected.

The second method involved the calculation of the average thickness of the coating from the densities and mean particle diameters of the coated and of the uncoated carbides. The coating thickness, T , is related to these parameters by either Eq. (1) or Eq. (2):

$$T = r \left[\left(\frac{D_c - D_g}{D_p - D_g} \right)^{1/3} - 1 \right], \quad (1)$$

or

$$T = R \left[1 - \left(\frac{D_p - D_g}{D_c - D_g} \right)^{1/3} \right], \quad (2)$$

where T = coating thickness, μ ;

r = average radius of uncoated carbide particles, μ ;

R = average radius of coated particles, μ ;

D_c = bulk density of uncoated carbide particles, g/cm^3 ;

664 012

D_p = bulk density of coated particles, g/cm^3 ;

D_g = bulk density of pyrocarbon coating, g/cm^3 .

These equations were obtained from a mass balance for a coated particle, assuming spheres. Either equation could be used, depending on whether the average diameter of the uncoated or of the coated particles was measured.

The particle size distribution of uncoated or coated particles was determined by screen analysis, and the average particle diameter was determined from a cumulative-size distribution plot (see Fig. 6). The bulk densities of the uncoated or coated particles were determined as described in the ~~previous~~ ^{next} section.

The results of the techniques employed on duplicate samples may be compared from the curves in Fig. 7. The agreement between Battelle Memorial Institute (BMI) and General Atomic results by metallographic techniques is only fair. The difficulty of measuring coating thickness on irregular-shaped particles may be the cause of this discrepancy. The calculated values from density measurements also do not agree well with the metallographic data.

Coating-thickness determinations by metallography at General Atomic and Battelle Memorial Institute and by calculation from density measurements at General Atomic on a variety of samples are compared in Table I. Fair agreement is shown, and the agreement improves among the samples containing spherical carbide particles. The accuracy in measuring coating thickness on spherical base particles was estimated to be $\pm 5 \mu$. The majority of the spherical particles observed had coatings very near the average thickness, and it is unlikely that the true average value would fall outside the maximum or minimum value for any measured particle.

Evaluation of Particles

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Bulk Density Determinations of Irregular Particles. Bulk densities of uncoated and coated carbide particles were measured by mercury dis-

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Fig. 6 -- Typical average particle diameter determined by screen analysis

Fig. 7--Comparison of coating thickness determination methods on duplicate samples from the same coating batch; particle size 105 to 250 μ

Table I.

COMPARISON OF COATING-THICKNESS DETERMINATIONS
MADE BY GENERAL ATOMIC AND BY BATTELLE
MEMORIAL INSTITUTE

Carbide Particles*		Coating Thickness (μ)		
Fabrication Method	Shape	Metallographic		Calculated at General Atomic
		General Atomic	Battelle Memorial Institute	
Sintered	Irregular	{ 4	--	2
		{ 12	10	6
		{ 16	12	14
		{ 32	30	41
		{ 37	22	26
		{ 40	--	29
		{ 42	33	48
		{ 46	--	41
		{ 57	--	90
		{ 67	55	114
		{ --	30	38
Melted	Spherical	{ 29	--	36
		{ 59	--	60
		{ 59	--	65

*The particle surfaces in all cases were not smooth.

664 016

placement, using a penetrometer assembly from a standard mercury porosimeter (see Fig. 8). The penetrometer consisted of a "bowl" connected to the top of a capillary stem. A cap was sealed to the top of the bowl and the assembly was placed within a filling apparatus consisting of a tapered bulb with connections to a vacuum pump, to air, and to a mercury reservoir.

The sample volume was selected to allow the capillary to protrude above the sample level, thus preventing the loss of material. The filling apparatus was evacuated, and mercury was introduced into the apparatus to a level that covered the bottom of the penetrometer stem. Air was admitted slowly to attain atmospheric pressure, which forced the mercury up the capillary and around the sample. The filled stem was then withdrawn and weighed. This operation was also carried out without the sample. By this method, the sample volume and density could be calculated, assuming that the mercury does not wet the carbide particles, from the relationship:

$$D_s = \frac{W_s D}{W_s + W_c - W_f}, \quad (3)$$

where D_s = density of sample, g/cm^3 ;

W_s = weight of sample, g;

W_c = calibration weight of penetrometer filled with mercury, without sample, g;

W_f = final weight of penetrometer (filled with mercury) and sample, g;

D = density of mercury, g/cm^3 , at temperature of test.

The density of copper shot determined with the above apparatus and procedure compared favorably with the handbook value (determined = 8.84 g/cm^3 ; handbook value = 8.92 g/cm^3).

Crushing-strength Tests on Coated Particles. In order to evaluate the coated particles for subsequent use in fabricating compacts, it was necessary to determine the relative strength of the coatings. Comparative crushing resistance among various batches of coated carbides was determined by subjecting the particles to a compressive load (1) individually and (2) as a

Fig. 8--Schematic diagram of apparatus for measuring bulk densities of bare and coated (Th, U)C₂ particles

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column in an aluminum die. In the first method, single particles were loaded until fracture occurred; in the second, the integrity of the column of particles after being subjected to load was evaluated by conducting weight-gain experiments. The number of particles that were ruptured during loading was assumed to be directly related to the subsequent weight gain of the column. This test was helpful in screening out coated particles of poor quality.

Only a few crushing tests were made on single particles; results are shown in Table II. The difficulty in applying the load to such a small sample (200- to 500- μ diameter) and the probable variation in the sample particles require that the data be considered qualitative; i. e., there was no apparent correlation between the crushing strength of an individual particle and the thickness of its coating.

The crushing resistance of a column of coated particles was measured by placing a 1-g sample in a die and applying compressive loads of 400, 600, 800, and 1000 lb to the column. The sample was exposed to air for 24 hr after each loading, to allow hydrolysis of any exposed carbide particles whose coatings had fractured. Weight gain from hydrolysis of the $(\text{Th}, \text{U})\text{C}_2$ was observed after each load was applied. This procedure was repeated until several points were obtained.

The weight gain increased in varying amounts, depending on the quality of the coated sample, as additional load was applied. Typical weight-gain to load curves for samples of varying coating thicknesses prepared under similar conditions are shown in Fig. 9. The scatter in the weight-gain data decreased with increasing coating thickness. The slope of each curve was used as a comparative measure of the crushing resistance of the coated sample, small values of the slope indicating good crushing resistance. The curve in Fig. 10 shows the rough correlation between crushing resistance, as determined by the slope of the crushing-test curve, versus coating thickness for carbide particles coated under similar conditions.

Table II
CRUSHING-STRENGTH DATA ON SINGLE PARTICLES

Base-carbide Fabrication Method	Coating Thickness* (μ)	Number of Particles Tested	Load to Fracture (g/particle)	Uncertainty in Data (%)
Sintered	42	4	725	± 13
Melted	30	6	1340	± 8
Melted	30	5	658	± 40
Melted	60	5	1140	± 14
Melted	60	5	935	± 18

* Measured by metallography, on a sample taken from an entire coating batch.

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Fig. 9.--Typical weight gain versus load curves for coated carbides

Fig. 10--Correlation between crushing resistance and coating thickness

Structural Studies of Coating

X-ray diffraction measurements were obtained on samples of crushed carbon. The coated particles were crushed in a mortar, and the $(\text{Th, U})\text{C}_2$ was allowed to hydrolyze in laboratory air. Carbon fragments were soaked in hot HNO_3 to leach out the carbide. The carbon powder was encapsulated in a 0.2-mm glass capillary, and Debye-Scherrer powder patterns were obtained. A densitometer was used to scan the exposed films. The patterns from a commercial moderator graphite (AGOT) and from a commercial pyrolytic carbon, for comparison, are shown in Fig. 11.

The carbon coatings deposited at 1400°C at a pressure of about 160 mm Hg produce diffuse bands in the region of the (00ℓ) and (hk) Bragg reflections and no general (hkl) reflections. The structure of the commercial pyrolytic carbon is quite similar to that of the carbon deposited at 1400°C , except that the bands are much sharper and are shifted to higher Bragg angles. In contrast, the AGOT graphite pattern shows that the (hk) bands are replaced by the (101) and (100) lines, the three-dimensional (112) peak is present, and the (00ℓ) lines are much sharper and are shifted to higher Bragg angles. The higher angle reflection, (006) , was sufficiently intense to be recorded on the film.

The data in Fig. 11 indicate the structure of the deposited carbons is two-dimensional, consisting of layers stacked in parallel groups but not otherwise mutually oriented; i. e., the layers show a completely random orientation with respect to rotation about the layer norm. The three-dimensional patterns observed for AGOT graphite were not observed for the deposited pyrocarbon.

Increasing the temperature of deposition above 1400°C and decreasing the CH_4 pressure to about 100 mm Hg changes the appearance of the deposited carbon. Photomicrographs of carbon deposited at 1400° , 1800° , 2000° , 2200° , and 2400°C are shown in Fig. 12. The 1400°C picture was repeated for comparative purposes. X-ray diffraction traces of these

Fig. 11--X-ray diffraction patterns of AGOT, commercial pyrolytic carbon, and experimental pyrocarbon

(615x)

(a)

(1000x)

(b)

Fig. 12--Macrostructure of carbon deposited at various temperatures from methane: (a) at 1400°C; (b) at 1800°C

(470×)

(c)

(640×)

(d)

Fig. 12 (cont.)--Macrostructure of carbon deposited at various temperatures from methane: (c) at 2000°C; (d) at 2200°C

(530×)

(e)

Fig. 12 (cont.)--Macrostructure of carbon deposited at various temperatures from methane: (e) at 2400°C

samples are shown in Fig. 13. The traces for commercial pyrocarbon and for the 1400°C specimen are included for comparative purposes.

The macrostructure of the coatings appears to change abruptly from a layered structure when deposited at 1400°C to a mottled or grainy structure when deposited at 1800° and 2000°C . Further increasing of the temperature of deposition to 2200° and 2400°C also resulted in the layered structure. The significance of the change in the appearance of the structure is not clear. Examination of the X-ray traces in Fig. 13 indicates only that the two-dimensional structure becomes better developed as the temperature of deposition is increased. Because of the diffuse character of the (00ℓ) bands, the position of their maxima and thus the layer spacing (C_0) cannot be calculated with any degree of certainty. It is safe to say, however, that the layer spacing (C_0) for all carbon structures shown in Fig. 13, including the commercial pyrolytic carbon, is greater than $6.80 \text{ \AA}$. A considerable amount of incoherent scattering and scattering by noncrystalline carbon is present.

The X-ray diffraction traces of powder samples of crushed coatings annealed for 6 hr at 2400°C in helium are presented in Fig. 14. The annealing of these coatings at 2400°C has a marked effect on their structures. The effects observed directly on the X-ray diffraction patterns (compare Figs. 13 and 14) are:

1. The (10) two-dimensional band splits into the two- and three-dimensional peaks--the (100) and the (101) .
2. The three-dimensional (112) peak appears.
3. The (00ℓ) lines sharpen and move toward higher Bragg angles.

The degree to which these changes occur depends on the temperature at which the carbon coatings were originally deposited.

The modulation of the (10) band and the appearance of the (112) reflection is apparent for the coatings deposited at 1400° , 1800° , 2200° , and 2400°C (Fig. 14), whereas the coating deposited at 2000°C retained its two-dimensional character. This indicates that annealing the coatings

Fig. 13--X-ray traces of carbon structure deposited in the temperature range from 1400° to 2400° C

Fig. 14--X-ray traces of carbon structure deposited in the temperature range from 1400° to 2400° C and annealed for 6 hr at 2400° C

664 030

deposited at 1400° , 1800° , 2200° , and 2400° C transforms at least a portion of the carbon to three-dimensional graphite structure. The transformation is not complete for graphite deposited at 2000° C. Further, the pattern for the carbon coatings deposited at 1400° C and subsequently annealed at 2400° C indicates a slightly better graphitic structure than that formed from material deposited at 2200° or 2400° C. (See Figs. 13 and 14 and compare the relative perfection of the (101) and (112) reflections in the annealed 1400° C sample with the AGOT sample.)

Lattice parameters A_0 (unit cell dimension in the A direction) and C_0 were calculated directly from line spacings on the films, using the (110) and (004) reflections, respectively. The results are shown in Fig. 15. The layer spacings are approaching those of a highly graphitic structure for the annealed coatings deposited at 1400° , 2200° , or 2400° C, whereas the layer spacing for annealed coatings deposited at 1800° or 2000° C are considerably larger.

To supplement the X-ray data, additional samples deposited at 1400° or 1800° C were annealed at 2000° , 2200° , and 2400° C and examined metallographically. Photomicrographs of the results are shown in Figs. 16 and 17. The surface appearance of the specimen deposited at 1400° C apparently has not undergone a major change. The layered or shell-like structure still persists, although for the annealed specimens these layers appear to be thicker.

Although the X-ray trace of the 1800° C sample annealed at 2400° C for 6 hr was quite similar in appearance to that of the 2200° C sample (see Fig. 14), the (002) and (004) lines occur at lower Bragg angles. Note also that the macrostructure of the 1800° C coating (see Fig. 17) has not undergone a major change; i. e., the mottled, or grainy, structure persists. In the annealed samples, the grains appear larger and contain so-called "growth cones," with the apex of the cone oriented toward the substrate.

Fig. 15--Lattice parameters of carbon structures deposited in the temperature range from 1400° to 2400°C and annealed 6 hr at 2400°C

(615x)

(a)

(470x)

(b)

Fig. 16--Macrostructure of carbon deposited
(a) at 1400°C at 160 mm Hg and annealed
(b) at 2000°C for 2 hr

(c)

(d)

Fig. 16 (cont.)--Macrostructure of carbon deposited at 1400°C at 160 mm Hg and annealed (c) at 2200°C for 1 hr and (d) at 2400°C for 0.5 hr

(1000×)

(a)

(640×)

(b)

Fig. 17-- Macrostructure of carbon deposited
 (a) at 1800°C at 100 mm Hg and annealed
 (b) at 2000°C for 2 hr

(530×)

(c)

(490×)

(d)

Fig. 17 (cont.)--Macrostructure of carbon deposited at 1800°C at 100 mm Hg and annealed (c) at 2200°C for 1 hr and (d) at 2400°C for 0.5 hr

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III. DISCUSSION OF RESULTS AND CONCLUSIONS

The macrostructures of the pyrocarbons presented here are quite different from those published by Brown, Clark, and Eastabrook,³ which were deposited on a commercial-graphite substrate. Their photomicrographs show a block-like or perhaps a conical appearance in all specimens deposited at 1400°, 1600°, 1700°, 1900°, and 2100°C, with the 2100°C deposits having the appearance of a fibrous structure similar to that shown in Fig. 8. Their lower-temperature deposits appear to have highly conical grains. The macrostructure of the samples in our present studies indicate that the 1400°C sample has large macrolayers that follow the circumference of the substrate. As the deposition temperature is increased to the range of 1800° to 2000°C, the macrostructure changes to relatively fine grain or conical, with an appearance similar to Brown's photomicrographs.³ These grains are diminished but not completely obliterated, and the ringed macrostructure reappears as the deposition temperature is raised to 2200° to 2400°C. X-ray data on similar deposits in the temperature range of 1600° to 2100°C by Brown and Watt² in an earlier publication are similar to that observed for the deposits in our study. Their samples of lowest (1600°C) and highest (2100°C) deposition temperature when annealed at 2500° or 2800°C showed some degree of transformation to graphitic structure; the X-ray lines of all deposits become sharper except for the sample deposited at 1900°C, which showed no (hkl) reflections even when annealed at 2800°C. This behavior corresponds to that of our 1800° and 2000°C samples after annealing at 2400°C.

Meers⁵ and Stover⁶ annealed carbon deposits at temperatures above 2500°C. Meers indicates that carbons deposited in the range of 1800° to 2000°C have relatively low densities; annealing these samples at 2800°C failed to increase their densities to values that compared with annealed samples deposited at 1600°C and at 2100° to 2200°C. Stover found essentially the same phenomenon, indicating that a sample deposited at

1750°C transforms to graphite structure at lower annealing temperatures than one deposited at 2100°C.

In general, our data (although the samples appear quite different in macrostructure) agree with the work of the above investigators. The annealing studies all indicate that carbon deposited in an intermediate-temperature range is deposited in such a manner that it resists graphitization. There is some disagreement on the exact temperature range that produces nongraphitizable carbon. The observation of temperature-dependence on graphitizability apparently holds even though the various experiments were carried out under widely different conditions of pressure, hydrocarbon species, substrate material, geometry, and size.

Our results indicate that material deposited at 1400°C is essentially a graphitizable carbon, while that deposited in the intermediate range of 1800° to 2100°C is essentially nongraphitizable. According to Franklin's⁷ classification, carbons prepared by pyrolysis of carbonaceous materials of high hydrogen content give particles with small crystallites that are somewhat ordered and positioned in such a manner that further structural development may be accomplished by further heating. This was particularly true of materials that pyrolyzed at least partially in the liquid state--such as coal tar or petroleum pitch. Also, carbons prepared from materials of high oxygen content or those that pyrolyze in the solid state produce essentially crosslinked structures which resist further graphite formation. Akamatu and Kuroda⁸ in their studies of petroleum coke particles and carbon blacks by electron diffraction patterns show that the crystallites of petroleum coke are in fact aligned and that this alignment is necessary for ease of graphitization during subsequent heating. If Franklin's classification of carbons is applied to the data on carbon structures formed by deposition, one would expect the carbon structures prepared at a temperature near 1400°C to be better developed than those prepared at 1800° to 2000°C; however, from analysis of X-ray diffraction patterns, this apparently is not the case.

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FOOTNOTES

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⁴A commercial name given to deposited carbon bodies by some commercial producers.

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664 040

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664 041