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Nondestructive Testing of Graphite at the
Los Alamos Scientific Laboratory*

80428

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ABSTRACT

The Los Alamos Scientific Laboratory has investigated and used radiography, ultrasonics, sonics, eddy current, radiation gaging, and beta-backscatter gaging for the inspection of graphite and its protective coatings. Some applications and limitations of these techniques are reviewed. The use of ultrasonics, sonics, eddy current, and radiation gaging to determine physical characteristics, aside from the normal flaw detection shows promise in the inspection of graphite. Electron microscopy has proved to be a valuable tool for the evaluation of the findings of nondestructive testing.

KEY WORDS

Nondestructive Testing, Graphite, Ultrasonics, Sonics, Eddy Current, Radiation Gaging, Beta Backscatter, Electron Microscopy, Physical Properties, Moduli, Sound Velocity, Grain Orientation, Protective Coatings

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INTRODUCTION

When the Los Alamos Scientific Laboratory started to use graphite in its experimental nuclear reactor programs, the need for more knowledge of the material was evident. One natural need was how to inspect it. Nondestructive testing is, of course, one of the inspections needed for any structural material and for the components made of it.

The safety and economic aspects inherent in nuclear reactor technology justify a more thorough investigation of all aspects of testing than in other potential fields.

Therefore, an extensive investigation of possible means of nondestructive testing of graphite has been and is being conducted. Most of the methods used on the more conventional structural materials have been investigated and various innovations have been devised. The search has not been confined to the normal flaw-detection approach but has been extended to a search for methods of nondestructively determining physical constants which might have bearing on the utility and suitability of the part. The methods investigated are covered topically along with the findings for each.

METHODS

Radiography

Radiography was one of the earliest methods of nondestructive testing to be investigated. The findings were reported thoroughly in a previous article.⁽¹⁾ Radiography is currently used for the inspection of a number of reactor components fabricated by, and for, the Laboratory and is used for flaw detection. The establishment of meaningful specifications for these inspections is a continuing process based on the performance of the parts.

Ultrasonics and Sonics

Graphite was found very early in the investigation to be a poor conductor of ultrasonic energy. Only relatively thin sections can be inspected for discontinuities with the normal pulse-echo techniques. A large variation in the degree of attenuation among the various types of graphite which have been tested is illustrated by the data in Table 1. These data were obtained by using a transmitted wave (two transducer) technique at 2.25 MHz. The relative attenuation factor is that amount of gain necessary to bring the received pulse height to that of Type 331 graphite.

Sound transmission measurements have not been used much for flaw detection, but they are useful in establishing sound velocity in the specimens. Sound velocity is of interest because of its connection with other physical attributes of the material.

Various moduli of a material can be determined from its sound velocity and density. The relationships between sound velocity and physical properties are given in a number of references, for example, the Nondestructive Testing Handbook.⁽²⁾ The density can be determined nondestructively with radiation gages which will be discussed later. Many have investigated methods of determining sound velocity.^(2,3,4)

The through-transmission, elapsed-time method of determining sound velocity is applicable to most graphites. A Sperry 721 Reflectoscope shown in Fig. 1 is generally used. The thickness of the sample is limited to about two inches due to the attenuation. Greater thicknesses could be measured with a more powerful pulser. In most cases alcohol is used as a couplant since it leaves little contamination.

The resonant frequency technique has also been used to measure sound velocity--in this case, the extensional velocity. A small, light-weight disk of magnetic material is attached to one end of the sample. The disk is excited by an electro-magnet driven by a variable frequency generator. A ceramic

pickup attached to the opposite end of the specimen generates a signal voltage proportional to the energy it receives. The specimen is supported at its first and third quarter-length points as shown in Fig. 2. To insure that the lowest resonant frequency has been found, the specimen is momentarily clamped in the center to severely damp the fundamental frequency. The specimen must have a length at least ten times its smallest cross-sectional dimension for this technique.

A similar setup can be used to determine the torsional velocity. The driving disk and the pickup are mounted on the circumference rather than at the ends of the specimen. Velocities have been determined in graphite samples up to five-feet long in this manner. We have been able to sort many graphite components into different manufacturing batches using these methods. In some instances, correlation indicated that the sound velocity had a bearing on the serviceability of the part. Fig. 3 is a graph showing cracking as a function of sound velocity in a group of components from a reactor test.

Although conventional electronic pulsing circuits are now available to produce considerably higher power than the reflectoscope, we are investigating sources with even more power to penetrate reasonable sections of low-density graphite. A block diagram of the first attempt is shown in Fig. 4. The spark gap produces a very strong shock front which can be received by a standard ultrasonic transducer. The 1 μ f capacitor bank in the block diagram was charged to about 3000 V and discharged through a thyatron.

Although this apparatus produced a shock wave capable of penetrating 3.5 in. of graphite having a density of 1.01 gm/cc, the dispersion of energy through the couplant, and echoes from the walls of the container precluded use because of the reduced signal-to-noise ratio. We think the device can be refined by collimating and focusing the source and coating the container walls with sound-attenuating materials.

Eddy Current

Primarily, eddy current inspection has been used for near-surface flaw detection in graphite. However, conductivity measurements have contributed other useful information. Such measurements have been used to sort graphites either as a function of composition or treatment, often a useful step in the fabricating sequence. They have also been used to determine grain orientation, as will be discussed later.

Our primary tool for eddy current inspection is a system which we assembled and named the "Universal Eddy Current Tester." A block diagram of this unit is shown in Fig. 5. The design is such that a coil may be driven at any frequency from 20 KHz to 1.5 MHz. The search coil signal can be balanced to a null in a chosen set of surroundings with the bridge circuit. The imbalance in the bridge, caused by changes in the coil impedance due to its surroundings, is amplified and displayed both on an oscilloscope and on a digital voltmeter.

The system can drive coils with a wide variety of designs. The smallest coil employed for routine inspection is 0.082 in. in diameter. This probe for inspecting graphite with a small diameter hole is shown in Fig. 6. With this coil and the system, we have consistently detected discontinuities equivalent to a 0.035-in.-diameter hole extending 0.025 in. radially from the bore surface.

The procedure for utilizing the system is as follows:

1. The bridge is adjusted to zero with the coil in air or with a calibration sample.
2. The amplifier gain is set at a selected voltage level with the coil in a standard containing a discontinuity of a known size.

3. Routine inspection proceeds on the premise that discontinuities producing voltages equal to or greater than that of the standard are worthy of consideration.

The tester has also been used to detect the presence of protective coatings on graphite. The bridge is nulled with the coil on bare graphite. The amplifiers are then adjusted to a five-volt level on a specimen with the greatest expected coating thickness. Components producing signals below 2 V are rejected for insufficient coating.

Although the frequency range of the tester, 20 KHz to 1.5 MHz, has been adequate for most graphite inspections, we are now in the process of increasing the upper limit to 180 MHz. We hope that this extended range will prove useful in the inspection of very thin protective coatings. At the higher frequencies, it should be possible to inspect the coatings with negligible energy penetration into the graphite substrate.

The Universal Eddy Current Tester is also used to make conductivity measurements. In this application, it is more flexible than a Magnaflux Magnatest FM-102. The range of conductivities which can be measured with this commercial equipment extends to a minimum of only 0.8 per cent of the International Annealed Copper Standard, a conductivity greater than that of most of the types of graphite used by the Los Alamos Scientific Laboratory. Furthermore, the FM-102 is relatively limited as to the type of probe which can be used with it.

The Universal Eddy Current Tester is calibrated with samples of known conductivity for making conductivity measurements according to the following procedure:

1. The bridge is adjusted to some convenient level.
2. Standards of known conductivity are placed beneath the probe and the output voltages of the tester are noted.
3. The output voltages are plotted as functions of the conductivities of the samples to form a calibration curve.
4. The output voltages from unknown samples are converted to conductivities through the calibration curve.

A typical calibration curve is shown in Fig. 7. Conductivities as low as 0.003 per cent IACS can be measured with the equipment. The low conductivity standards are fabricated from graphite carefully inspected to insure uniformity; their conductivities are determined by standard laboratory methods.

To detect grain orientation in graphite by conductivity measurements, a rectangular coil 0.125-in. wide and 0.25-in. long was constructed. The conductivity of a sample parallel to the longer dimension of the coil contributes the major portion of the signal from the coil. The change of conductivity as the coil orientation is changed has been found to be a function of grain orientation in graphite.

Various types of graphite exhibit different degrees of grain orientation. In some types, very little grain orientation is observed, while others show grain orientation in only one plane and some show it in all three planes. Some typical values of measured conductivity with the directional eddy current probe are shown in Table 2. We think that a change greater than 0.01 per cent IACS is indicative of significant grain orientation.

Radiation Gaging

One use of radiation gages is the determination of densities of samples. For this application, it has been found that high energy radiation (about 1 MeV) is desirable because the absorption coefficients are more nearly a function of density at that energy level. The Los Alamos Scientific Laboratory pioneered in the development of this type of gage and has published several articles on it.^(5,6,7)

The densities of graphite samples up to 4-in. thick have been determined with such a transmission gage using the gamma radiation from a cobalt-60 source. As in previously reported work, the gage was empirically calibrated using graphite standards, the densities of which had been measured by standard means.

The gage used for this work consists of a one curie cobalt-60 source mounted in a uranium shield, primary and secondary collimators designed to restrict the radiation beam to a diameter of approximately 3/16 in., a stage for scanning the graphite sample between the two collimators, a gamma detector utilizing a Na I(Tl) scintillating crystal and photomultiplier tube, and a dc signal amplification and recording system. Basically, the relative transmission of gamma rays through the sample is the measured variable in the equation:

$$T = e^{-\mu \rho t},$$

where

T is the per cent transmission (ratio of transmitted gamma intensity to initial gamma intensity)

μ is the mass absorption coefficient

ρ is the material density

t is the specimen thickness.

For a given material, μ is constant and t can be measured. Therefore ρ can be related to the measured T through the calibrated standards. These standards are selected from graphites of different densities machined to size and accurately weighed. The overall error in the density measurements of such standards is on the order of ± 0.25 per cent.

Since a two-dimensional scanner is normally used to position specimens in the beam of radiation, it is possible to determine density variations within the sample. A wide variety of scanning patterns can be selected to cover samples with simple geometrical shapes. Figure 8 shows the gage described, and named DXT Gage since it measures density times thickness. Figure 9 is a close-up of the unit showing the detector collimator. Figure 10 is a reproduction of a typical scanning trace from the unit.

Density measurements have been made on a number of graphite samples in the research program of the Laboratory. The total error in the instrumentation and the calibration standards has been observed to be about ± 0.5 per cent.

Beta Backscatter

As previously mentioned, much of the graphite used in the Los Alamos Scientific Laboratory's programs is protected from corrosive atmospheres by suitable coatings. The nondestructive testing of these coatings must be as exacting as the inspection of the graphite. Beta particle scattering gages have been used extensively for the determination of the thicknesses of these coatings. Although the principle has been covered in the literature, (8,9,10) some of our engineering accomplishments to adapt the method to particular configurations are worthy of recording along with the improvements in accuracy achieved.

When materials are bombarded by beta particles, a portion of the particles are reflected or scattered back. The amount of reflection or scattering is a function of the atomic number of the materials upon which the beam impinges (directly proportioned to Z^2 and to density). The application of this method to measurement of coating thickness is dependent on the existence of an appreciable difference in the atomic numbers of the coating material and the graphite, a condition easily met by the materials normally used by the Laboratory.

Most of the work has been done so far using strontium-yttrium-90 beta sources and scintillating detectors coupled to photomultiplier tubes. These gages have been made to inspect the coatings on the bores in small diameter holes as well as to inspect coatings on easily accessible surfaces. A cross-sectional schematic diagram of a beta probe for inspecting easily accessible surfaces is shown in Fig. 11. Figure 12 is a similar view of a small diameter bore probe. A quartz light pipe is used to couple the scintillator to the photomultiplier in these probes. The scintillator is shielded against direct radiation from the source by a 0.015-in. gold foil so that only the scattered radiation can contribute to the signal.

In the routine inspection of coatings, it has been found that accuracies of from ± 3 to ± 10 per cent can be achieved depending on the material and conditions. Again, an empirical calibration of the instrument is made for each coating and substrate. This is accomplished in two ways; first, by the introduction of foils of the coating material, and second, by metallographic measurement of coatings measured. A typical calibration chart is shown in Fig. 13.

Electron Microscopy

The data obtained from nondestructive tests are frequently not completely understandable, particularly in cases where new

materials are involved. We have frequently resorted to various destructive tests to evaluate the findings of nondestructive tests. Electron microscopy has been found to be a very useful tool in these efforts. The measurements from such tests as eddy current and ultrasonics can be explained by the fine structure of the material revealed by the electron microscope. Figure 14 is such a picture of a sample of nuclear graphite of the type normally used by the Laboratory; that is, an extruded graphite made of graphite flour with a binder. The flake structure in the sample is the flour and the granular regions are the graphitized binder. The orientation of the grains can easily be detected. Figure 15, an electron micrograph of a carbide coating on a pyrocarbon matrix, illustrates the laminar nature of pyrocarbon deposits, explaining the vastly different nature of nondestructive test signals being returned as compared to those from conventional graphite. Electron microscopy is indispensable in the evaluation of our protective coating results. Figures 16 and 17 are micrographs showing adherent and nonadherent niobium carbide coatings on graphite, respectively. Note the intrusion of the adherent coat into the open pores in the graphite matrix.

CONCLUSION

This paper describes some of the work which has been done in developing and applying nondestructive testing methods to graphite at the Los Alamos Scientific Laboratory. Radiography, ultrasonics, sonics, eddy current, radiation gaging, and beta-backscattering methods are described. Further development of most of these methods continues. An area which must be explored more thoroughly is the appraisal of the findings from these methods. This is perhaps the most difficult portion of the general field of nondestructive testing. In most instances, it involves a tremendous amount of very tedious assimilation of data.

The nondestructive determination of the mechanical properties of graphite is an area of much promise which is also being pursued by other workers.⁽⁴⁾ It appears to have more applicability to graphite than to most of the more conventional materials of construction.

Much work remains in the development of nondestructive tests for graphite, but meaningful tests are now available.

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Table 1 - - Relative Ultrasonic Attenuation Factors of Various Graphite Types

Graphite Type	Relative Attenuation Factor ¹
331	1
GG	0.001
AUC	4.5
H4LM	49
CS-312	80
Reactor Grade	120
ATL	210
SX-5	310
AGOT	370

¹ These values are approximate.

Table 2 - - Typical Conductivity Measurements with a Rectangular
Eddy Current Probe to Determine Grain Orientation ¹

Graphite Type	Apparent Conductivity		
	XY Plane	% IACS XZ Plane	YZ Plane
E24	0.047	0.045	0.051
EP192	0.080	0.085	0.084
SX-4	0.225	0.225	0.170
P03	0.110	0.115	0.142
SX-5	0.152	0.150	0.152

¹ Probe dimensions were 0.125 x 0.25 in.

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FOOTNOTES

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- Fig. 1 - - Model 721 Reflectoscope setup for determining longitudinal sound velocity.
- Fig. 2 - - Block diagram of resonance test.
- Fig. 3 - - Bar charts sound velocity vs cracking of modules.
- Fig. 4 - - Block diagram of high power wave generation experiment.
- Fig. 5 - - Block diagram of the Universal Eddy Current Tester.
- Fig. 6 - - Small diameter eddy current probe.
- Fig. 7 - - Calibration curve for conductivity measurements, Universal Eddy Current Unit.
- Fig. 8 - - Radiation gage for measuring graphite density.
- Fig. 9 - - Close-Up of radiation gage showing detector collimator.
- Fig. 10 - - Typical scanning trace from radiation gaging of a graphite cylinder.
- Fig. 11 - - Schematic beta-backscatter probe for accessible surfaces.
- Fig. 12 - - Schematic beta-backscatter probe for small diameter bores.
- Fig. 13 - - Calibration chart for beta-backscatter coating thickness measurement.
- Fig. 14 - - Electron micrograph of extruded graphite (X6000).

Fig. 15 - - Electron micrograph of pyrocarbon deposit (X6000).

Fig. 16 - - Electron micrograph of adherent coating on
graphite (X6000).

Fig. 17 - - Electron micrograph of nonadherent coating on
graphite (X6000).

Fig. 1

BLOCK DIAGRAM OF RESONANCE TEST

MOTION OF SAMPLE - FUNDAMENTAL FREQUENCY

FIG. 2

Fig. 3

BLOCK DIAGRAM OF HIGH POWER WAVE GENERATION
EXPERIMENT

FIG. 4

BLOCK DIAGRAM OF THE "UNIVERSAL EDDY CURRENT TESTER"

FIG. 5

Fig. 6

Fig. 7 - - Calibration Curve
for Conductivity Measurements
Universal Eddy Current Unit

Fig. 8

Fig. 9

DENSITY TIMES THICKNESS SCAN
GRAPHITE SAMPLE NO. 10Z-1

Fig. 10

Fig. 11

(DIMENSIONS
IN INCHES)

PROBE ASSEMBLY

Fig. 12

Fig. 13

Fig. 14

Fig 15

2 μ

Fig. 16

2μ

Fig 17