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NONDESTRUCTIVE EVALUATION TECHNIQUES FOR HIGH-TEMPERATURE CERAMIC COMPONENTS

Quarterly Report

October—December 1977

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ARGONNE NATIONAL LABORATORY, ARGONNE, ILLINOIS

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LIST OF FIGURES

NONDESTRUCTIVE EVALUATION TECHNIQUES
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D. S. Kupperman, A. Sather, N. P. Lapinski, and G. M. Dragel

I. INTRODUCTION

The overall objective of this program is to assess and develop nondestructive evaluation procedures for high-temperature ceramics. We are currently evaluating ceramic heat-exchanger tubing. Ceramic heat exchangers would be useful, for example, in coal-fired Brayton conversion or waste heat-recovery systems. The use of ceramic heat exchangers will allow working fluids to reach temperatures up to 1230°C, and, with further materials development, possibly 1650°C. If superalloys were employed, working fluids would be limited to ∿800°C. The use of working fluids at higher temperatures would result in more efficient systems. Furthermore, ceramic components are lighter than metallic ones and are made from less costly and more abundant elements. In addition, ceramic heat exchangers would be more resistant to corrosion. In the current NDE effort, several acoustic, optical, and radiographic techniques are being examined for their effectiveness in testing silicon carbide tubing. Some results employing dye-enhanced radiography are discussed in the present report.

II. DISCUSSION

Several silicon carbide tubes have been sent to Argonne National Laboratory for evaluation. The tubes are from the Garrett AiResearch Manufacturing Division's high-temperature heat-exchanger program. The tubes are nominally 200 mm long, 25 mm in diameter, and have wall thicknesses ranging from 1.5 to 3 mm.

Silicon carbide tubes for the present study have been fabricated by several manufacturers. Two of the tubes are shown in Fig. 1. The companies and type of tube made are as follows:

(a) Carborundum; Super KT and sintered α SiC,
(b) Norton; NC430,
(c) Materials Technology Corporation (MTC); chemical vapor deposit (CVD), and
(d) Deposits and Composites Incorporated (DCI); CVD.

In this quarter, progress has been made in the development of a dye-enhanced radiographic technique for SiC tubing. When this method is used, improvements can be made over the conventional radiographic and dye-penetrant methods currently being employed.

The objective of the dye-enhanced radiographic method is to fill surface defects with a substance (doping agent) that will absorb penetrating radiation more effectively than the base material. Thus, although normal radiographs may not reveal surface defects such as cracks, these flaws may become detectable when dye is present. Dye enhancement has been successfully used with neutron radiography. In this case, gadolinium nitrate is mixed with acetone and a wetting agent to form the penetrant. The dye can reveal cracks even if the neutron beam is not parallel to the crack plane. Thus, a surface crack may be mapped from its shadow. Since gadolinium has a mass absorption coefficient for neutrons three orders of magnitude greater than iron, for example, the technique is particularly useful. Neutron radiography techniques can be used for ceramics; however, the main effort in the present study has been to find a doping agent with sufficient contrast so that conventional x-rays could be used effectively. This seemed feasible because of the nature of silicon carbide (low atomic number and low density).

The absorption of a parallel narrow beam of monochromatic x-rays in a plane-parallel layer of homogeneous isotropic material can be described by the relationship

$$I = I_o \exp(-\mu t), \qquad (1)$$

where I is the emerging intensity, I_o is the incident intensity, μ is the total linear absorption coefficient, and t is the material thickness in cm. The mass absorption coefficient (μ/ρ) is generally used in Eq. (1) because the intensity reduction is determined by the quantity of matter traversed. The quantity (μ/ρ) is essentially independent of the physical state of the material and is additive with respect to the elements that make up the material.

$$\mu/\rho = \sum_i g_i \ (\mu/\rho)_i, \qquad (2)$$

where g_i is the mass fraction contributed by the element i with mass absorption $(\mu/\rho)_i$. Thus,

$$I = I_o \exp(\frac{-\mu}{\rho} \cdot \rho t). \qquad (3)$$

The mass absorption coefficient is dependent upon wavelength. For silver nitrate ($AgNO_3$) at 0.3 Å,

$$\mu/\rho_{(0.3\ \text{Å})} = 10.5\ \text{cm}^2/\text{g}.$$

For silicon carbide,

$$\mu/\rho_{(0.3\ \mathring{A})} \approx 0.5\ cm^2/g.$$

The ratio of mass absorption coefficients is approximately the same even for shorter wavelengths. Thus, the silver nitrate absorbs x-rays 20 times more strongly than silicon carbide. Surface flaws not visible by ordinary x-rays may be observed by a dye-enhanced technique.

In previous radiography work with silicon nitride, many commercially available enhancing agents were tried including hypaque-M 90% and hypaque-Na 50%, cholografin meglunine (iodine 42%, sodium 18.2%), and gastrografin (sodium 10% and iodine 30%). A dye consisting of lead oxide mixed with alcohol was also investigated as an agent.

The most successful doping agent, however, was a silver nitrate solution formed from equal parts, by weight, of silver nitrate and water plus a small amount of Photoflow for a wetting agent. The procedure for generating the dye-enhanced radiographs is as follows:

(a) clean the component in an ultrasonic bath,
(b) place in hot (80-90°C) silver nitrate solution,
(c) remove and x-ray, using conventional techniques, and
(d) clean component in ultrasonic bath.

An 80-kV x-ray machine with a film-to-object distance of 100 cm and Type SR Kodak Film was used.

As an example to demonstrate the enhancement technique, a layer of silver nitrate, nominally 0.05 mm thick, was placed on a 3.8-mm-thick flat plate of hot-pressed silicon nitride. Although the silver nitrate represents an increase in thickness of only 1.3%, the film density increased by almost 10%.

The advantages of the dye-enhanced radiography technique have been demonstrated using a piece of SiC tubing (MTC-CVD) with several inner surface cracks (Fig. 1). In the sample of interest, only one crack could be observed with conventional x-radiography techniques. After treatment with silver nitrate dye, five cracks were clearly revealed by dye-enhanced radiography. After sectioning, only the five cracks could be observed microscopically (~100X). These ID cracks penetrated approximately half way through the wall (~25 mils deep), were tight (<10 μm wide), and several were slanted with respect to the tube axis. The tightness and the skew prevented detection by conventional radiography. Figure 2 shows a cross section of the piece of tube after sectioning and Fig. 3 is a higher magnification of Fig. 2. In Fig. 3, the smaller crack was detected only via the dye-enhanced radiography technique. X-ray fluorescent analysis showed silver close to the crack tip, indicating the dye had penetrated almost to the maximum crack depth. The x-ray spectrum is shown in Fig. 4. A scanning-electron micrograph (SEM) of the entire crack is shown in Fig. 5.

The largest peak (Fig. 4) is the result of the penetration of silver at the crack tip. Other peaks are from silicon and gold. The gold was deposited as part of the procedure for SEM photography. Fluorescent dye-penetrant techniques have also been used on the specimen used for dye-enhanced radiography. With the fluorescent dye, the cracks could be seen. However, under normal dye-penetrant examination, cracks located on the inner surface of the tube would not be detected easily, if at all.

Three additional NDE methods were evaluated during this past quarter: acoustic emission, holographic interferometry, and internal friction. An apparatus has been assembled that will allow the tubes to be pressurized to observe the holographic interferograms and to detect acoustic signals generated by the initiation or propagation of defects in SiC tubes. The preliminary results will be analyzed.

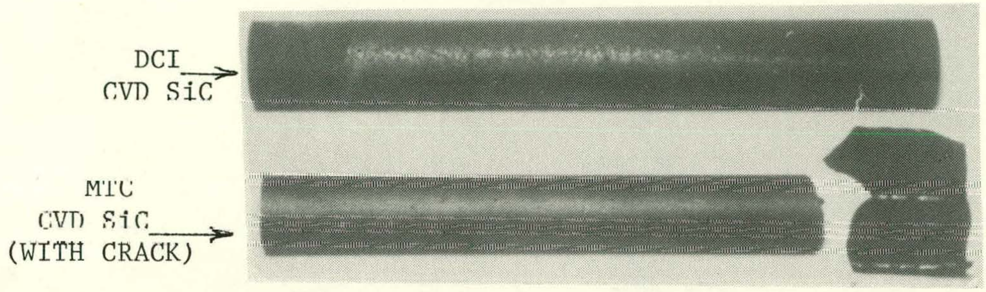


Fig. 1. SiC Tubes Used for Nondestructive Evaluation.

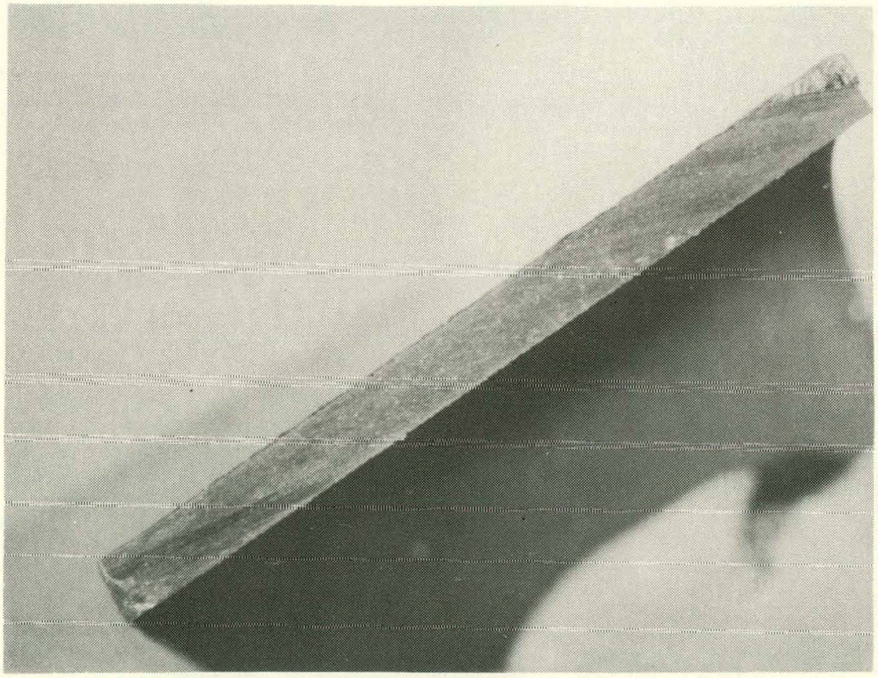


Fig. 2. Section of SiC Tube after Dye-enhanced Radiography Showing Several Cracks. Mag. ∿7.5X.

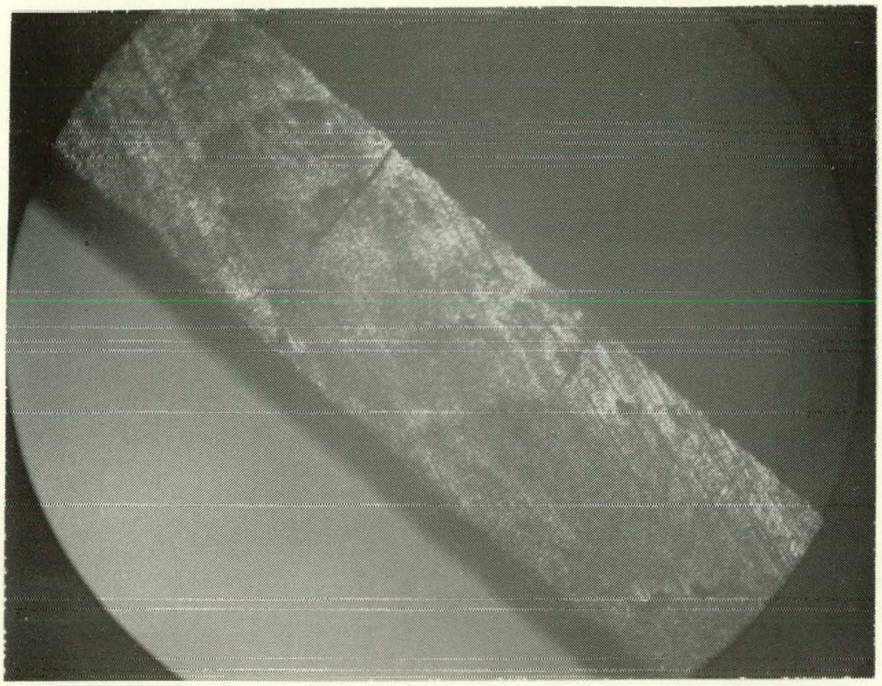
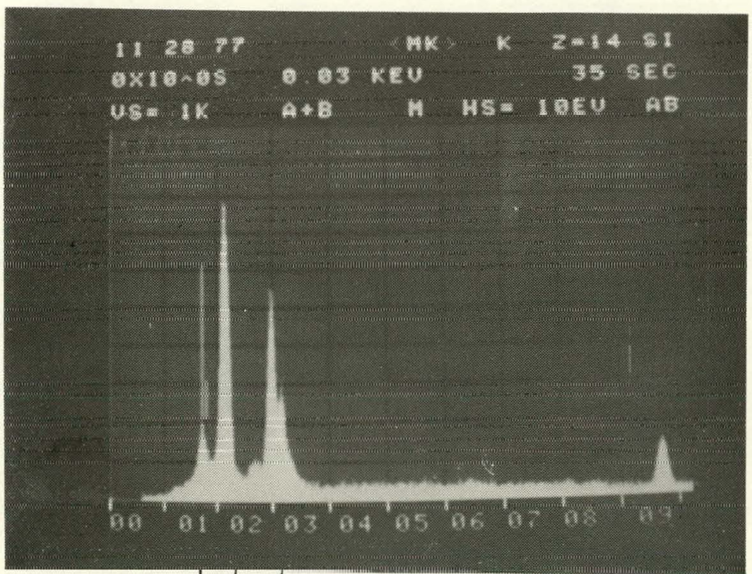


Fig. 3. View of SiC Sample Shown in Fig. 2. The largest crack was detected by conventional and dye-enhanced radiography. The smaller crack was detected only by dye-enhanced radiography. Mag. ∿25X.



Crack 2-1 Si Au Ag

Fig. 4. X-ray Fluorescent Analysis of One of the Crack Tips of the SiC Specimen Shown in Fig. 2. The analysis was carried out after the specimen had been soaking in silver nitrate dye. The largest peak is due to the presence of silver.

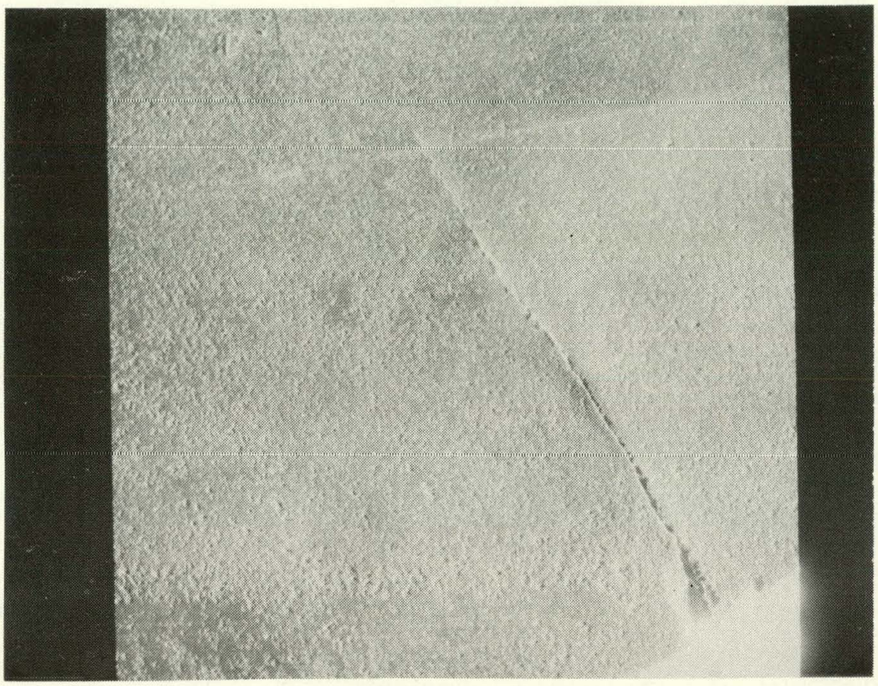


Fig. 5. Electron Micrograph of the Crack Examined by X-ray Fluorescent Analysis. Mag. 100X.