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OXIDATION OF TSX GRAPHITE OVER TEMPERATURE RANGE 450 TO 1200°C

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OXIDATION OF TSX GRAPHITE OVER THE
TEMPERATURE RANGE 450 TO 1200°C

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Although studies have been performed on the oxidation of many grades of graphite over a wide temperature range, the oxidation rate of the much purer nuclear graphites is best-known under conditions in an operating gas-cooled reactor. Thus, much past work has involved helium containing minor amounts of oxygen and water vapor, such as would result from a small leak in the cooling circuit. The accident at Chernobyl during 1986 and the resulting safety analyses of N Reactor prompted interest in obtaining data on the oxidation rate of TSX graphite, the type from which N Reactor was built, under conditions more relevant to those during a postulated accident. These conditions involve large air flows, both in the presence and absence of steam.

Work to date has concentrated on oxidation of TSX graphite by air, using thermogravimetric techniques to follow the progress of the reaction. Oxidation rate measurements have been conducted over a temperature range from 450 to 1200°C. In order to cover this range, two types of thermobalances were used, one a traditional high-sensitivity balance used up to a temperature of about 800°C. The other thermobalance was a conventional top-loading electronic balance, provided with an output permitting an analog signal for recording. This balance could follow the weight loss at high rates observed above 800°C but with less sensitivity. In addition to the traditional kinetics measurements, changes in density resulting from oxidation were measured by a technique in which radiographs of sections cut from oxidized specimens were digitized by a microdensitometer. By using a standard step wedge, it was then possible to relate film density to the density of the oxidized graphite. -

A summary of low-temperature oxidation rate measurements is shown in Figure 1, which includes a least-squares line drawn through the data. Also shown is a line representing a best fit to the data of Dahl,⁽¹⁾ measured using CS graphite, another nuclear grade. As would be expected for these high purity grades, the differences in rates are probably within collective

FIGURE 1. Low-Temperature Data with Least-Squares Fitted Line as Compared to that Resulting from Dahl's Work

sample-to-sample and grade-to-grade differences, even neglecting experimental error. The activation energies measured from the line slopes are essentially identical.

Some experiments were conducted at higher air flow rates, in order that reactant supply not be the limiting factor for oxidation rate. These experiments clearly demonstrated that the reaction rate was lower than predicted by an extrapolation of the line fitted to the low-temperature data. The rate-limiting step was probably diffusion of gas through surface layers, as has been concluded by many previous investigations.

The radiographic technique for measuring density resulted in plots like those shown in Figure 2. Two specimens are represented, one oxidized at a low temperature where the oxidation is homogeneous throughout the graphite and the other at a higher temperature where oxidation restricted to the surface is evident. The first of these exhibits a general decrease in density throughout the sample, and the second shows more attack in the outer regions. These two behaviors are shown in the figure, which also includes a hypothetical trace from an unoxidized specimen at full density.

Work is also in progress on measuring the CO/CO₂ ratio resulting from air oxidation of TSX graphite, for the purpose of supplying basic information to computer codes designed to model accident scenarios. The reaction of graphite with water vapor is also being measured, particularly as regards hydrogen production because of the possible secondary hazards created by this gas. These measurements are in progress at the present, and it is expected that more data will be available at the meeting.

FIGURE 2. Density Profile of Two Oxidized Samples as Calculated from Radiographic Density Measurements

REFERENCES

1. Dahl, R. E. Oxidation of Reactor Graphite Under High Temperature Reactor Conditions," USAEC Report HW-68493, July 1961.