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Memorandum

THE DIFFUSION OF OXYGEN IN NITROGEN IN THE PORES OF GRAPHITE

Preliminary results on the effect of oxidation on diffusivity

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THE DIFFUSION OF OXYGEN IN NITROGEN IN THE PORES OF GRAPHITE:
PRELIMINARY RESULTS ON THE EFFECT OF OXIDATION ON DIFFUSIVITY

by

G. F. Hewitt and E. W. Sharratt

Abstract

Preliminary results are reported from an experimental study of the effect of burnoff on the diffusivity of oxygen in nitrogen within the pores of graphite. It is found that the ratio of effective diffusivity to "free gas" diffusivity changes about four-fold in the range 0-9% total oxidation. The viscous permeability, B_0 , increases in almost the same proportion over the same range.

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A/I

1. Introduction

In a previous report⁽¹⁾ measurements are described on the diffusion of oxygen in nitrogen in the pores of nuclear grade graphite. Variations of the diffusion coefficient with position in a cross section of a block of pile A graphite were studied with the object of providing information relevant to the diffusion control of the oxidation of nuclear grade graphite. It was realised that the diffusion coefficient itself would be affected by the extent to which the graphite had been oxidised and a limited study of this was made; it was shown that the diffusion coefficient probably did not change significantly in the range 0-0.5% total oxidation. These results, however, were rather inconclusive since it was necessary to reduce the diameter of the sample somewhat between successive oxidations.

In the present report, initial results are given from a more extensive study of the effect.

2. Experimental

The samples of graphite used in the present tests were all taken from off-cuts of a block of pile A graphite (Ref. No. 7387), the rest of which was used by the Graphite Oxidation Section for oxidation tests. These off-cuts consisted of a slab of rectangular cross section cut from the base of the block and a short cylinder 6" O.D. x 1.1/8" I.D. cut from the end of the virgin machined cylindrical section used in the oxidation tests. Six radial samples (Rad 1 to Rad 6) were cut at approximately 60° radial spacing from the cylindrical piece and three samples (L, W and D) from the base slab. The axes of these latter samples were parallel, respectively, to each of the three sets of parallel faces of the slab. The samples were machined to 0.5" diameter and 2" length, the latter being about the minimum length for which consistent data are obtained for permeability⁽²⁾.

Results from only three samples are presented here; these are Rad 1, Rad 2 and L, the latter being parallel to the direction of extrusion of the block. (The other samples have not been oxidised to the same extent yet but results to date show similar trends.)

The samples were mounted in a silica holder and oxidised by atmospheric air in a silica tube furnace at 500 $\pm$ 5°C. They were allowed to cool and stand

in the atmosphere for at least 24 hours before the diffusion coefficient was measured. The furnace temperature of 500°C was chosen to ensure uniform oxidation throughout the graphite; it is well below the temperature where, in a sample of this size, diffusion becomes limiting and the centre of the specimen starved of oxygen⁽³⁾. The oxidation rates were found to be about 1×10^{-4} gm/gm. hr. This is of the same order as those found elsewhere⁽³⁾. In spite of the fact that the specimens were handled carefully, however, the rate of oxidation varied considerably even in successive tests on the same sample. Again, this is not surprising, since great precautions must be taken to ensure consistent results even with purified graphite⁽⁴⁾. Since the object was merely to cause successive changes in the total oxidation of the sample rather than to study those changes themselves, the data on oxidation rate are not important and are not given in detail.

The apparatus and techniques used in measuring diffusion coefficient for the present tests were essentially the same as those described previously⁽¹⁾. Oxygen and nitrogen respectively were made to stream past opposite ends of the sample and the amount of oxygen picked up in the nitrogen stream was recorded by a Hersch meter. As before, oxygen permeability was also measured by the pressure rise method.

As mentioned above, it had previously been necessary to reduce the diameter of the sample in successive tests. This difficulty arose from the fact that the samples were encapsulated in Araldite resin for the diffusion measurement; some of the graphite had to be removed to ensure complete removal of the Araldite before each stage of oxidation. This procedure has been avoided in the tests reported here by the development of a new method of sealing. The graphite samples were sealed in a pressurised rubber sleeve; further details will be given in the final report on these tests.

3. Results

The results of the diffusion experiments are presented in Figure 1 and the viscous permeabilities, determined on the same specimens, are given in Figure 2. The diffusion results are presented as the ratio (m) of the effective to the "free gas" diffusivity. It is seen that both m and B_0 , the viscous permeability, increase continuously with total oxidation. It seems that, over the range investigated, the viscous permeability increases in about the same proportion

as the diffusivity ratio. All the samples appear to be showing similar trends with oxidation but the values of m and B_0 are rather higher for the sample cut in the direction parallel to the direction of extrusion.

4. Conclusion

The four-fold change in m in the range 0-9% total oxidation is certainly significant and should be taken account of in applying theories of diffusion control in oxidation. More complete data are now being obtained.

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