

PREFERRED ORIENTATION OF COKE PARTICLES

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Introduction

The evaluation of different cokes as precursors for carbon and graphite products usually involves the fabrication of small-scale artifacts for the determination of physical properties. The coefficient of thermal expansion (CTE) of the graphite artifacts has been widely employed to assess the suitability of anisotropic cokes for graphite electrodes and more isotropic cokes for specialty graphites.

Microscopy techniques have been applied directly to coke particles in order to assess the carbonized mesophase domain structure and provide a measure of the coke anisotropy [1]. However, these methods have been difficult to quantify. In this paper, we report on the use of x-ray diffraction to quantitatively determine directly the degree of preferred orientation in a coke particle. The preferred orientation parameter is a direct measure of coke anisotropy and correlates with the CTE value for its derived graphite.

Experimental

A custom assembled diffractometer system was used to measure the average (002) diffracted intensity. Typically, in x-ray diffraction the volume of sample measured is too small to effectively respond to the type of anisotropy present in coke particles. This limitation was overcome using molybdenum or silver target tubes and a combination of translation and rotation required for anisotropy measurement. This arrangement greatly enhanced the sample volume in the diffraction experiment. Other salient features of the apparatus designed for measurement of coke particle preferred orientation are the HOPG monochromator, a soller slit system and the associated PC control data acquisition and control.

A. Sampling and Measurement

The commercial manufacture of coke is a large scale batch operation which can impart significant coke property variations within different parts of the coker. Therefore, in order to obtain a representative assessment, it is essential that multiple samples are obtained followed by careful blending and sizing. Another important factor for selection of coke particles for anisotropy determination is the effect of coke particle size on the particle anisotropy and the diffracted x-ray intensity. Very small particles are highly anisotropic and yield low diffracted intensities and very large particles tend to become isotropic for even the most highly oriented cokes.

Based on experimental trials, calcined coke particles were sieved through 3/6 Tyler screen and 40 particles were selected for the anisotropy measurement. These coke particles were then graphitized, since graphitization does not change the coke particle anisotropy but increases the x-ray diffracted intensity and, thereby, improves the measurement statistics.

Figure 1 is a typical representation of the intensity versus orientation as a function of rotation angle ϕ . The intensities observed at the minima between the orientation maxima are points used for the evaluation of the background scattering. Since the background is primarily due to instrumental scattering, the normal procedure is to evaluate the background for ten highly anisotropic particles. To be useful, the background corrected scattering must be transformed into a characterization parameter. For the type of orientation observed in coke particles the integral parameter "R" represented by the following equation was selected [2]:

$$R = \frac{\int I(\phi) \sin^3 \phi d\phi}{\int I(\phi) \sin \phi d\phi}$$

where $I(\phi)$ is the scattered x-ray intensity as a function of the rotation angle ϕ . The range for "R" encompasses a lower limit $2/3$, for a completely isotropic scattering intensity, and a maximum value of 1.0 for the delta function intensity obtained from a particle with perfect alignment.

B. Applications

The x-ray preferred orientation was measured on a variety of cokes obtained from different commercial sources. The validity of the measurements was tested by comparing them to the CTE values determined for graphite produced from the same precursor cokes. It should be mentioned that the graphite artifact, used for CTE measurements in this study, was prepared from coke flour. The relationship between the measured R values for a series of 3/6 coke particles and the corresponding CTE values are plotted in Figure 2. For the broad range covered, the R values do correlate with the coke CTE. However, an important trend to be observed in Figure 2 is that for lower CTE values the trend of higher anisotropy is not reflected in a continuous reduction in the graphite artifact CTE. Thus, a progressive increase in anisotropy is not associated with a lowering of the fine grain CTE.

References

1. Zeng, S. M. and Eser, S., 22nd Conference on *Carbon*, (1995) 260.
2. Price, R. J. and Bokros, J. C., *Appl. Phys.*, 36, 1897 (1965).

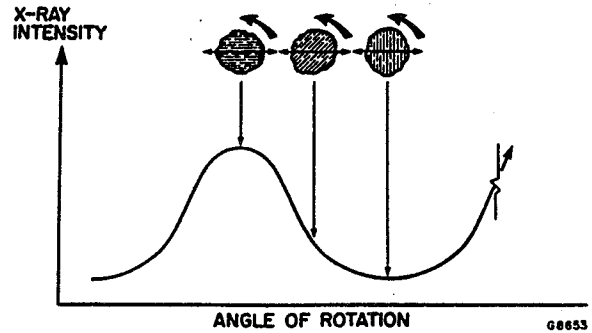


Figure 1. Diffraction intensity trace for sample with internal preferred orientation.

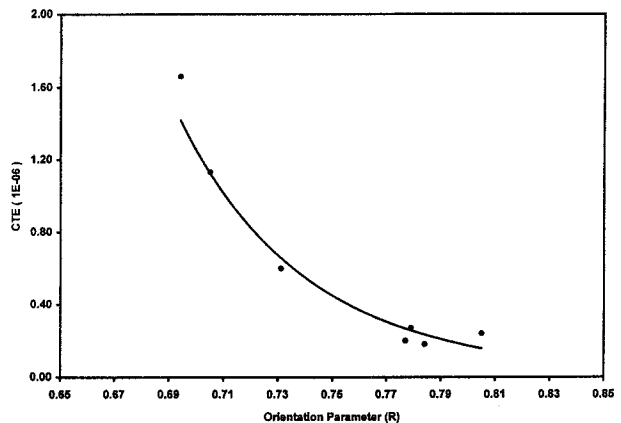


Figure 2. Relationship between the particle orientation and CTE of the graphite artifacts produced from the precursor cokes.