A COMPARISON OF OPTICAL METHODS FOR DETERMINING PREFERRED ORIENTATION IN NEARLY ISOTROPIC PYROCARBON

D. W. Stevens
General Atomic Company
San Diego, California 92138

Introduction

Nearly isotropic pyrocarbon deposited in fluidized beds has found important application as a coating on fuel particles for high-temperature gas-cooled reactors [1]. It has been known for more than a decade that the dimensional stability of these coatings during fast neutron irradiation requires an initially low preferred crystallite orientation [2]. Only recently has it been quantitatively shown by Kaae et. al. [3] that the required preferred orientation is indeed very low. The difficulty of making accurate x-ray measurements of low preferred orientation is largely responsible for the long time lapse between these developments. The use of optical reflectivity methods has alleviated this problem somewhat [4].

A highly sensitive optical method has been under development at General Atomic Co. [5,6]. This approach involves the use of a reflecting microscope with both a polarizer and an analyzer (i.e. a micropolarimeter). Recently this method was modified which resulted in a more practical and more powerful micropolarimeter [7]. This device is operated in a manner analogous to synchronously rotating the polarizer and the analyzer, and is referred to as a synchronous micropolarimeter (SMP). At the same time, General Atomic has been operating a device which is essentially a duplicate of the instrument described in Ref. 4. This microscope is used to measure reflected intensities as a function of the polarization direction of normally incident light, and is referred to as a polarizing microreflectometer (PMR). The subject of this report is a comparison of preferred orientation measurements obtained from the SMP and the PMR.

Experimental

The methods used to deposit pyrocarbon in fluidized beds have been described previously [8]. The carbons were deposited onto small graphite discs which were incorporated into the fluidized bed. The source gas was propylene, and the deposition temperature ranged from 1300° to 1500°C. (Although deposition temperature remained essentially constant throughout each individual coating run.) Deposition conditions were controlled so that several deposits were produced at widely different coating rates, but with essentially constant densities. Sample series of this nature were prepared at six different densities (see results below). The deposits were removed from the graphite substrates and mounted in polyvinylchloride. They were then sectioned perpendicular to the plane of the substrate and mechanically polished. Final polishing was done using 0.05μm alumina on an automatic polishing machine.

The SMP employs a He-Ne laser for a light source and the test region on the sample surface was 21μm in diameter. The PMR uses a white light source and the test region was 24μm in diameter. The sample surface was in immersion oil in both instruments. Local inclinations of the sample surface are corrected in the SMP by tilting the sample. This capability is not available in the PMR. Each test region was identified with fiducial marks. Using these marks, measurements in the two instruments were obtained from essentially the same test regions. Optical anisotropy measurements from each instrument are transformed to preferred orientation in terms of the Bacon Anisotropy factor (BAF). BAF is unity for uniformly distributed crystallites and increases without limit as the crystallites approach parallel alignment.

Results and Discussion

Measurements were made at six test regions on each of 33 disc samples. The results from the SMP are shown in Fig. 1. At a given density, BAF falls off rapidly with increasing coating rate initially, and changes very slowly at high coating rates. For constant coating rate, BAF increases with increasing density, and appears to be comparatively sensitive to density at high density values.

The results from the PMR fell in about the same range and exhibited a similar dependence upon coating rate, but a clear systematic variation with density could not be resolved. This comparative characteristic of the two instruments apparently arises from the improved sensitivity of the SMP. The higher sensitivity of the SMP is demonstrated in the two optical micrographs in Fig. 2. These are micrographs of the same pyrocarbon coated fuel particle. A polished metallographic section through the midpoint of the particle is viewed in reflection using horizontally polarized light at normal incidence. Without an analyzer, a slightly higher reflected intensity is observed at the top and bottom positions on the coating (i.e. 12:00 o'clock and 6:00 o'clock positions) as compared to the intensities at the rightmost and left-most positions (i.e. 3:00 o'clock and 9:00 o'clock positions). This variation which corresponds to the signal detected by the PMR, is very subtle and difficult to resolve even though the preferred orientation of this pyrocarbon is more than an order of magnitude beyond the range of interest.

However when an analyzer is inserted into the reflected light with verticle transmission axis, a pronounced variation in intensity is observed at different positions along the circumference of the coating. If reflection from the sample surface does not cause the plane of polarization to shift from its original horizontal direction, then the analyzer will absorb the light. This condition occurs at four positions on the pyrocarbon coating, causing dark bands at the 12:00 o'clock position and at 90 degree intervals from that position. At intermediate positions, a rotation of the polarization direction allows a component of the reflected light to be transmitted by the analyzer. This rotation is directly related to the variation in reflected intensity observed with only a polarizer [9], and the net effect is that the analyzer has provided a pronounced improvement in sensitivity.

BAF measurements from the two instruments were correlated using three "least-squares" criteria. The line $Y=a+bX$ was fit to the data X_i, Y_i, where X_i is the value of (BAF-1) from the SMP, and Y_i is the value of (BAF-1) from the PRM, each from the i^{th} test region. The parameters "a" and "b" were chosen so as to minimize Σd_i^2 where d_i is the distance between the i^{th} data point (X_i, Y_i) and the line fit to the data. The path over which d_i was obtained was determined by the least-squares criterion: for criterion 1, the

path was parallel to the Y-axis, for criterion 2, the path was parallel to the X-axis, and for criterion 3, the path was perpendicular to the line being fit to the data (i.e. for criterion 3, d_i was the shortest distance between the point and the line). The results are shown in Table 1. In each criterion the correlation coefficient was 0.87 ± 0.02, but the variation in the parameters "a" and "b" according to the criterion might be a better indication of how well the relationship is known.

Criteria for choosing between the three lines are not completely clear. When X is well known and scatter is attributed only to Y, then criterion 1 is chosen, (vise versa, then criterion 2 is accpeted). But when scatter is equally attributable to X and Y, then criterion 3 should be chosen. Very little statistical information is available for making a choice. However, there is a physical basis for expecting $BAF_{PMR} \cong BAF_{SMP}$, and since the third criterion yields nearly unit slope, it is the recommended choice.

References

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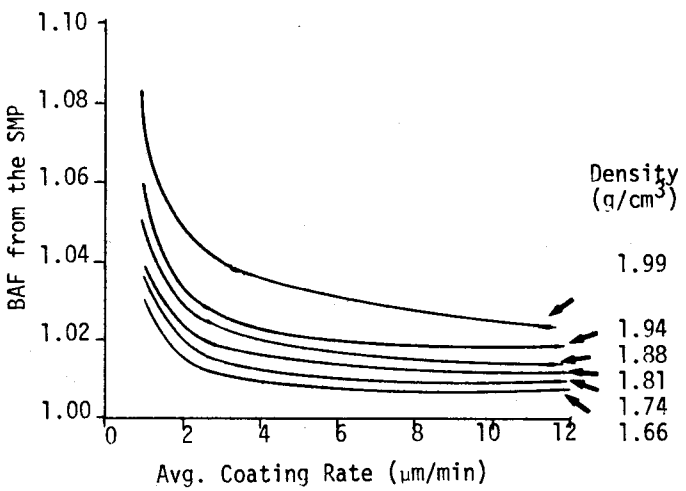

Fig. 1. Influence of Coating Rate and Density on BAF from the SMP

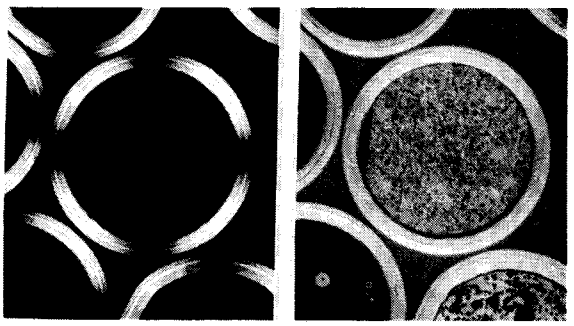

Fig. 2. Micrograph of an anisotropic pyrocarbon coating on a fuel particle, showing the effect of an analyzer on the optical signal.

TABLE 1
Least Squares Parameters for Correlation Between Preferred Orientation Data from the SMP and the PMR

Criterion	a	b	σ_x (†)	σ_y (‡)
1	0.014	0.90	0.0116	0.0104
2	0.005	1.18	0.0102	0.0119
3	0.010	1.03	0.0105	0.0108

† σ_x is the standard deviation from the line in the direction parallel to the x-axis.

‡ σ_y is the standard deviation from the line in the direction parallel to the y-axis.