SMALL ANGLE X-RAY SCATTERING INVESTIGATION OF PYROLYTIC CARBON

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Introduction

Silicon carbide alloyed isotropic pyrolytic carbons (PyC) which are deposited in a bed of fluidized particles are the principle material used in the manufacture of mechanical heart valves. The microstructure of these pyrolytic carbons are composed of roughly spherical growth features. These growth features contain both SiC particles and turbostratic carbon "crystallites" whose c-axis's are oriented perpendicular to the growth feature boundary [1].

Previous investigations of PyC microstructure have relied primarily on the use of transmission electron microscopy and x-ray diffraction. This work represents an effort to develop a possibly faster bulk method for measuring the average sizes of the growth features, SiC particles, and porosity by use of small angle x-ray scattering (SAXS).

Experimental

Alloyed and unalloyed PyC samples were coated in a fluidized bed reactor process described previously [2]. The graphite substrate on which the PyC coatings were deposited were subsequently removed. The substrate free samples were then ground to a powder.

SAXS data was collected on two instruments at the University of New Mexico Center for Advanced Microceramics. The low scattering vector ($0.0002<q<0.1$ Å^{-1}) data was collected on a Bonse-Hart system, and the high ($0.03<q<0.7$ Å^{-1}) data was collected on a short geometry pinhole system coupled to a two dimensional detector.

For this set of experiments the Guinier model was chosen. This model assumes a single phase material with discrete particles of a single shape but different sizes. The void fraction and alloying constituent (2-3atomic %) for this type of PyC are very low, thus, allowing a single phase assumption; furthermore TEM results [3,4] showed this type of PyC is composed of spherical growth features.

The theoretical intensity for the Guinier model can be expressed as

$$I(q) = I(0)e^{\frac{-q^2 R_g^2}{3}}$$

where $I(0)$ is the scattering intensity at the scattering vector with zero magnitude, $q=(4\pi/\lambda)\sin\theta$, and R_g is the scattering radius of gyration. This R_g is a measure of the size of the particles equivalent to the radius of gyration defined in mechanics. For spherical particles R_g is defined as

$$R_g^2 = \frac{3r^2}{5}$$

where r is the radius of the sphere.

Results

Figures 1 and 2 show the plots of the logarithm of the intensity verses the logarithm of the magnitude of the scattering vector for unalloyed and alloyed PyC, respectively. Initial inspection of these plots showed a q^4 dependence at low q and a q^2 dependence at high q with a smoothly changing curvature in between.

The radius of gyration of the scattering features was calculated from linear regions of the plots by fitting the experimental results to the above model using the method of least squares. Table 1 shows a summary of the calculated R_g and their interpretation assuming spherical particles.

Discussion

Reviewing the results of published work [3,4] shows that the growth feature radii ranging in size from 1000 - 5000 Å agrees well with the larger of the scattering particles in Table 1. The difference in the growth feature size between the alloyed and unalloyed PyC could be due to different coating conditions. A TEM examination will need to be done to confirm these growth feature sizes.

Comparing alloyed to unalloyed PyC shows an additional R_g value which is clearly due to the alloying constituent SiC. For simplicity, Table 1 assumes that the SiC is spherical in shape yielding a particle radius of 62 Å which is on the order of that observed in reference 4.

For the radii of gyration in the 16.5 to 17 Å range, it is tempting to attribute this to the turbostratic crystallites since their out of plane coherence lengths (L_c) are 40 to 45 Å exactly the diameter calculated in Table 1. It is, however, doubtful that these turbostratic "crystallites" create a sharp enough interface to be considered scattering particles. The interpretation here will consider these scattering particles to be mesopores.

Finally, both alloyed and unalloyed SAXS results revealed a radius of gyration in the 3 to 4.5 Å range interpreted as spherical particles with radii on the order of 4 to 6 Å. No feature of this size is known to exist in PyC, and, therefore, it is assumed that scattering is due to micropores.

Conclusion

SAXS was used to characterize an alloyed and unalloyed from of PyC. Comparisons with previous work shows generally good agreement on the size of growth features and SiC particles. Additionally, it was assumed that the two other scattering sites where possibly due to mesopores and micropores.

Acknowledgments

The authors wish to acknowledge the contributions of Richard Magrum, Randy Weldy, and Cephas Wozencraft in preparing the samples.

References

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2. Bokros, J.C., *Carbon*, 1965, 3, 17.
3. Kaae, J.L., *Carbon*, 1975, 13, 55.
4. Kaae, J.L., *Carbon*, 1975, 13, 51.

Figure 1: Unalloyed Pyrolytic Carbon

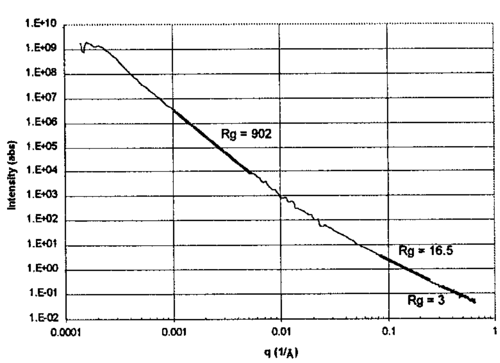

Figure 2: SiC Alloyed Pyrolytic Carbon

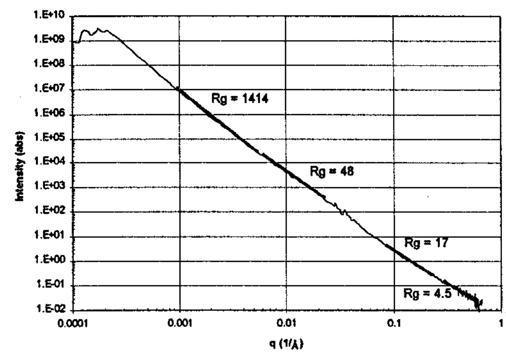

Table 1

Unalloyed PyC *Figure 1*		Alloyed PyC *Figure 2*	
R_g (Å)	r (Å)	R_g(Å)	r (Å)
902	1164	1414	1825
16.5	21	48	62
3	4	17	22
		4.5	6