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Texture analysis of pyrolytic carbon by polarized light microscopy and selected area electron diffraction: A quantitative model for the correlation between extinction angle and orientation angle

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Abstract

Many properties of pyrolytic carbon depend strongly on the degree of texture which is frequently analyzed by polarized light microscopy (PLM) and selected area electron diffraction (SAED). PLM allows the fast and simple determination of the extinction angle Ae. SAED exhibits a higher spatial resolution which is mainly determined by the diameter of the SAED aperture and allows the determination of the orientation angle.

A quantitative model for the relationship between the extinction angle determined by PLM and the orientation angle determined by SAED is presented and applied to our experimental data. The distribution of the orientation of coherent domains is derived from SAED data and the reflection coefficients of pyrolytic carbon are calculated as the sum of the reflection coefficients of the coherent domains. The only fit parameters in this model are the ratio of the reflection coefficients of the coherent domains for extraordinary and ordinary rays r_e/r_o and the relative phase shift Δ. A good agreement between calculation and experiment is achieved for a r_e/r_o of 0.33 and a phase shift |Δ| of 75°. The results allow a quantitative comparison of extinction angles with orientation angles.

Keywords: Pyrolytic Carbon, Chemical Vapor Deposition, Optical Microscopy, Electron Diffraction, Texture

1. Introduction

Different methods are used for the determination of the degree of texture of pyrolytic carbon. Besides x-ray diffraction, polarized light microscopy (PLM) and selected area electron diffraction (SAED) in a transmission electron microscope are the most popular techniques.

Only empirical data for the relationship between orientation angle (OA) determined by SAED and extinction angle (Ae) determined by PLM [1, 2] have been given in literature. Here [3] we present a quantitative model describing this relationship. Extinction angles are calculated from SAED profiles and compared with experimental data.

2. Theoretical Description

Pyrolytic carbon is composed of turbostratic domains of nearly parallel, slightly distorted graphene layers. In the model presented here, azimuthal SAED intensity profiles I(α) are interpreted as the distribution of twisting of these turbostratic domains. For pyrolytic carbon layers deposited on plane substrates the carbon layer is symmetric against rotation with respect to the growth direction: it can therefore be assumed that the distribution of twist angles α is identical with the distribution of tilt angles β (this assumption also holds for pyrolytic carbon layers on fibers for sufficiently small SAED apertures, for details see [3]). This leads to the following expression for the distribution of twist α and tilt β of the turbostratic domains:

$$P(\alpha,\beta) = \frac{I(\alpha)\cdot I(\beta)}{\int_{-90^\circ}^{90^\circ}\int_{-90^\circ}^{90^\circ} I(\alpha)\cdot I(\beta)\,d\alpha\,d\beta} \qquad \alpha,\beta \in [-90^\circ, 90^\circ]$$

The expected average optical properties of the pyrolytic carbon sample can be calculated by assuming that the reflection coefficients correspond to the sum of the reflection coefficients of all turbostratic domains. This summation is carried out by integration taking into account the orientation of the turbostratic domains. The ratio of reflection coefficients of the turbostratic domains for extraordinary and ordinary rays r_e/r_o and their relative phase shift Δ are not known a priori and are considered as fit parameters in the model.

Based on the calculated reflection coefficients the extinction angle of the sample Ae_{SAED} can be calculated [1] and be compared with the extinction angle Ae_{PLM} measured by PLM.

3. Experimental

In this study pyrolytic carbon layers deposited on plane substrates by chemical vapor deposition as well as the pyrolytic carbon matrices deposited on carbon fiber bundles by chemical vapor infiltration were investigated (for details see [3]).

SAED was carried out using a 120 keV Zeiss EM 912 Omega transmission electron microscope equipped with an electron-energy spectrometer integrated into the projection lens system [4].

PLM was carried out using an optical microscope

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equipped with a polarizer and a rotating analyser. The plane pyrolytic carbon layers were cut parallel to the growth direction. For the investigation of infiltrated carbon fiber bundles, polished cross sections perpendicular to the fiber axis were used (for details see [1, 2]).

3. Results and Discussion

Based on the azimuthal intensity scans the corresponding extinction angles Ae_{SAED} were calculated as described above for three samples of plane pyrolytic carbon layers and for ten samples of infiltrated carbon fiber bundles and compared with extinction angles Ae_{PLM} measured by PLM (see Fig. 1).

Good agreement between Ae_{SAED} and Ae_{PLM} is achieved using as values for the fit parameters: ratio of reflection coefficients r_e/r_o = 0.33 and phase shift $|\Delta|$ = 75° for the turbostratic domains.

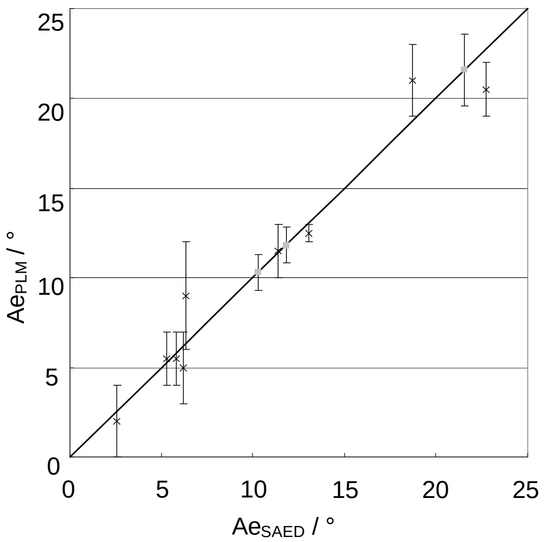

Fig. 1. Comparison between extinction angles determined by polarized light microscopy Ae_{PLM} and calculated from SAED data Ae_{SAED}. Ae_{SAED} was calculated from SAED data using a ratio of reflection coefficients r_e/r_o = 0.33 and a phase shift $|\Delta|$ = 75° for the turbostratic domains. The black crosses correspond to infiltrated fiber bundles; grey squares correspond to pyrolytic carbon layers on plane substrates.

In the following the optical properties obtained by fitting will be used to calculate the relationship between extinction angle and orientation angle. For this calculation it is assumed that the distribution of twist and tilt follows a Gaussian distribution.

The required optical parameters are taken from the fit presented in Fig. 1: r_e/r_o = 0.33 and $|\Delta|$ = 75°. The calculation is carried out for different orientation angles resulting in the correlation between orientation angle and extinction angle illustrated in Fig. 2.

Pyrolytic carbon is classified by polarized light microscopy into three categories for samples with local rotational symmetry: low textured (LT, Ae_{rot} < 12°), medium textured (MT, 12° < Ae_{rot} < 18°) and high textured (HT, Ae_{rot} > 18°) carbon. According to the presented model, these classes correspond to the following ranges of orientation angles (OA): low textured (OA > 71°), medium textured (54° < OA < 71°), high textured (OA < 54°).

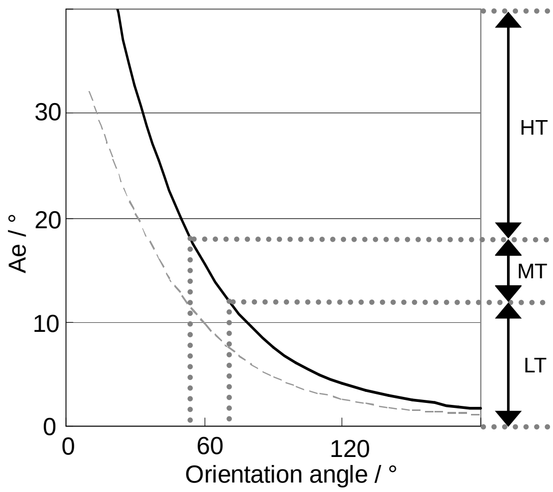

Fig. 2. Correlation between extinction angle Ae determined by polarized light microscopy and orientation angle determined by SAED calculated for a ratio of reflection coefficients r_e/r_o = 0.33 and a phase shift $|\Delta|$ = 75° for graphite. The black solid line was calculated for samples with rotational symmetry, the dashed line was calculated for flat samples.

4. Conclusions

A quantitative model for the relationship between extinction angle determined by PLM and the orientation angle measured by SAED was presented for pyrolytic carbon. It is based on the assumptions that the distribution of the orientation of turbostratic domains can be derived from the SAED azimuthal intensity distribution and that the reflection coefficients of pyrolytic carbon can be considered as the sum of reflection coefficients of the turbostratic domains taking into account their orientation. A good agreement between extinction angles calculated from SAED data and those measured by PLM is thus achieved for a ratio of reflection coefficients r_e/r_o of 0.33 and a phase shift $|\Delta|$ of 75°. These fit parameters were used to derive the relationship between extinction angle determined by PLM and orientation angle determined by SAED.

The described model represents the first quantitative description of the relationship between extinction angle and orientation angle measured for pyrolytic carbon.

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