Elastic constants of high-texture pyrolytic carbon measured by ultrasound phase spectroscopy [Letter]

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Elastic constants of high-texture pyrolytic carbon measured by ultrasound phase spectroscopy

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Pyrolytic carbon (PyC) is one of the commonly used micro constituents of carbon/carbon (C/C) composites. Although this material has been extensively studied since the 1960s, there is a lack of information on the correlation between the elastic properties and microstructure of this anisotropic graphite-like material. In 1963, Papadakis and Bernstein [1] measured elastic properties of pyrolytic graphite blocks by ultrasonic pulse-echo testing. However, these studies did not provide any structural information such as the texture degree of pyrolytic carbon.

Some of the elastic moduli were measured by Pappis and Blum [2] by tension, compression and bending tests. The effective elastic moduli of PyC can be extracted from results

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of nano- and microindentation [3–6]. However, these results are not sufficient for the numerical modeling of the mechanical behaviour of the composites including PyC.

The present study aims to obtain all independent elastic constants for a PyC material with a well-documented texture degree. For this purpose the texture degree has been comparatively studied by polarized light microscopy (PLM) and high-resolution transmission electron microscopy (HRTEM) while the elastic constants have been determined by ultrasonic measurements.

The PyC investigated in present study was obtained from Schunk Kohlenstofftechnik GmbH, Heuchelheim, Germany, in the form of discs with a diameter of 100 mm and a thickness of 1.5 mm. According to the manufacturer the total amount of impurities is reported to be less than 10 ppm.

First, the spatial anisotropy of the PyC disc has been analyzed at the micrometer scale. For this purpose, the texture degree has been studied by an improved PLM technique [7,8] allowing monitoring the texture gradients in planar deposits at the broad scale ranging approximately from 0.001 to 10 mm. The perfection and texture degree of the graphene stacks at the nanometer scale along the disc thickness has been studied by HRTEM coupled with image analysis.

Fig. 1 is a pair of optical micrographs showing typical microstructures of the PyC disc viewed with polarized light. Overlapping elongated cones appear in the cross-section prepared parallel to the deposition direction (a). The disc surface topography appears as a superposition of half-sphere units (b). Both cross-sections manifest that the studied material is a polycrystalline solid.

The results of texture examination are presented in Fig. 2. Fig. 2a shows a PLM micrograph of a polished disc cross-section. The optical anisotropy of the PyC disc has been measured within three areas corresponding to its top, centre

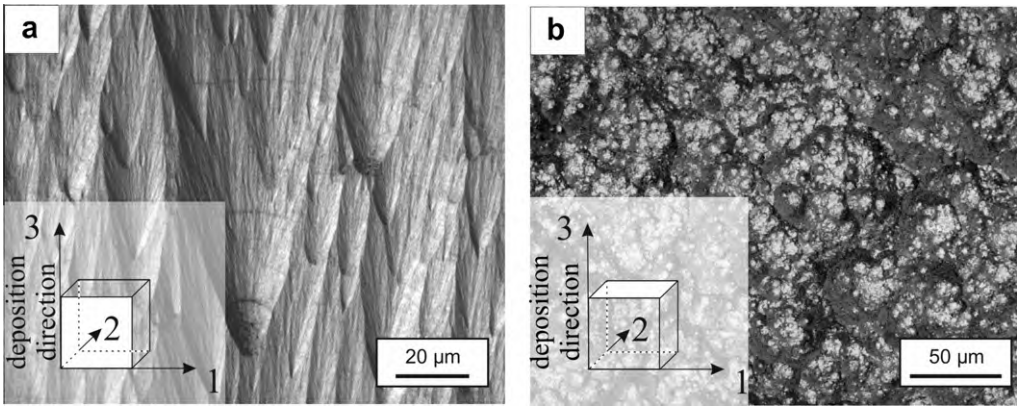

Fig. 1 – Polished cross-sections of the pyrolytic carbon disc viewed in polarized light. (a) Cross-section parallel to deposition direction and (b) cross-section perpendicular to deposition direction.

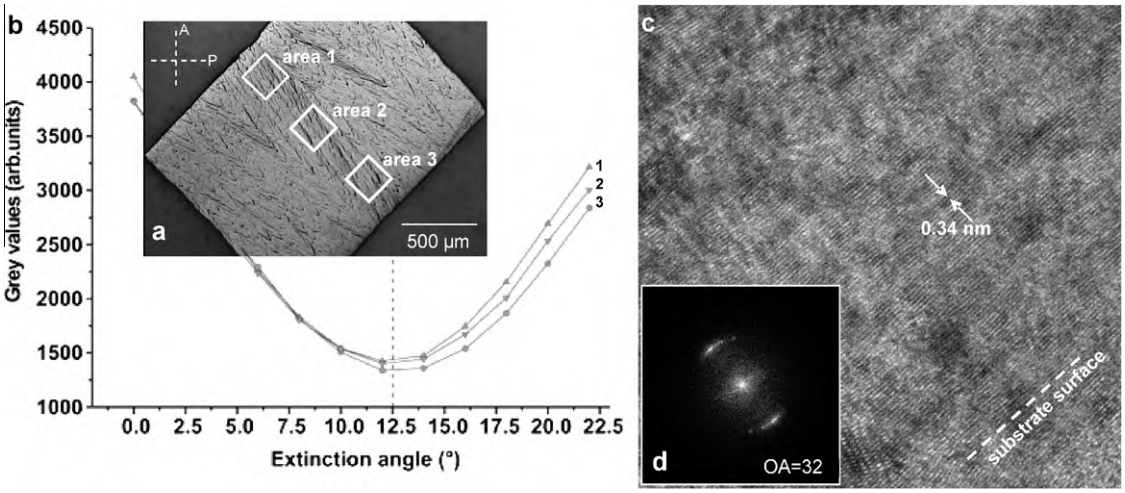

Fig. 2 – Texture examination through the thickness of the PyC disc. (a) Polarized-light optical micrograph of a cross-section. The sample is rotated to 45° with respect to the crossed analyzer (A) and polarizer (P) shown by the dashed lines. The white square-labeled areas were selected for measurements of extinction angle. (b) Photometric plots (values of the extinction angle) from the selected areas shown in (a). (c) HRTEM image showing typical for high-textured PyC graphene stacks. The dashed line in (c) shows the approximate substrate orientation. (d) FFT pattern from (c) indicating orientation angle (OA) of 32° ± 5° corresponding to the high-textured PyC [9,10].

and bottom (areas 1–3). Fig. 2b demonstrates that in all areas the extinction angle is about 12.5° corresponding to high-textured PyC [7,8] and the absence of the textural gradients through the disc thickness.

Similar to PLM (Fig. 2a), three areas corresponding to the top, centre and bottom of the PyC disc have been selected for HRTEM studies. About 3–5 HRTEM in each point image have acquired and compared. Fig. 2c shows a representative HRTEM image of graphene stacks taken from the selected disc areas across its thickness. The lattice distance between single graphene layers is about 0.34 nm. The texture degree of the stacks has been analyzed by a Fourier image analysis (FFT) of HRTEM images [9]. The corresponding to the HRTEM image (Fig. 2c) FFT pattern (Fig. 2d) shows two arc-shaped diffraction maxima exhibiting orientation angle of 32° ± 5° which is typical for the high-textured pyrolytic carbon [9,10]. Therefore, both PLM and HRTEM studies demonstrate that the studied material is a high-textured, gradient free PyC.

For ultrasonic studies, beams with cross-section $1.5 \times 1.5\ mm^2$ were cut from the middle of the disc using a diamond wire saw according to Fig. 3a. The two outer beams were cut into 36 samples (dimensions about $1.5 \times 1.5 \times 5.0$ mm), for which the density was determined according to the Archimedes principle. Measurements of 36 samples show a mean density of $2.178\ g/cm^3$ and a standard deviation of $0.0078\ g/cm^3$ and it can be concluded that the PyC material can be considered to be homogeneous. For ultrasound phase spectroscopy measurements, the inner beams were cut into samples with faces perpendicular to the 2-axis (Fig. 3b) and into samples rotated under 45° to the 3-axis (Fig. 3c). The obtained samples were sufficient for wave velocity measurements to determine all five independent elastic constants for transversal isotropic material.

The velocities were measured with ultrasound phase spectroscopy [11–13]. An Advantest network analyzer (R3754A, ADVANTEST®) was used to generate continuous harmonic waves. Two identical pairs of transducers were used for longitudinal (Panametrics V122, PANAMETRICS®) and shear waves (Panametrics V155, PANAMETRICS®) to measure the phase delay in the frequency range of 10 kHz–15 MHz for longitudinal waves and 10 kHz–10 MHz for shear waves. Water solvable syrup was used to couple the transducers to the samples.

The results of the ultrasonic measurements are presented in Table 1. When averaging the results for U_{11} and U_{22}, for U_{12} and U_{21}, and for U_{23}, U_{32}, U_{13} and U_{31}, very small relative standard deviations are obtained, which indicates the presence of transverse isotropy. The velocity U_{1313} is a longitudinal wave

Table 1 – Results of ultrasound phase spectroscopy. The right column of the wave velocity shows the standard deviation of all measurements divided by the mean velocity.

	Wave velocities considered for averaging	Average values for velocities (m/s)	Relative standard deviations (%)
Four samples according to Fig. 3b	U_{11}, U_{22}	4304	3.3
	U_{33}	2902	2.4
	U_{12}, U_{21}	2151	5.5
	U_{23}, U_{32}, U_{13}, U_{31}	907	2.6
Six samples according to Fig. 3c	U_{1313}	3370	4.3

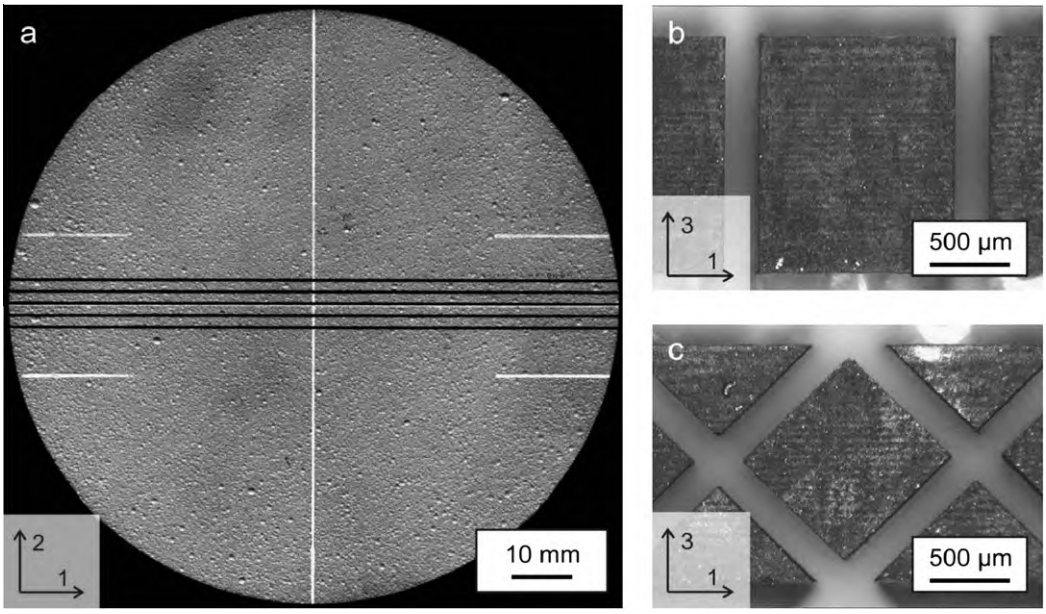

Fig. 3 – Preparation of samples for ultrasound phase spectroscopy: (a) Beams cut from the middle part of the PyC disc (view from the top, white lines are guidelines for preparation), (b) cut of the beam parallel to deposition direction and (c) cut of the beam under angle 45° to the deposition direction.

Table 2 – Calculated elastic constants from the wave velocities presented in Table 1 and comparison to literature values. The values in brackets for the elastic constants represent the standard deviation divided by the mean value for the presented results.

PyC	ρ (g/cm^3)	Measurement method	C_{11} [GPa]	C_{12} [GPa]	C_{13} [GPa]	C_{33} [GPa]	C_{44} [GPa]	E_1 [GPa]	E_3 [GPa]
Present studies	2.178	Ultrasound phase spectroscopy (continuous waves technique)	40.0 (7%)	20.0 (17%)	13.1 (46%)	18.2 (5%)	1.8 (6%)	27.1	12.8
[1]	2.16	Ultrasonic (impulse-echo technique)	52.1 (5%)	18.8 (23%)	18.0 (56%)	25.8 (5%)	1.5 (5%)	38.6	16.7

velocity propagating in the bisectrix in the 1–3-plane according to Fig. 3c.

Based on the wave velocities presented in Table 1, the elastic constants are calculated [14] and compared to literature values in Table 2. It can be seen that the magnitude of the elastic constants is in general smaller compared to the results of Papadakis and Bernstein. Probably the texture degree of the material studied by Papadakis and Bernstein [1] and in the present study was not identical. The relative inaccuracy of the elastic constants is in the same range compared to Papadakis and Bernstein (values in brackets).

The authors explained the high inaccuracy of C_{13} with the error propagation for this constant. By comparison to destructive compression tests, they concluded that the value of C_{13} is quite realistic and the error should be much smaller than estimated on the basis of the ultrasonic measurements and error propagation. Instead of estimating the maximum experimental error [1] we used the standard deviations of the corresponding velocities as a value of inaccuracy.

All independent elastic constants for pyrolytic carbon with a well-documented texture are measured experimentally. These results are of especial importance for the numerical modeling of mechanical behaviour of composites including PyC as micro constituents.

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