ELSEVIER

Nuclear Instruments and Methods in Physics Research A 467–468 (2001) 384–387

NUCLEAR INSTRUMENTS & METHODS IN PHYSICS RESEARCH Section A

www.elsevier.com/locate/nima

Recent developments of high quality synthetic diamond single crystals for synchrotron X-ray monochromators

A.K. Freund[a,*], J. Hoszowska[a], J.P.F. Sellschop[b], R.C. Burns[c], M. Rebak[c]

[a] *European Synchrotron Radiation Facility, B.P. 220, 38043 Grenoble, France*
[b] *University of the Witwatersrand, P.O. Box WITS, 2050 Johannesburg, South Africa*
[c] *De Beers Diamond Research Laboratory, P.O. Box 1770, 2135 Southdale, South Africa*

Abstract

For several years now, the ESRF, the University of the Witwatersrand and Messrs. De Beers Industrial Diamonds (Pty) Ltd. through their Diamond Research Laboratory, have pursued a development programme to assess and improve the quality of synthetic diamonds. Recently, in an effort to study the influence of nitrogen impurities on the defect structure, X-ray excited optical luminescence and spatially resolved double-crystal diffractometry were employed as new techniques. The correlation between nitrogen impurities and the raw defect structure was clearly visible. It was confirmed that concentration variations are related to lattice imperfections, where tilts are much more important than lattice constant variations. High reflectivity was observed and quite large zones of a sample bigger than 1 cm^2 showed to be perfect to within better than 0.5 arcsec.

PACS: 07.85.q; 07.85.y; 41.50 + h

Keywords: X-ray monochromators; Heat-load; Diamond crystals; Synchrotron instrumentation

1. Introduction

Diamond single crystals are of increasing importance as monochromators for third and fourth generation X-ray sources, i.e. for synchrotron radiation from storage rings (SR) and future free electron lasers (FELs) because of their excellent thermal and crystallographic properties, see, e.g. Ref. [1]. Their application still remains limited to a few experiments where their mosaicity, at present about 2 arcsec, is acceptable. Therefore, it is crucial to further improve the crystalline quality of synthetic diamond. In type Ib diamond the major impurity is nitrogen, whereas the colourless type IIa diamond is effectively free of or very low in nitrogen.

Many type Ib samples have been characterised in a collaboration between the ESRF, the University of the Witwatersrand and De Beers Industrial Diamonds (Pty) Ltd. by means of traditional X-ray rocking curve measurements and topography [2,3]. In a recent study of several type Ib samples, an average mosaic spread of 2 arcsec was found [3]. For 12.4 keV X-rays the measured (1 1 1) reflectivities were 60% and 50% when convoluted with the Si (2 2 0) reflection and with another diamond (1 1 1) reflection, respectively, much lower than the theoretical perfect-crystal value of 82%. The as-

*Corresponding author. Tel.: +33-476-88-2040; fax: +33-476-88-2542.

E-mail address: freund@esrf.fr (A.K. Freund).

0168-9002/01/$ - see front matter
PII: S0168-9002(01)00331-X

cleaved crystals were 1.2 mm thick and showed strains from the cleavage as clearly seen in the topograph (Fig. 1a). Ongoing work at Spring-8 on (1 0 0)-oriented crystals had shown that some type Ib samples had a mosaic spread between 5 and 15 arcsec, while type IIa crystals had about 2 arcsec wide rocking curves [4]. The (1 1 1)-oriented type IIa crystals tested at the Argonne National Laboratory had a mosaic spread of 2.5–4 arcsec for a beam size of 1–2 mm^2 [5].

In an effort to study the influence of nitrogen impurities on the defect structure, we applied X-ray excited optical luminescence (XEOL) and spatially resolved double-crystal diffractometry [6] as new techniques.

2. Experimental techniques and results

2.1. Optical characterisation

Fig. 1b shows a XEOL picture of the second (1 1 1)-oriented sample ($9.5 \times 12 \times 1.2\,mm^3$) exposed to the white synchrotron beam on beamline BM5 at the ESRF. It was cleaved from the same big stone and adjacent to the sample whose topograph is represented in Fig. 1a. Depending on nitrogen concentration, the colour changed from dark yellow to green and blue from regions with and without nitrogen impurities. The yellow lines correspond to light reflection from cleavage steps on the surface and cannot be associated with nitrogen.

The first sample was then thinned down to 0.42 mm. Figs. 1c and d are a polarised light micrograph and a close-up photograph, respectively. The first image reveals the overall strain pattern in the specimen. The topograph, the micrograph and the XEOL picture show very similar structural features that seem all to be closely related to the yellow nitrogen distribution seen in the photograph.

2.2. Double-crystal X-ray diffractometry

Double-crystal rocking curves of the thinned diamond plate were taken on the ESRF optics beamline with 10 keV X-rays. Downstream of the (1 1 1) double-crystal monochromator a symmetrical silicon (2 2 0) crystal was mounted on the first axis of the diffractometer. The diamond crystal was mounted on the second axis in almost non-dispersive setting. A Si-PIN diode ($10 \times 10\,mm^2$) fully collected the reflected beam. For beam footprints of 1×1, 1.8×2 and $3 \times 3\,mm^2$ the FWHM of the diamond rocking curve was 1.9 mdeg or 6.8 arcsec, i.e. 1.3 arcsec wider than the theoretical width of 5.5 arcsec. The peak reflectivity recorded was $84 \pm 3\%$, the highest ever seen by us and in very close agreement with the theoretical value of 82%.

To further evaluate the micro-defect structure of the same diamond crystal, we performed spatially resolved X-ray diffractometry.

2.3. High resolution spatially resolved double-crystal diffractometry

We replaced the Si-PIN diode with a high-resolution area detector, namely the FReLoN (Fast-Readout-Low-Noise) 1024×1024 pixel CCD camera developed at the ESRF [7]. With the 10 μm optics the field was $10 \times 10\,mm^2$, the distance from the diamond crystal was 0.5 m. Since the camera was fixed the scans were ω-scans and represented the distribution of lattice tilts. A software package [6] was used for calculating the rocking curve parameters such as width, peak reflectivity and peak position for each pixel. In this way a tremendous gain of five orders of magnitude in measuring time was obtained with respect to the serial rocking curve mapping technique [3].

A rocking curve with the fully illuminated diamond was taken with 10 s of exposure for each of the 40 points of the rocking curve. The result of the rocking curve width is shown in Fig. 1e. The picture was recorded at a Bragg angle of 17.52° and then stretched using the computer. The structure seen is similar to that of the previous pictures and related to the defect structure consisting mainly of growth bands. The tilts or lattice orientation changes caused by growth bands have rotation axes that run parallel to the six crystal edges. Therefore maximum tilt angles were observed for the two lower and the two upper growth sectors, whereas no contrast was seen for

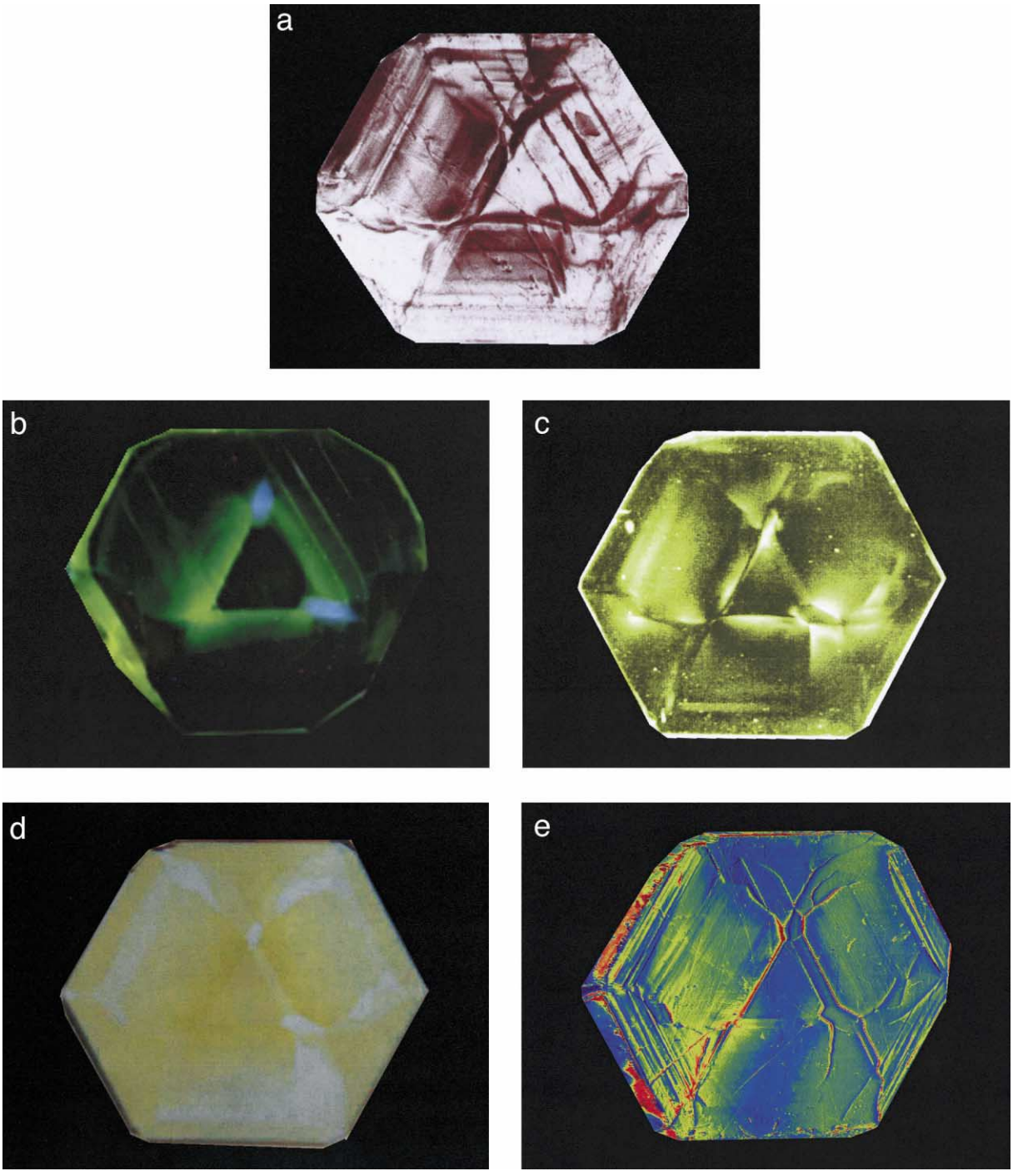

Fig. 1. Results of diamond characterisation, size: $10 \times 12\,mm^2$. (a) X-ray white beam topograph of an as-cleaved diamond crystal. (b) XEOL picture of a sample adjacent to the one shown in (a). (c) Polarised light optical micrograph of the sample thinned down to 0.42 mm. (d) Close-up photograph. (e) Rocking curve FWHM contour maps of the thinned diamond recorded with the FReLoN camera; colours (mdeg): blue: 1.25, green: 1.5, yellow: 1.75, orange: 2, red: 2.25.

the left and right growth sectors. Here and in the crystal centre a minimum rocking curve width of about 1.25 mdeg or 4.5 arcsec was measured. On average the rocking curve width was about 1.6 mdeg.

The measured minimum rocking curve width (blue colour) corresponded exactly to the theoretically perfect crystal width given by the spectral distribution of the beam impinging on the diamond crystal, convoluted with the angular resolution of the recording device: the perfect crystal rocking curve width was calculated as 1.27 mdeg, in excellent agreement with the observed width of 1.25 mdeg as derived from Fig. 1e. Then the average mosaic spread was the observed, average rocking curve width of 1.6 mdeg deconvoluted with this width giving 1 mdeg, again in excellent agreement with the value of 1.1 mdeg derived from the integrated rocking curve measurement. More experimental results will be published elsewhere [8].

3. Conclusions

The X-ray diffraction studies using a 2D high-resolution camera have contributed a wealth of new details on the crystalline quality of synthetic diamond crystals and allowed us to obtain a very substantial gain in time. Although there is a strong correlation between nitrogen concentration and lattice deformation it is still unclear whether a non-uniform incorporation of substitutional nitrogen atoms was at the origin of the growth defects.

Acknowledgements

The authors thank Mr. R. Hustache for assistance at the experiments on BM5 and Ms. A. Rommeveaux for helping with image processing.

References

[1] A.K. Freund, Opt. Eng. 34 (1995) 432.
[2] A.K. Freund, J.P.F. Sellschop, K. Lieb, S. Rony, C. Schulze, L. Schroeder, J. Teyssier, SPIE Proc. 3448 (1998) 53.
[3] A.K. Freund, J. Hoszowska, J.P.F. Sellschop, R. Burns, M. Rebak, AIP Proc. CP521 (2000) 242.
[4] N. Toda, H. Sumiya, S. Satoh, T. Ishikawa, SPIE Proc. 3151 (1997) 329.
[5] P.B. Fernandez, W.K. Lee, D.M. Mills, G. Tajiri, L. Assonfid, Nucl. Instr. and Meth. A 459 (2001) 347.
[6] D. Lübbert, T. Baumbach, J. Härtwig, E. Boller, E. Pernot, Nucl. Instr.and Methods B 160 (2000) 521.
[7] J.-C. Labiche, J. Segura-Puchades, D. Van Brussel, J.-P. Moy, ESRF Newslett. 25 (1996) 41.
[8] J. Hoszowska, A.K. Freund, E. Boller, J.P.F. Sellschop, G. Level, J. Härtwig, R.C. Burns, M. Rebak, J. Baruchel, Proceedings of the Conference "XTOP 2000", Ustron-Jaszowiec, 13–15 September 2000.