

X-ray Phase-contrast Imaging Study of Voids and Fibres in a Polymer Matrix

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Abstract

We report X-ray phase-contrast images of voids and glass fibres in a polymer matrix. The phase-contrast images are obtained using both a monolithic 3-reflection asymmetric channel-cut monochromator and analyser with 1.54 Å X-rays. The mechanisms of contrast formation in the images of voids and glass fibres are discussed in detail and numerical simulations of the images are found to agree with the experiments.

1. Introduction

Phase gradients are produced in an X-ray beam when it passes through a medium containing refractive index variations. The phase gradients perpendicular to the direction of propagation of the X-ray beam can be resolved using an analyser of high angular sensitivity. The method of image formation by resolving these phase gradients has been called X-ray phase-contrast imaging (Davis *et al.* 1995a). Recently substantial image contrast in weakly absorbing materials has been reported using X-ray phase-contrast imaging (Nikulin 1990; Beliaevskaya *et al.* 1991; Semenov *et al.* 1991; Wilkins 1993a, 1993b; Ingal *et al.* 1994; Davis *et al.* 1995a, 1995b). Here we report the X-ray phase-contrast imaging of voids and a glass fibre in a polymer matrix, using the combination of a channel-cut asymmetric monochromator and analyser. The results are discussed in detail and compared with numerical simulations of the images.

2. Experimental Geometry

The schematic representation of the experimental geometry is shown in Fig. 1. A Kevex microfocus X-ray source (with nominal 10 µm diameter source size) was used with a monochromator and an analyser in a double-axis X-ray diffractometer. We used a monochromator and analyser employing successive asymmetric Bragg reflections for obtaining nearly parallel X-ray beams (see e.g. Kohra and Kikuta 1968; Matsushita *et al.* 1971). In our experiment, the monochromator and analyser crystals were of similar design. The only difference was that the analyser was larger to accommodate the expanded X-ray beam from the monochromator. The monochromator and analyser consisted of monolithic channel-cut silicon crystals designed for three successive 422 asymmetric Bragg reflections. The asymmetry factor for a given reflection is defined as

$$b = \sin(\theta - \gamma) / \sin(\theta + \gamma), \quad (1)$$

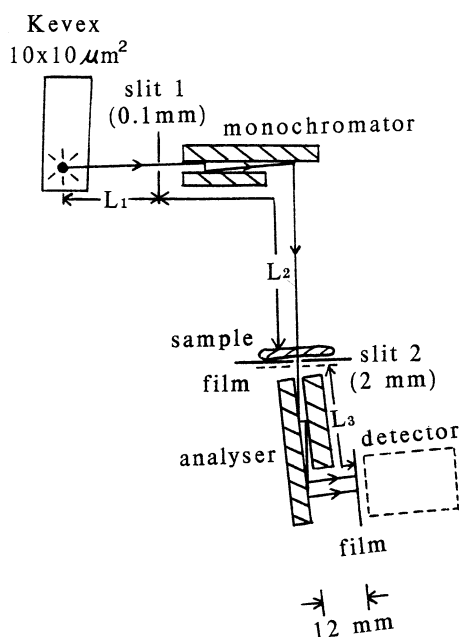


Fig. 1. Schematic representation of the experimental geometry. In the present experimental setting, the distance L_1 between the source and slit 1 is 80 mm; the (average X-ray path) distance L_2 between slit 1 and the sample is 230 mm; and the (average X-ray path) distance L_3 between the sample and the film is 40 mm.

where θ is the Bragg angle; γ is the angle between the Bragg planes and the crystal surface. The asymmetry factors for the three successive 422 Bragg reflections for both the monochromator and the analyser are $b_1 = 0.0625$, $b_2 = 16$ and $b_3 = 0.0625$. In Fig. 1 the width of slit 1 is approximately 0.1 mm and the width of slit 2 is 2 mm. In the present work, Cu $K\alpha_1$ radiation was used for the X-ray reflection because it offers the possibility of high spatial resolution and is also a convenient standard in the laboratory. Kodak dental film was used for recording the images. In this report, we refer to a conventional 'radiograph' when the film is placed at the entry to the analyser, approximately 2 mm from the sample, while on the other hand 'phase-contrast images' are obtained with the film placed approximately 12 mm after the third face of the analyser and perpendicular to the beam.

3. Experiments and Results

The sample was made by mixing an acetone-based glue ('Tarzan's Grip') on a sheet of plastic film which was removed after the glue had dried. Two air bubbles were trapped in the glue as shown in Fig. 2a. The thickness of the sample is approximately $300 \mu\text{m}$. The diameters (in the direction perpendicular to the incident X-ray beam) of the bubbles were measured using an optical microscope. One of the bubbles was found to have a diameter of $800 \mu\text{m}$ and the other $960 \mu\text{m}$. The air bubbles are approximately $200 \mu\text{m}$ thick, in the direction parallel to the X-ray beam. A glass fibre of diameter $13 \mu\text{m}$ was laid over the top of the glue surface.

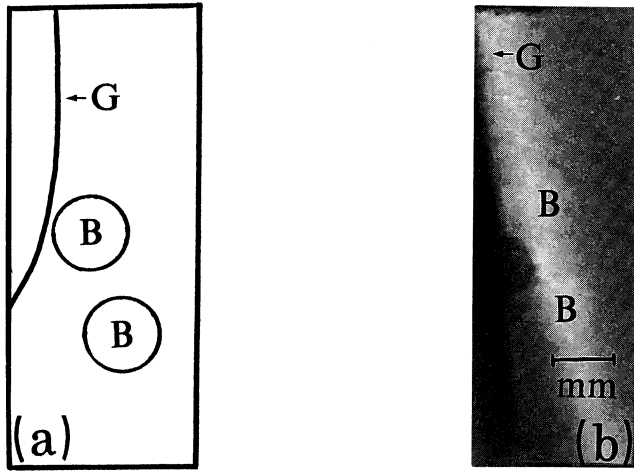


Fig. 2. (a) Schematic representation of two air bubbles (with a glass fibre) as the sample. (b) Conventional radiograph of the sample taken with $\text{Cu K}\alpha_1$ radiation, in which the air bubbles are indicated by B and the glass fibre is indicated by G.

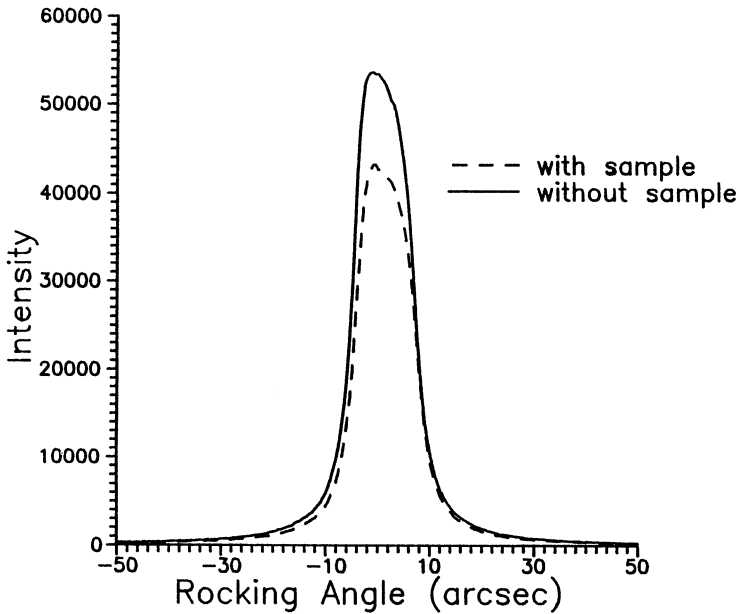


Fig. 3. Rocking curves of the reflection off the analyser with and without the air bubble sample.

A schematic representation of the sample and its radiograph are shown in Fig. 2. Some very weak absorption (positive) contrast of the voids and weak negative contrast of the glass fibre are seen in Fig. 2b. In the present work, positive (negative) contrast refers to an excess (reduction) of the diffracted

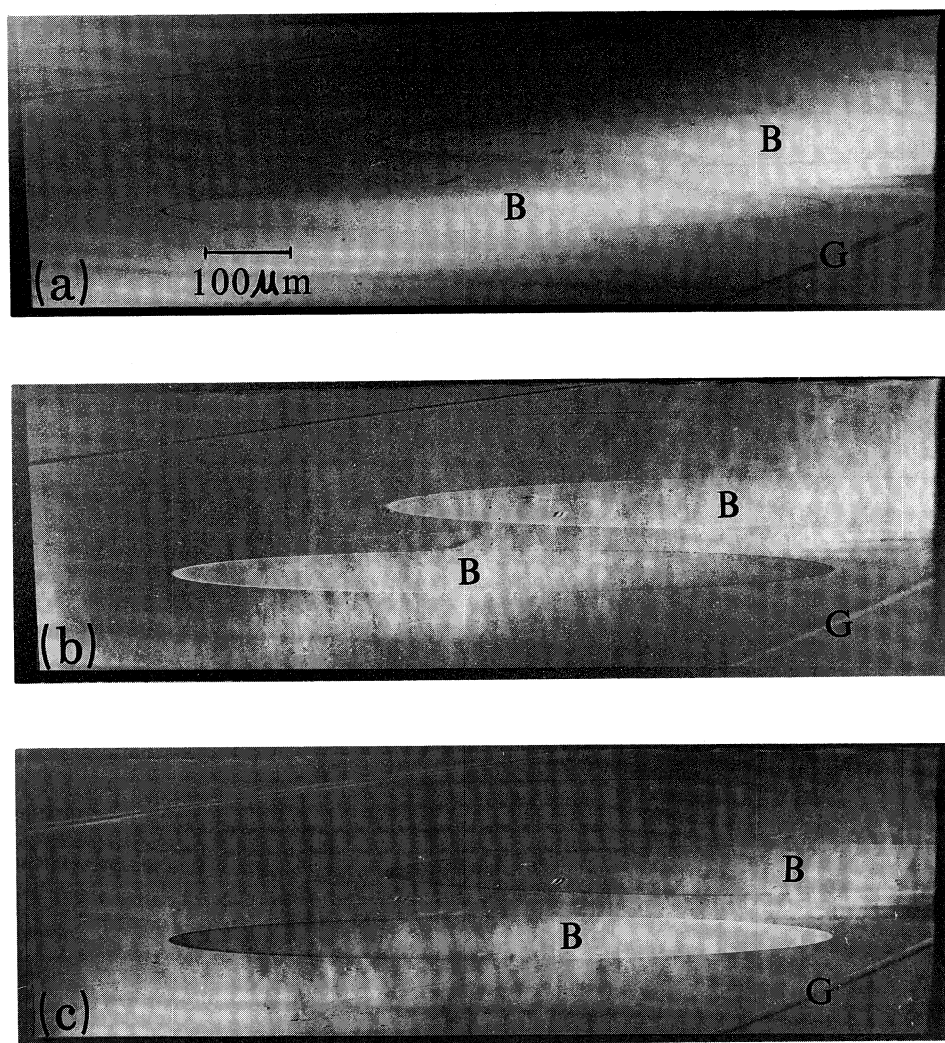


Fig. 4. With $\text{Cu K}\alpha_1$ radiation, phase-contrast images are shown of the air bubble sample taken at (a) the peak position ($0''$) of the rocking curve of the reflection of the analyser; (b) the positive shoulder ($+6''$) of the rocking curve of the reflection of the analyser; and (c) the negative shoulder ($-6''$) of the rocking curve of the reflection of the analyser. Here B denotes a bubble and G a glass fibre.

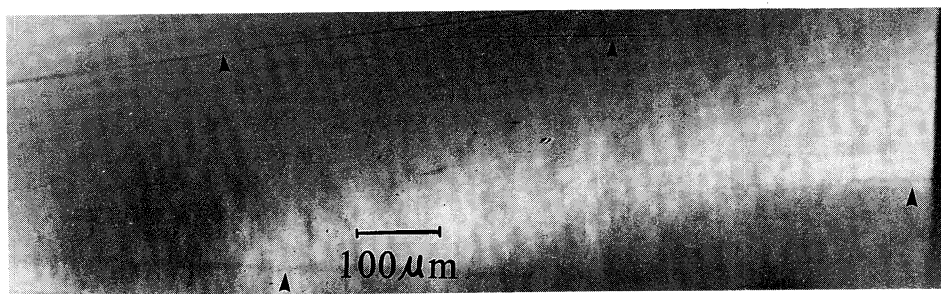


Fig. 5. Topographic image of the monochromator and the analyser without the sample, which shows the background of the phase-contrast images in Fig. 4a.

intensity relative to the perfect crystal background (Lang 1978*a*). That is, a white region in an image means positive contrast, while black means negative contrast. The rocking curves of the reflection off the analyser both with and without the sample were taken (see Fig. 3). The rocking curves show that the sample is a weakly absorbing material for hard X-rays (approximately 80% transmission with Cu $K\alpha_1$ radiation). The FWHM of the rocking curve of the reflection of the analyser without the sample is 12.3 ± 0.1 arcsec, while the FWHM with the sample is nearly same, i.e. 12.5 ± 0.1 arcsec. This implies that the overall divergence of the X-ray beam changes very little when the beam is transmitted through the sample.

The phase-contrast images were taken at the Bragg peak ($0''$ offset corresponding to 'bright field') and off-the-peak ($+6''$, $-6''$ corresponding to 'dark field') orientations of the analyser (see Figs 4*a*–4*c*). It should be noted that the order of the bubbles and fibre in Fig. 4 is the mirror image of that in Fig. 2 due to the experimental geometry in recording the phase-contrast images, and also that the features in Fig. 4 are magnified by a factor of about 15 in the horizontal direction due to the asymmetric (beam-expanding) analyser crystal. It is apparent that, in the case where the film is placed after the analyser, very strong and sharp positive (white) and negative (black) contrast of both the voids and the glass fibre is formed. We notice that when the phase-contrast images are taken at the peak position of the reflection off the analyser (see Fig. 4*a*), both the left and the right boundaries of the bubble images and the glass fibre show black contrast; when the phase-contrast images are taken at the positive shoulder of the rocking curve of the reflection of the analyser (see Fig. 4*b*), the left side of the void image shows white contrast and the right side shows black contrast. The contrast of the glass fibre image is reversed when compared with that of the voids. When the phase-contrast images are taken at the negative shoulder of the reflection of the analyser (see Fig. 4*c*), the left side of the void image shows black contrast and the right side shows white contrast. Again the contrast of the image of the glass fibre is the reverse of that for the bubble.

Fig. 5 shows the topographic image of the reflection off the monochromator and the analyser without the sample and at the exact matching Bragg condition, which is the background image corresponding to Fig. 4*a*. By combining these observations with those for other settings of the analyser (not shown here), two types of crystal defect images can be identified from Fig. 5. One type (indicated by the arrows) originates from the monochromator and the other from the analyser.

4. Discussion

Short wavelength X-rays passing through a non-absorbing medium suffer a phase change $\Delta\phi$ related to the dielectric susceptibility χ of the medium (Davis 1995)

$$\Delta\phi = \frac{\pi}{\lambda} \int \chi(s) ds, \quad (2)$$

where λ is the wavelength and s measures the distance in the dielectric medium along the path of the X-ray beam. The gradient of this phase is related to the

gradient in the electron density in the medium. A phase gradient transverse to the incident X-ray propagation direction represents a change in the direction of the transmitted beam. This is equivalent to refraction of the X-rays in the medium (Davis 1994). The relation between the angle of incidence and the angle of refraction across an air–solid interface is given by Snell’s law (Davis 1994; Wilson 1962), which is approximately

$$\sin\alpha = (1 - \delta)\sin\beta, \quad (3)$$

where $1 - \delta$ is the refractive index, and $-\delta = \Delta\chi$ is the difference in the dielectric susceptibilities across the interface between two uniform media. Here we observe that X-ray phase variations arise from changes in the refractive index. Usually δ amounts to only a few parts per million.

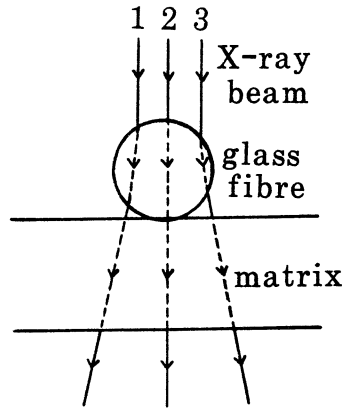


Fig. 6. Schematic explanation of the formation of contrast for phase-contrast imaging of the glass fibre on the polymer matrix.

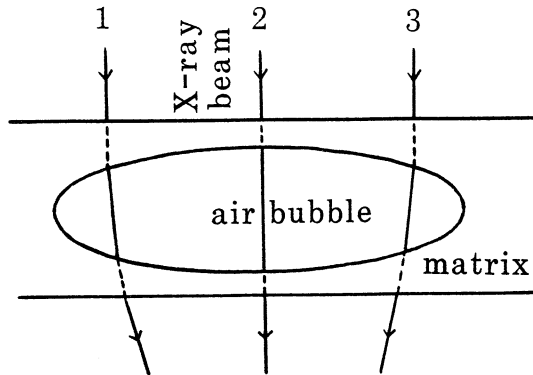


Fig. 7. Schematic explanation of the formation of contrast for phase-contrast imaging of an air bubble in the polymer matrix.

In order to help understand the contrast features in the images, schematic representations of the effect of refraction are shown in Figs 6 and 7. First we consider the phase-contrast imaging of the glass fibre. To help illustrate the principle of contrast formation in the images, we consider three beams of X-rays

incident on the top surface of the glass fibre, as shown in Fig. 6. When phase-contrast images are recorded at the peak position of the reflection of the analyser, only beam 2 satisfies the Bragg condition and is diffracted by the analyser. The beams 1 and 3 do not satisfy the Bragg condition due to the effect of refraction by the glass fibre. This is observed in the phase-contrast image of the glass fibre (Fig. 4a), in which a narrow positive line is embedded in wide negative strips. When a phase-contrast image is taken at the positive shoulder of the reflection of the analyser, beam 1 is further from the Bragg diffraction condition but beam 3 is closer and is diffracted. Therefore, the negative-positive (black-white) images of the glass fibre can be seen (see Fig. 4b). A similar situation occurs on the negative shoulder of the reflection of the analyser and the positive-negative (white-black) image can be seen (see Fig. 4c).

Next we consider the contrast arising from air bubbles in the sample. A similar assumption for the X-ray beams is applied to the voids (see Fig. 7). We notice that the directions of the refraction of the X-ray beams from the air bubbles are opposite to those from the glass fibre. Therefore, the contrast effect is reversed in the images of the air bubbles, compared with that of the glass fibre (see Figs 4a-4c). A very sharp, strong contrast image is seen along the boundaries of the air bubbles and the polymer matrix. This is due to the effective focusing action of the bubbles and to the relatively small radius of curvature at the extrema of the bubbles which tend to strongly refract the incident beam, compared with the refraction near the centre of the bubbles. Therefore, the diffraction intensity along the boundaries recorded by the analyser increases (or decreases) sharply. Numerical simulations were performed to calculate the images formed by the voids and the glass fibre. Both refraction and absorption were taken into account by using complex dielectric susceptibilities. The data used in the simulation are given in Table 1.

Table 1. Data used in the numerical simulation

Material	Real χ	Im χ	Thickness (μm)
Glue	-7.9×10^{-6}	-1.5×10^{-8}	300
Fibre	-2.1×10^{-5}	-4.0×10^{-7}	13
Bubble	0	0	200

The X-rays passing through the 800 μm diameter bubble are spread over 12 mm on the analyser surface as a result of the asymmetry. The incident X-ray beam was assumed to be a monochromatic plane-wave and the phase change in the beam transmitted through the sample was calculated from (2). The effect of the 3-reflection asymmetric analyser crystal was calculated using a Fourier transform method (Davis 1995) which relates the amplitude profile of the diffracted X-ray beam at the surface of the crystal to the incident profile and the crystal rocking curve. The numerical simulations of the phase-contrast images are shown in Figs 8a-8c which correspond to Figs 4a-4c. The numerical simulations of the images are consistent with the results of the experiments and confirm the general principle of image formation by the phase-contrast mechanism.

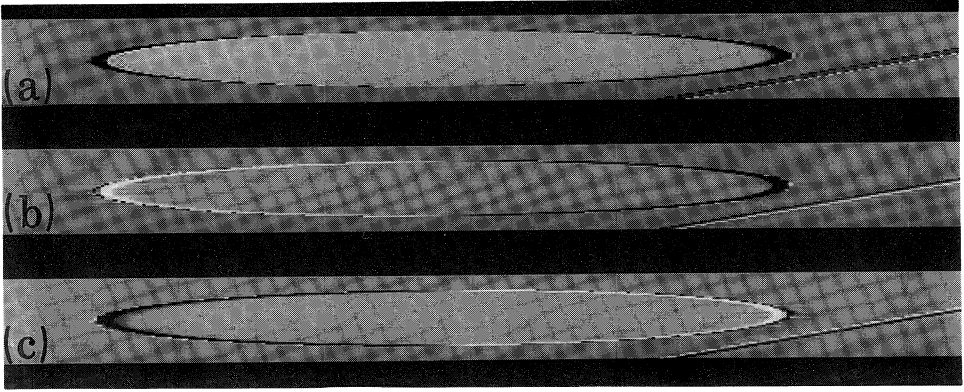


Fig. 8. Numerical simulations of phase-contrast imaging for an air bubble and glass fibre on the polymer matrix. Figs 8a–8c correspond to Figs 4a–4c respectively.

Next we discuss the resolution of phase-contrast imaging. Similar to spatial resolution in X-ray topography (Yoshimatsu and Kohra 1960; Austerman and Newkirk 1968; Lang 1978*b*), spatial resolution of phase-contrast imaging is determined by the size of the image of a point on the sample. In the vertical plane (normal to the plane of diffraction), the spatial resolution of an image is determined by

$$A_v = L_3 \frac{F_v}{L_1 + L_2}, \quad (4)$$

where F_v is the vertical dimension of the X-ray focal spot used and L_1 , L_2 , L_3 are defined in Fig. 1. For our experiment, the vertical spatial resolution of the image is approximately $1.3 \mu\text{m}$. Several factors may affect the horizontal spatial resolution of the image. In the present geometry, the effect of dispersion (i.e. $\Delta\lambda/\lambda$) is negligible due to the 422 monochromator and analyser which operate non-dispersively. The theoretical divergence of the X-ray beam for a given λ is 0.026 arcsec in the present experimental setting, which is negligible compared with other factors. Therefore, the horizontal spatial resolution of the image is mainly determined by the resulting angular width of the reflection of the analyser. So the horizontal spatial resolution of the image is given by

$$A_h = L_3 \Delta\theta_r, \quad (5)$$

where $\Delta\theta_r$ is the angular width of the rocking curve of the analyser. The horizontal spatial resolution for phase-contrast imaging in the present configuration is calculated to be approximately $2.4 \mu\text{m}$. We should point out that the spatial resolution of the topographs originating from the monochromator may be worse than $6 \mu\text{m}$ in both vertical and horizontal planes, while those originating from the analyser may be better than $1 \mu\text{m}$, due to the effective value of the distance L_3 .

5. Remarks

Significant phase-contrast imaging of voids and fibres in a polymer matrix was studied using a microfocus source and a combined system of monolithic 3-reflection asymmetric channel-cut Si 422 crystals as X-ray monochromator and analyser. Numerical simulations of the images are generally in good agreement with the results of the experiments. The contrast and the resolution of the images were discussed in detail and clear evidence was obtained for the observed contrast as arising via the phase-contrast mechanism. In order to improve the resolution of the phase-contrast imaging technique, more highly coherent X-ray sources and better quality crystal optics are desirable.

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