Direct Observations of Cracks and Voids in Structural Materials by X-ray Imaging Using Ultra-bright Synchrotron Radiation

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Refraction contrast X-ray imaging experiments were conducted on acrylic resin with an artificial cylindrical hole, A7075 aluminum alloy, A6063 aluminum castings, mild steel with cracks or voids, and low alloy steel with inclusions, using a ultra-bright synchrotron radiation X-ray beam in BL24XU hutch C of SPring-8. Conventional absorption contrast X-ray imaging experiments were also done for the comparison. The X-ray beam was controlled to be monochromatic by Si double-crystals and collimated by a slit. The distance between the sample and the detector was changed from 0 to 3 m, and the X-ray energy was 15 to 25 keV. Photographs were taken by X-ray film and/or X-ray CCD camera. As a result, the refraction imaging method gave a much more distinct image of the artificial cylindrical hole in acrylic resin as compared with the absorption method. The fatigue cracks in aluminum alloy and mild steel were also distinctly observed. The X-ray imaging revealed the presence of MnS nonmetallic inclusions in low alloy steel. Void defects in aluminum castings were clearly detected by the imaging. In addition, in-situ observation of tensile fracture of aluminum alloys using a high resolution X-ray CCD camera system was successfully conducted. The observations by use of asymmetric reflection technique for X-ray imaging experiment were also well performed. From above, the X-ray imaging method using ultra-bright synchrotron radiation is concluded to be very useful for fracture research of materials.

KEY WORDS: synchrotron radiation; X-ray diagnosis; structural materials; fracture; refraction imaging.

1. Introduction

In recent years, weight reduction has been an increasingly important requirement in many fields, from skyscrapers, long-span bridges, and other large items of infrastructure, to trains, ships, cars, and other vehicles, and to electronics and information systems. One practical approach to reduce weight is to increase the strength of their structural materials. When making the materials stronger, however, the susceptibility to environmental assisted cracking such as delayed fracture, corrosion fatigue, and so on considerably increases. That is why, numerous studies on environmental assisted cracking have been carried out so far. Herein, non-destructive diagnosis methods are required to investigate directly such fracture phenomena in the matrix in connection with diffusive hydrogen, stress and strain states, microstructure of materials, and so on.

X-ray radiography is thought to be useful as one of non-destructive diagnosis methods. It is well known that the image contrast in conventional X-ray radiography results from differences in absorption, that is, differences in density, thickness and/or elemental composition of a sample. In this sense, conventional X-ray radiography is often called ‘absorption contrast’ imaging.

On the other hand, much attention has been recently devoted to synchrotron radiation, which is emitted from an electron traveling at close to the speed of light when its path is bent by a magnetic field, as an ideal artificial X-ray source for the X-ray radiography. This is because the emitted X-ray beam is bright, directional, stable, and its energy is arbitrarily controllable and selectable for the experiment. Recently, what is called, the third-generation large-scale synchrotron radiation facilities, which are regarded as the facility having more than 5 GeV electron energy, have also been operated. There are three third-generation large-scale synchrotron radiation facilities in the world, that is, SPring-8 (Super Photon ring, 8 GeV, Japan), APS (Advanced Photon Source, USA) and ESRF (European Synchrotron Radiation Facility, France). For example, SPring-8, which is the largest and brightest, produces radiation that is about one billion times more brilliant than conventional X-ray sources. It is thought that such ultra-bright synchrotron radiation source could provide us, in principle, completely...
different information from conventional methods such as absorption contrast X-ray imaging, because much highly directional property is also available. That is why, based on the fact that the phase shift cross section for light element is almost a thousand times larger than the absorption one, especially in a hard X-ray region, new X-ray imaging techniques such as refraction method, diffraction method, inter-ferometric method have recently been studied and proposed actively.

The principle of refraction contrast X-ray imaging is shown in Fig. 2, as compared with conventional absorption X-ray imaging, illustrating the X-ray intensity profiles of the absorption contrast and refraction contrast for a void in material. In the conventional absorption method, the X-ray beam passing through the void is less absorbed, and the intensity pattern of the void area is simply stronger due to increased density of the X-ray transmission beam. On the other hand, the refraction contrast imaging, which is obtained by setting the sample some distance apart from the detector, forms a very weak line along the X-ray absorbed region (strong intensity region) due to the distribution of X-ray direct beam differentiated from the refraction effect. As a result, this method provides a higher resolution image than the conventional absorption method. It is thought that these refraction contrast X-ray imaging can be well performed only in the third-generation large-scale synchrotron radiation facilities, that is, SPring-8, ESRF and APS. This is because the refraction effect needs X-ray beam with not only brilliant property but also highly directional property obtained from these ultra-bright synchrotron radiation source, according to the principle mentioned above.

So, such phase contrast imaging has been intensively and successfully applied as a high resolution medical diagnosis method to observe the internal structure of biological materials such as the respiratory organ, cancers, capillaries, blood vessel in tumors, and so on. For example, in SPring-8, efforts are being made in the field of medical diagnosis, and the development of cancer diagnosis is being actively carried out at the experimental station of BL20B2. These observations utilize a high spatial resolution of 10 μm imaging method, confirming that the accurate diagnosis of tumors using synchrotron radiation is possible. A real-time observation system of blood flows in small blood vessels is also under development in SPring-8. On the other hand, Buffiere et al. have studied the evolution of damage in an Al/SiC composite material during monotonic tensile tests, using high resolution X-ray imaging techniques in ESRF. In addition, in-situ observation of foaming behavior of an aluminum foam sandwich structure for lightweight application, etc. have also been attempted in ESRF, indicating that X-ray imaging techniques using ultra-bright synchrotron radiation source is useful for the studies on fracture and formation of materials. However, such application to structural material studies including fracture phenomena in the matrix has not much carried out whereas refraction contrast imaging is thought to be very promising as a unique non-destructive diagnosis method.

The present study has been conducted to compare absorption images with refraction images by changing the distance between samples and detector, and then, to observe cracks and voids in selected structural materials in connection with microstructure by refraction contrast imaging, using ultra-bright synchrotron radiation by SPring-8. In addition, a high resolution X-ray imaging has been attempted.
to observe cracks and voids in the materials using an asymmetric reflection technique. Furthermore, in-situ observation has also been tried to observe directly crack initiation and tensile fracture of materials by high resolution X-ray imaging using SPIn-8.

2. Experimental

Acrylic resin (20 mm in thickness) with an artificial cylindrical hole, A7075 aluminum alloy (0.5 to 2 mm in thickness) with a fatigue crack, mild steel (0.5 mm in thickness) with a fatigue crack, 0.35C–0.25Si–0.6Mn–0.018S–1.5Ni–1.5Cr–0.2Mo forged steel (0.3 to 0.5 mm in thickness) with MnS non-metallic inclusions, and A6063 aluminum castings (1 to 2 mm in thickness) with porosity were used as materials for the X-ray imaging experiments. The artificial cylindrical hole of acrylic resin was machined with a cutting drill to have two internal diameters with 1.5 and 3.0 mm. The fatigue cracks of A7075 aluminum alloy and mild steel were both introduced from a slit with 0.5 mm in width of compact tension specimen by cyclic loading using a hydraulic universal testing machine. After fatigue test, A7075 aluminum alloy and mild steel were mechanically cut to 2 mm in thickness and 0.5 mm in thickness with a slicing machine.

The experiment of X-ray imaging was performed at the experimental hutch C of BL24XU (Hyogo-BL) 24 of SPIn-8, which has the electron energy of 8 GeV and the brilliance of $1 \times 10^{19}$ photon/s/mm$^2$. In the BL24XU, the X-ray beam was first collimated with a four-quadrant slit with an aperture size of 1 mm x 1 mm installed in the front end at a distance of 30 m from the undulator radiation source. The X-ray beam was then controlled to be monochromatic by silicon double-crystals with (111) symmetric reflection. The X-ray beam was then expanded by using two types of asymmetric reflection optical systems as shown in Figs. 3(a) and 3(b). The asymmetric factor $b$ is defined as:

$$ b = \frac{\sin(\theta_B - \alpha)}{\sin(\theta_B + \alpha)} $$

where $\theta_B$ is the Bragg angle and $\alpha$ is the inclined angle between the diffraction plane and the crystal surface, respectively. 25,26 Since the crystal surface and diffraction plane chosen were (100) and (511), respectively, the asymmetry factor $b$ was 0.207. 13,24 Finally, the beam was vertically expanded in the same way as in the horizontal case. The total factor of magnification in each direction, $1/b^2$, was about 23. 13 Therefore, the enlargement of observing area and the high resolution observation were obtained from setting (a) and (b) in Fig. 3, by making use of the asymmetric reflection in both horizontal and vertical directions. 13,21 The X-ray energy was set to 15 keV for acrylic resin and aluminum alloy samples, and to 25 keV for steel samples. Photographs of the images were taken by an X-ray film and/or X-ray CCD camera as an image detector. Refraction contrast images were obtained by setting a 3 m gap between the sample and the image detector. In the case of absorption contrast imaging, the sample was placed just in front of the image detector.

On the other hand, the facility system with a tensile test machine and an imaging stage for in-situ observation of crack initiation and propagation in structural materials were specially designed and constructed in the experimental hutch C of BL24XU (Hyogo BL) of SPIn-8. High resolution X-ray CCD camera with a pixel size of 6.45 mm x 6.45 mm and exposure time from 0.000134 to 10 s were also specially prepared. Using these facilities, in-situ observation was tried to directly investigate the crack initiation and tensile fracture of aluminum alloys by the high resolution X-ray imaging.

3. Results and Discussion

3.1. Observation of an Artificial Cylindrical Hole in Acrylic Resin

Figures 4(a) and 4(b) present X-ray absorption contrast image and X-ray refraction contrast image, respectively, of an acrylic resin (20 mm in thickness) with an artificial cylindrical hole of 3.0 and 1.5 mm in internal diameter. Both X-ray imaging tests were conducted in the experimental hutch C of BL24XU of SPIn-8, using setting (a) in Fig. 3. The absorption contrast image was obtained by setting the X-ray detector just behind the sample. The refrac-

![Fig. 3. Schematics of two optical systems by asymmetric reflection Bragg reflection for X-ray imaging experiment at BL24XU hutch C of SPIn-8. (a) System for enlargement of observation area. (b) System for high resolution observation.](591)
tion contrast image was obtained by setting a 3 m gap between them. The X-ray energy was 15 keV and the photographs were taken by X-ray film. It was confirmed that the refraction contrast image is much more distinct, especially at the edge of the cylindrical hole, than the absorption contrast image. A pockmarked pattern on the internal-surface of the artificial cylindrical hole, presumably formed at the time of drill hole cutting, is also clearly seen only in the refraction image.

Figure 5 shows X-ray images of the acrylic resin with an artificial cylindrical hole containing water as a function of distance from 0 to 3 m between the sample and the X-ray detector. With the increase in the distance, the contrast of the X-ray image was improved, indicating that the refraction contrast image is better than the absorption contrast image. Especially in the case of the distance of 3 m between them, the X-ray image clearly detected not only the interface boundary between water and acrylic resin, but also small air bubbles with decade μm size in water. These facts also confirm that the distance between the samples and the X-ray image detector is one of the key controlling parameters in the X-ray refraction contrast imaging method.

3.2. Observation of Fatigue Cracks in Aluminum Alloy, Mild Steel and Titanium Alloy

X-ray imaging experiments using setting (a) in Fig. 3 were conducted for detecting fatigue cracks in A7075 aluminum alloy (2 mm in thickness) and mild steel (0.5 mm in thickness). The fatigue cracks were both preliminary introduced from the notched slit before the imaging tests. These observed samples were schematically shown in Fig. 6. X-ray images of fatigue crack initiated from notched slit of A7075 aluminum alloy are shown in Fig. 7, in which (a) is the absorption contrast X-ray image and (b) the refraction contrast X-ray image. The absorption contrast image was obtained by setting the X-ray detector just behind the sample, whereas the refraction contrast image was obtained by setting a 3 m gap between them. The X-ray energy was 15 keV for both cases and the photographs were taken by X-ray film. It is obvious that the refraction contrast image of the fatigue crack seen in Fig. 7(b) was thick and dark, while the absorption contrast image in Fig. 7(a) was thin and bright, confirming that the former image is much more distinct than the latter. The refraction contrast image also unveils the fatigue crack branching at the tip. Figure 8 shows the refraction contrast X-ray images of the fatigue

![Fig. 4](image1)

**Fig. 4.** X-ray images of acrylic resin (20 mm in thickness) with a cylindrical artificial hole. (a) Absorption contrast image, in which distance between sample and X-ray detector was adjusted to be 0.05 m. (b) Refraction contrast image, in which distance between sample and X-ray detector was adjusted to be 3.0 m.

![Fig. 5](image2)

**Fig. 5.** X-ray images of acrylic resin with artificial cylindrical hole containing water as a function of distance between sample and X-ray detector.

![Fig. 6](image3)

**Fig. 6.** Schematics of observation samples of aluminum alloy (Fig. 7) and mild steel (Fig. 8) with fatigue crack for X-ray imaging experiments at BL24XU hutch C of SPring-8.
crack in mild steel. The refraction contrast image was obtained by setting a 3 m gap between the sample and the X-ray detector. The X-ray energy was 25 keV in this case and the photograph was taken by X-ray film. It is obvious that the fatigue crack is clearly detected with \( \mu m \) resolution as a dark contrast image. It is thought that the bright contrast band in the vicinity of the dark contrast crack image originates in the leakage of synchrotron radiation X-ray beam through the fatigue crack.

On the other hand, Fig. 9 compares X-ray images of the fatigue crack in Ti–6Al–4V alloy (1 mm in thickness) for settings (a) and (b), each corresponding to the experimental setting conditions (a) and (b) in Fig. 3. So, both setting (a) and (b) in Fig. 9 should have enlargement of the observing area and high resolution image, by making use of the asymmetric reflection in both horizontal and vertical directions. The X-ray energy was 25 keV and the photographs were taken by X-ray film for both cases. It was confirmed that setting (a) provides enlargement of the observing area, say at least 5×5 mm square area, and setting (b) higher image resolution. In fact, the branched fatigue cracks are clearly seen at setting (b) by the enlargement of X-ray beam, while such cracks are seldom seen at setting (a).

3.3. Observation of MnS Nonmetallic Inclusions in Low Alloy Steel

Figure 10 shows MnS nonmetallic inclusions observed in the vicinity of the fracture surface of 0.35C–0.25Si–0.6Mn–0.018S–1.5Ni–1.5Cr–0.2Mo forged steel (0.5 mm in thickness). The image was obtained by the refraction contrast X-ray imaging experiment using setting (a) in Fig. 3. The X-ray energy was 25 keV and the photographs were taken by X-ray film. The refraction contrast X-ray imaging was applied to the steel after the tensile test by setting a 3 m gap between the sample and the X-ray film. It is obvious that non-metallic inclusions, which were confirmed to be MnS by means of EPMA, were clearly observed in steel. The dark contrast of the edge parts of the inclusion is thought to be refraction image due to the formation of micro-cracks between the inclusion and matrix.

On the other hand, Fig. 11 presents the X-ray images of MnS nonmetallic inclusion and voids in the low alloy steel (0.5 mm in thickness). Both images were obtained with the asymmetric reflection method using setting (b) in Fig. 3. As compared with the X-ray image in Fig. 10, the MnS nonmetallic inclusion seen in Fig. 11 is much large and more distinct with \( \mu m \) scale resolution owing to the enlargement of X-ray beam by asymmetric reflection method of setting (b) in Fig. 3. Voids in Fig. 11 are also well observed, indicating that voids coalesce with each other in the steel matrix just before fracture.

3.4. In-situ Observation of Tensile Fracture of Aluminum Alloys

Figure 12 shows the facility system with a tensile test machine and an imaging stage for in-situ observation of crack initiation and propagation in structural materials, which was specially designed and constructed in the experimental hutch C of BL24XU (Hyogo BL) of SPring-8. Using this facility and high resolution X-ray CCD camera, a couple of in-situ observation experiments were attempted to directly investigate crack initiation and tensile fracture of
A7075 aluminum alloys with a cylindrical hole-notch and A6063 aluminum castings with porosity (cavity voids).

Figure 13 shows the tensile test specimen with 0.5 mm in thickness of A7075 aluminum alloy for in-situ observation of tensile fracture. As seen in Fig. 13, the tensile test specimen has a cylindrical hole-notch with two sizes in the depth direction. The size of the shallow hole was 0.2 mm in diameter and 0.15 mm in depth, and that of the deep one 0.03 mm in diameter and 0.3 mm in depth. The refraction contrast image was obtained by setting a 1 m gap between the sample and the X-ray CCD camera. The X-ray energy was 15 keV in this case and the photographs were taken at every fifth of a second by the X-ray CCD camera. Figure 14 shows X-ray images of the fracture process in the vicinity of the cylindrical micro-notch of the tensile test specimen; (a) presents the image before tensile loading, (b) the image just before tensile fracture, and (c) the image at the moment of tensile fracture. It is obvious that the diameter of the cylindrical micro-notch of the image (b) is wider than that of the image (a), indicating that the test specimen was plastically deformed to the tensile direction by loading. In addition, the image (c) indicates the fracture crack propagation in the vertical direction against tensile loading from the bottom of the cylindrical micro-notch. The interpretation of wavy patterns seen around the crack in the image (c) must be further examined, though it seems to originate in

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Tear pieces on the fracture surface and/or heavy plastic deformation.

Figure 15 shows X-ray images of in-situ observation of tensile fracture of A6063 aluminum casting (2 mm in thickness) with porosity (cavity voids) before and after tensile fracture. The in-situ experiment was conducted using the tensile test facility. The X-ray energy was 15 keV and the photographs were taken by X-ray film in this case. The refraction contrast images were obtained by setting a 3 m gap between the sample and the X-ray film. It is obvious that the porosity (cavity voids) of 0.05 to 0.3 mm size are clearly observed in both images (a) and (b). It is also seen from the comparison between images (a) and (b) that tensile stress enlarges the size of the voids to the tensile direction. In addition, micro-cracks are observed in the vicinity of the cavity voids in the image (a), indicating that the crack initiates from the cavity voids and propagates almost to the vertical direction against tensile loading. Thus, in-situ observation of fracture process has also been successfully carried out in this study by X-ray imaging using SPring-8.

4. Conclusions

The refraction contrast X-ray imaging, which was performed by setting a few meters distance between the sam-
ple and X-ray detector in the experimental hutch C in BL24XU of SPring-8, was concluded as a very useful technique to sensitively diagnose cracks and voids in materials, as compared with conventional absorption method. Further high resolution was obtained by the use of asymmetric reflection technique for X-ray imaging. In fact, cracks and voids in aluminum alloys and mild steel, and MnS non-metallic inclusions in a low alloy steel were much more distinctly observed by using these X-ray imaging techniques. In-situ observation experiments of fracture process of aluminum alloys were also successfully conducted, by using a specially designed tensile test facility system and high resolution X-ray CCD camera.

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