A Quantitative Study of Hydride in Titanium by the Diffraction-enhanced X-Ray Imaging Method

The diffraction-enhanced X-ray imaging (DEI) method was applied to the quantitative study of hydride in titanium. The diffusivity and activation energy of hydrogen diffusion were determined by the direct observation of the hydride layer formed by an electrolytic-charge. Intensity profiles obtained from a model calculation were compared to those obtained from the experimental results, and the fitted diffusivity was determined by trial and error. The activation energy thus obtained, 0.54 eV, shows good agreement with the widely accepted value obtained by internal friction studies.

Considerable effort has been made towards investigating the interactions between hydrogen and metals for various applications, such as the development of hydrogen-storage materials. Although titanium and its alloys are promising materials for storing hydrogen, until now it has been impossible to visualize hydrides in titanium using non-destructive methods such as projection X-ray radiography. In the field of synchrotron radiation imaging, X-ray refraction-contrast imaging methods have been developed and used as a diagnostic tool in many disciplines, such as biology, the medical sciences and industrial applications since the early 1990's. However, only a few quantitative studies, such as the determination of physical constants have been reported. We have applied the X-ray diffraction-enhanced imaging (DEI) method to visualize hydride formed on a titanium surface by electrolytic-charge, and determined the activation energy of hydrogen diffusion in titanium-hydride.

Hydride was formed on a 99.99% pure α-titanium surface by electrolytic charging at various temperatures. A 1 mm thick slice of the titanium-hydride specimen was cut for cross-sectional observations at the vertical-wiggler beamline BL-14B. A schematic of the beamline and the experimental setup is shown in Fig. 1(a). The photographs were taken at two positions with small offset angles from the Bragg condition, on the low-angle and high-angle sides of the rocking curve. The X-ray energy of the incident beam was tuned to 30 keV using the beamline monochromator.

Figure 1(b, c) shows the cross-sectional profile of the specimen charged by the electrolytic method. The hydride layer is revealed as black or white lines parallel to the surface, and the contrast of the image is reversed between Fig. 1 (b) and (c) [1].

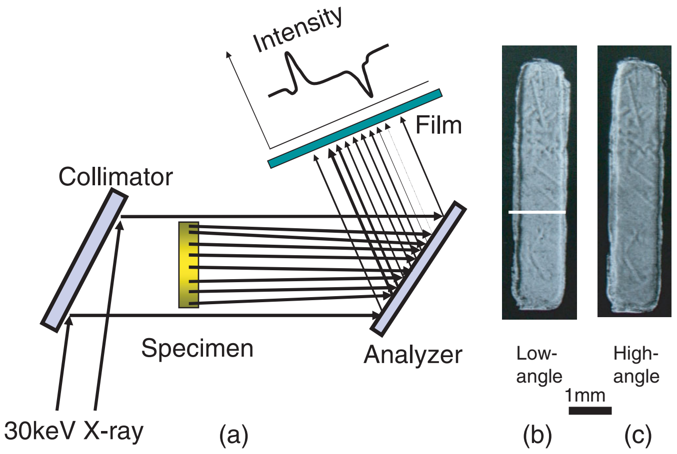

Figure 1
Experimental setup and diffraction-enhanced images of the cross-section of the hydrogen charged specimen. The intensity distribution in (a) corresponds to the contrast along the white line shown in (b).

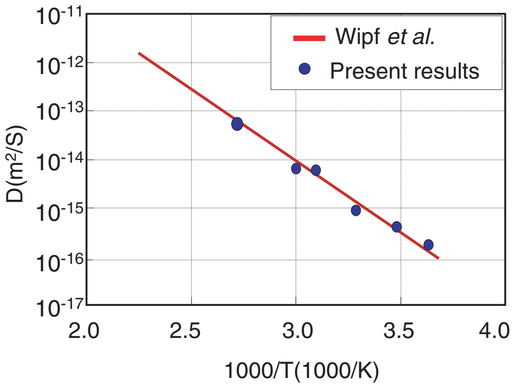

Figure 2
Diffusion coefficients of hydrogen in titanium-hydride for different charging temperatures.

The principle of the generation of refraction-contrast in Fig. 1 (b) is schematically shown in Fig. 1(a). The intensity distribution in Fig. 1(a) corresponds to the contrast along the white line shown in Fig. 1 (b). Since the refraction index of titanium for 30 keV X-rays is greater than that of titanium hydride, the distribution of titanium acts as a converging lens. Note that the direction of deflection is not the same as that for visible light because the index of refraction for X-rays is slightly smaller than unity. In this figure, the inward deflection causes a white and black fringe at the edge of the specimen, as shown in the X-ray intensity distribution. This may be attributed to the fact that the inward deflection angle varies as a function of the concentration of hydrides.

The X-ray intensity of refraction was determined from the difference between the intensity distributions in the low-angle and high-angle images. The intensity profile was compared with that obtained from a model calculation of hydrogen atom diffusion, and the fitted diffusivity was decided by trial and error. The activation energy of hydrogen diffusion was determined from the diffusivity obtained from different charging temperatures [2] which are shown as an Arrhenius plot in Fig. 2. A value for the hydrogen diffusion activation energy in titanium-hydride of 0.54 eV was obtained, showing good agreement with the widely accepted value obtained by internal friction studies. [3].

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BEAMLINE

14B

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X-Ray Interferometer Based Phase-contrast X-Ray CT Image of Cancer Specimens: Comparative Study with 4.74-Tesla MRI

Phase-contrast X-ray CT using an X-ray interferometer can reveal the inner soft tissue structures of biological objects clearly without the use of a contrast agent. Observing formalin-fixed colon cancer specimens, phase-contrast X-ray CT was compared with 4.74 tesla magnetic resonance imaging (MRI) in order to examine the properties of the technique. The phase-contrast X-ray CT image with a spatial resolution of 18 μm clearly demonstrated the inner structures of the cancer masses, such as the cancer, necrosis, and the surrounding tumor vessels and skin. Approximately a 4.0-fold higher signal to noise ratio was obtained compared to MRI with a spatial resolution of 75 μm.

Phase-contrast X-ray imaging with an X-ray interferometer, which has about a 1000-fold higher sensitivity than the conventional absorption-contrast X-ray method, can reveal the inner soft tissue structures of biological objects (Fig. 1) [1]. Using phase-contrast X-ray CT, detailed morphological structures of formalin-fixed cancerous lesions in rabbits [2] and humans [3] were visualized clearly without the use of contrast agents. At present, magnetic resonance imaging (MRI) is popularly used in clinical studies and biological research because it has excellent image contrast for differentiating soft tissue lesions. The mechanism of image generation is quite different for the two techniques. The image contrast in the phase-contrast technique is generated by the density difference, whereas that of MRI is determined by the difference of the inherent spine-density and the inherent relaxation time of water within the tissue. Here, a comparative study between phase-contrast X-ray CT and 4.74-tesla micro-MRI was performed to examine their differences and the properties of these imaging techniques using formalin-fixed colon cancer specimens from nude mice.

For the phase-contrast X-ray CT, the beam exposure time was 15 sec/projection for 250 projections over 180 degrees, and data acquisition time was about 2 hrs. For the MRI, images were achieved with a 3D gradient echo sequence (FLASH). In the 3D sequence, a matrix of 128 × 128 × 64 pixels was recorded with a TR(repetition time)/TE(echo time) of 50 ms/6 ms, a flip angle of 22 degrees, and 16 repetitions, requiring a data acquisition time of 1.8 hrs. Spatial resolutions and slice thicknesses were set to 18 μm and 18 μm for the phase-contrast X-ray CT, and 75 μm and 75 μm for MRI, respectively. To improve the signal to noise ratio of the MRI image, the spatial resolution was set to 75 μm in this study. The samples were approximately 12 mm in diameter formalin-fixed colon cancer specimens implanted in nude mice. The experiment was approved by the Medical Committee for the Use of Animals in Research of the University of Tsukuba, and conformed to the guidelines of the American Physiological Society.

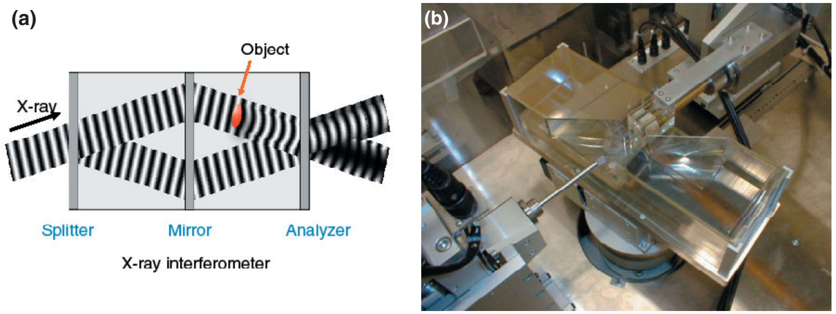

Figure 1
(a) Schematic picture of monolithic X-ray interferometer. (b) Object setting picture.

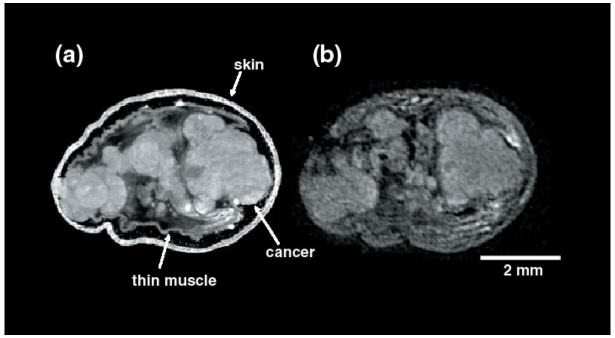

Figure 2
Phase-contrast X-ray CT image with a spatial resolution of 18 μm (a), and MRI image with a spatial resolution of 75 μm (b) of the nude mouse formalin-fixed colon cancer specimen. Data acquisition time was set about 2 hrs in both imaging techniques.

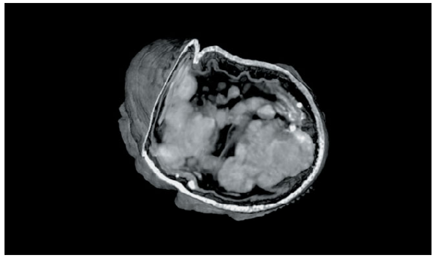

Figure 3
Three dimensional image of the colon cancer mass.

The phase-contrast X-ray CT image clearly revealed the detailed structures of the colon cancer, including cancer lesions, necrosis, surrounding tumor vessels, the subcutaneous thin muscle layer, the subcutaneous tissue and the skin (Fig. 2(a)). The bulging of the cancer from the thin muscle layer and the surrounding bleeding were well demonstrated. However, the 4.74 T-MRI image only revealed the cancer mass, the subcutaneous thin muscle layer, and the skin, and the image quality was poor compared to the phase-contrast X-ray CT images (Fig. 2(b)) [4]. The signal to noise ratio of the colon cancer lesions was about 48 in the phase-contrast X-ray CT images, about 4.0 times higher that that of the MRI images. By the phase-contrast X-ray CT, the colon cancer lesion was enable to observe three dimensionally. (Fig. 3). Using a fast imaging camera [5], images of the same quality could be acquired in about 25 minutes. For formalin-fixed biological samples, phase-contrast X-ray CT was found to exhibit higher image quality than MRI, and is thought to be suitable for imaging the detailed inner structure of soft tissue with a high spatial resolution.

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