Imaging by using proton-induced quasi-monochromatic X-ray emission

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Abstract

Quasi-monochromatic X-ray emission induced by MeV-proton bombardment onto metallic targets was applied to radiography of small samples. The X-ray energy was adjusted in the range of ≈4–23 keV by changing the target material. The radiograms were recorded on an imaging plate. A small phantom made of Lucite was prepared for the test of photography using a contrast medium (RuCl₃·H₂O; ruthenium chloride). To clearly observe the image of the contrast agent, the photon energy was chosen to be 23.17 keV (Cd Kα) which is slightly above the K-absorption edge of Ru at 22.12 keV. Only the image of the contrast agent was successfully extracted by subtracting the image taken by 21.18-keV X-rays (Pd Kα) from that obtained by the 23.17-keV photons. Also images of a small fish sample were taken using a point-like X-ray source excited by a proton microbeam with a diameter of ≈10 μm. In this experiment, the X-ray energy was adjusted to 4.51 keV (Ti Kα) to obtain a strong attenuation by Ca in the bones. A fine structure of the bones in the thin caudal fin (<100 μm) was highly contrasted by this method. The spatial resolution of the picture was ≈20 μm. Performance of the above technique based on the proton-induced quasi-monochromatic X-ray emission is discussed in comparison to conventional methods.

Keywords: Radiography; K-absorption edge; Particle-induced X-ray emission; Characteristic X-ray; Proton microbeam; Electrostatic accelerator

1. Introduction

Since the discovery of penetration capability of X-rays, application of X-ray imaging has been expanding into various scientific fields ranging from medical use to industrial testing equipment. In conventional X-ray radiography, the image contrast of a two-component sample is obtained simply by the difference in absorption properties between the components. For instance, owing to large absorption coefficient for high-Z elements, high contrast is available when heavy elements, such as Ca in bones, are localized in the sample. From this point of view, difference in absorption properties between these two components should be large enough. To realize a higher contrast, we should optimize the wavelength, or the energy of the X-ray so that the difference in the absorption coefficients can be as large as possible [1].

As shown in Fig. 1, if the energy of the X-rays is slightly above the absorption edge of the element of interest, we obtain very high attenuation by the element, and therefore very high contrast in the transmission image. One can adjust the spectra of X-ray tubes by changing the tube voltages, the anode materials, and the filters [1,2]. However, spectra of X-ray tubes always suffer from broad Bremsstrahlung continua. If an energy-variable monochromatic X-ray source is available, the contrast of the image will be further improved. Such a source can be available at synchrotron radiation facilities [3]. However, such a huge facility is not suitable for routine analysis in hospitals or research laboratories. Quasi-monochromatic X-ray emissions can be obtained also by the diffraction of polychromatic emissions produced by X-ray tubes [4]. In this case, however, high monochromaticity is not always compatible with high intensities needed for high quality radiography.
Also the size of objects is limited by the emission angle of diffracted X-rays.

Another way to obtain monochromatic X-rays is to apply particle-induced X-ray emission. When a target is bombarded with energetic heavy charged particles, such as MeV protons, the particles interact with the electrons bound in the target atoms, creating inner shell vacancies. The energy of the X-rays emitted when the vacancies are refilled is characteristic for the element from which they originate. This phenomenon is known as a tool for elemental analysis, where spectra of the characteristic X-rays emitted from the sample are measured to identify and quantify the trace elements contained in the sample [5]. If one uses an electron beam instead of the proton beam, as in the case of ordinary X-ray tubes, the energy spectrum of the X-rays is degraded by a continuum [6], because of the high Bremsstrahlung background resulting from the strong deceleration of incident electrons. In the case of the excitation by protons, the energy of the secondary electrons produced in the target is much lower, and thus dramatically reduces the background continuum under the characteristic X-ray peaks. Moreover, particle-induced X-rays have the advantage that their large emission angle allows us to handle larger objects than those in the diffracted X-ray radiography.

The final goal of our research is to establish a novel technique based on the proton-induced X-ray emission, which allows low-dose (<0.1 mSv/shot), high-resolution (<100 μm) and high-contrast radiography for clinical and/or industrial applications. Also, for practical use, the exposure time must be short enough (<1 s) and contrast media such as iodine should be applicable. However, as long as protons with energies up to a few MeV are used, it is impossible to produce strong characteristic X-rays with energies above ≈50 keV, which are needed for the radiography of human body or thick industrial samples. Hence, in this paper, our study is dedicated to the investigation on the scientific feasibility of using these quasi-monochromatic emissions to improve the quality of X-ray radiography. The effectiveness of dual-energy subtraction imaging technique based on the strong attenuation near the K-absorption edge is emphasized. We present preliminary results of experiments using a phantom filled with a contrast medium. Also application of a proton microprobe for radiography of fine structures of small biological samples is demonstrated.

2. Experimental

2.1. Experiments using conventional millimeter beams

Fig. 2 shows the experimental setup. As sources of primary MeV proton beams we used a 1.7-MV tandem pelletron at Research Laboratory for Nuclear Reactors (RLNR), Tokyo Institute of Technology and a 3 MV single-ended electrostatic accelerator at Takasaki Radiation Chemistry Research Establishment, Japan Atomic Energy Research Institute (JAERI) [7]. The incident proton energies are 3.0 and 2.6 MeV for experiments at RLNR and JAERI, respectively. The beam current was ≈10–100 nA. The beam diameter was ≈1–2 mm, depending on experimental conditions. We used thick 46Pd, 47Ag, 48Cd and 49In targets. The X-rays emitted from these metallic targets were extracted from a Mylar window with a thickness of 100 μm.

The radiograms were recorded by an imaging plate with a size of 30×50 mm². We used an imaging plate readout system (Combix 2000, Crossfield corp.) dedicated to dental use. For protection of the surface of the imaging plates, we covered them with a Mylar envelop (60 μm in thickness) during exposures. Exposure time was ≈10²–10³ s, depending on the X-ray yield and the sensitivity of the imaging plate. The energy spectra and irradiation doses of X-rays during the exposure were monitored by a CdTe detector.

The structure of the phantom is illustrated schematically in Fig. 3. Holes with a diameter of 2 mm were drilled in a Lucite block. Four holes (B, C, D and E) were filled with contrast media with different concentrations. We used aqueous solutions of RuCl₃·H₂O (ruthenium chloride)
as the contrast media. Two holes filled with air run parallelly (A) and perpendicularly (F) to others in order to simulate a background noise due to inhomogeneity in the sample. The imaging plate was directly put onto this phantom. The distance between the X-ray source and the bottom surface of the phantom was 30 mm. To clearly observe the image of the contrast agent, the X-ray energy was chosen to be 23.17 keV, which is slightly above the K-absorption edge of $^{44}$Ru at 22.12 keV \[8\]. These 23.17-keV photons were generated by bombarding the Cd target. Also an image was taken using Pd K$_{α}$ X-rays with an energy of 21.18 keV, which is just below the K-edge of Ru.

Together with the KX-rays, LX-rays are emitted by electron transitions to the L-shell. However, the energy of LX-rays is generally much lower than that of KX-rays. The phantom was thick enough to completely absorb these low-energy LX-rays from the target elements.

### 2.2. Application of a proton microprobe

The X-ray imaging of a biological sample with a very small source size was tested by using a proton microprobe system \[9,10\] at JAERI Takasaki. The experimental setup is illustrated in Fig. 4. The sample was a dried small fish (fry of Japanese sand lance, *Ammodytes personatus*). The distance between the X-ray source and the sample was 2 mm. A 100-μm-thick Mylar foil was used to support the sample. To obtain a magnified image, we put an imaging plate 34 mm behind the sample. The size of the primary proton beam was $\approx 10 \mu\text{m}$ (horizontal) $\times \approx 5 \mu\text{m}$ (vertical). The beam intensity was $\approx 10–20 \text{nA}$. The energy spectrum and irradiation dose of the X-rays during the exposure were monitored by a Si(Li) detector. To clearly observe the bones in this thin sample, the attenuation of the X-rays by Ca should be as large as possible. We used $^{22}$Ti as the target in this measurement, since the energy of the Ti K$_{α}$ X-ray is 4.51 keV, which is just above the K-absorption edge of Ca at 4.04 keV \[8\]. Also, a radiogram was taken using 8.05-keV K$_{α}$ X-rays emitted by a $^{29}$Cu target for comparison.

In this experimental geometry the metallic targets work not only as an X-ray source but also as a vacuum window. Thanks to this configuration we could put the sample very near to the source, although the sample was kept in the atmosphere. To eliminate self-absorption of the X-rays by the target layer we used metallic foils as thin as possible. We used Ti and Cu foils with a thickness of 20 μm. Nevertheless, the X-ray intensity was reduced by a factor of 30–50% due to the attenuation in the foil. LX-rays from these metallic targets were almost completely attenuated by self-absorption.

In this geometry, the sum of the mass thickness of the target (metallic foil, 20 μm), air (2 mm) and the sample holder (Mylar, 100 μm) in front of the sample was not enough to stop the incident protons. To protect the sample from the damage due to the proton irradiation, we put a 60-μm Mylar foil additionally behind the metallic foil.

### 3. Results and discussion

#### 3.1. X-ray spectra

Fig. 5 shows an example of X-ray spectra obtained by proton irradiation onto the different metallic targets. We used conventional millimeter-size beams available...
at RLNR. The incident proton energy was 3 MeV. We see that quasi-monochromatic X-rays with variable energies can be obtained by changing the target element. Not only Kα emissions but also small peaks of Kβ lines are observable. Nevertheless, we do not see background continua due to Bremsstrahlung. Generally, when a target is bombarded by MeV protons, characteristic X-rays with different energies can be observed in the energy spectrum owing to different electronic transitions. However, if the target is a simple substance, the strongest line may be used for radiography, since the spectrum is usually very simple. The contributions of other small peaks are negligibly small. Otherwise they can be attenuated by appropriate absorption-edge filters.

3.2. Application of strong attenuation above the K-absorption edge

Fig. 6(a) shows a radiogram of the phantom taken with 23.17-keV X-rays emitted from the Cd target. This picture was taken at the JAERI facility. The proton energy, beam current and the exposure time were, respectively, 2.6 MeV, 100 nA and 1000 s. The X-ray dose was 0.3 mSv. Fig. 6(b) shows another result, where the target was changed to Pd to produce 21.18 keV-X-rays. Compared with the picture taken with the Pd target, the images of the Ru contrast medium in holes B, C and D are highly contrasted when the Cd target was used. This result can be explained by the strong attenuation of 23.17-keV X-rays (Cd Kα) near the K-absorption edge of Ru at 22.11 keV.

In both photographs the structure of the Lucite block (holes A and F) are still visible as backgrounds. In order to erase these backgrounds, the image in Fig. 6(b) was logarithmically subtracted [1,2] from that in Fig. 6(a). The result is shown in Fig. 6(c). This subtraction process was performed by the following formula [11]:

$$\Delta L \equiv \ln I_H - \alpha \ln I_L$$

where \(I, \mu, x,\) and \(c\) are the image intensity, the attenuation coefficient, the equivalent thickness and a constant, respectively. The subscripts H and L mean higher and lower energy of the X-rays, respectively. The subscripts 1 and 2 stand for the Ru contrast medium and the Lucite, respectively. By choosing the weighting factor \(\alpha\) to be \(\mu_2H/\mu_2L\), the contribution of the Lucite can be removed from the image. The linear attenuation coefficients for Lucite at photon energies of 21.18 and 23.17 keV are, respectively, 0.601 and 0.543 cm\(^{-1}\), so we have \(\alpha = 0.543/0.601 \approx 0.90\). In Fig. 6(c), where \(\Delta L\) defined by the above equation is shown, we see that the backgrounds due to the structure of the Lucite block have been almost completely disappeared and only the images of the contrast media are more clearly observable. This noise reduction technique may be used to find very small tumors in non-uniform tissue structures [3] of small biological samples.
3.3. Imaging by using the proton microprobe

Fig. 7(a) shows a transmission image of the fish sample obtained by 4.51-keV photons emitted from the Ti target. Fig. 7(b) shows a picture taken by 8.05-keV photons from the Cu target. The exposure time was 3000 s for each sample. The radiation dose was 0.36 mSv for the Ti target and 0.18 mSv for the Cu target. For comparison a radiogram of the same fish is shown in Fig. 7(c), where conventional millimeter beam was used to produce the 4.51-keV X-rays. The detailed inner structure of the sample is observable only when the microbeam was used. The 8.05-keV emission seems suitable for observation of thick part, such as the spine (~400 µm in thickness) in this case. As expected, however, the fine structure of the very thin caudal fin (<100 µm) is more highly contrasted by 4.51-keV X-rays than that by 8.05-keV photons. This result is due to the strong absorption of 4.51-keV X-rays by Ca in the bones. The spatial resolution of the picture in this case was ~20 µm, whereas that for Fig. 7(c) was ~1 mm.

In the present experimental geometry, unsharpness of the radiographs is determined mainly by the size of the penumbra due to the finite source size. The penumbra size $P$ on the imaging plate is given by

$$P = (M - 1) r_s,$$

(2)

where $r_s$ denotes the source size which is roughly equal to the size of the primary proton beam. The parameter $M$ is the magnification defined by

$$M = \frac{l_1 + l_2}{l_1},$$

(3)

where $l_1$ and $l_2$ denote the source–object and the object–image distance, respectively. Using these parameters, the resolution of the image in actual size $\delta x$ is evaluated by

$$\delta x = \frac{P}{M} = \left(\frac{M - 1}{M}\right) r_s.$$

(4)

From Eq. (4), the theoretical resolution of the images obtained by the microbeam was calculated to be ~10 µm, whereas that by the conventional beam was evaluated to be ~1 mm. These are consistent with the image qualities seen in Fig. 7.

We could not apply dual-energy subtraction imaging technique, which is mentioned in the previous section, to this experiment because characteristic X-rays emitted from lower-Z targets such as Al, Si were strongly absorbed by the Mylar foils and the sample.

4. Conclusions

We have experimentally demonstrated the application of quasi-monochromatic X-rays produced by MeV protons to high-contrast X-ray imaging. Adjustment of the photon energy by changing the metallic target enabled us to use strong attenuation slightly above the K-absorption edge for enhancement of images of the contrast medium. Especially the dual-energy subtraction imaging technique has been successfully applied to diagnostics of a non-uniform sample in combination with these quasi-monochromatic X-rays.

We have found that a metallic target bombarded by a proton microbeam is potentially a useful point-like monochromatic X-ray source, which has sizes as small as those of recent sources with cold cathodes [12]. Ti Kα X-rays produced by a proton microprobe might be a powerful tool for observation of bones in small biological samples thinner than 100 µm.

Owing to the low intensity of the proton-induced X-ray emission, however, one needs so far ~$10^2$–$10^3$ s to take...
a picture with an acceptable quality. If a proton microprobe is used, this problem is more serious. Higher proton energies will be helpful to increase the X-ray yield [5]. To reduce the necessary exposure time dramatically, one needs not only a dedicated high-current proton accelerator, but also an innovative focusing technique that works well under strong space-charge effect of intense proton beams. More realistic way is to apply an X-ray imaging detector with a higher sensitivity. If the imaging plates were replaced by a CCD-based X-ray imaging device [13], the sensitivity would be further improved and the exposure time would be reduced.

Despite these problems, especially if a proton microprobe is used, the performance of the method presented in this paper is already enough for many kinds of high-resolution, high-contrast radiography of thin biological samples, if long exposure time such as \( \approx 1 \) h is permitted.

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