Source of luminescence of water lower energy than the Cerenkov-light threshold during irradiation of carbon-ion

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Abstract

Although luminescence of water during irradiations of proton and carbon-ion lower energy than the Cerenkov-light threshold were found recently, the sources of the luminescence were not yet obvious. To estimate the sources of the luminescence, we measured the light spectrum of the luminescence of water during carbon-ion irradiations and estimated the sources of the luminescence. Using an ultraviolet (UV) light sensitive charge coupled device (CCD) camera, we measured the luminescence images of water during carbon-ion beam irradiations by changing optical filters, derived the light spectra of the luminescence of water and compared with the calculated results. The intensity of the measured light spectrum of the luminescence of water at the Bragg peak region was decreased as the wavelength of light proportional to $\lambda^{-2.0}$ where $\lambda$ is the wavelength of the light, indicating the source of the luminescence of water can be electromagnetic pulse produced by the dipole displacement inside the water molecules. In the shallow part of the water prior to the Bragg peak, where the Cerenkov-light is included, the spectrum showed steeper curve that is proportional to $\lambda^{-2.6}$, which was similar to the calculated spectrum of Cerenkov-light including the refractive index changes of water with the wavelength of light. From these results, the luminescence of water is thought to be mainly come from electromagnetic pulse produced by the dipole displacement inside the water molecules.

1. Introduction

It was widely believed that luminescence was not emitted from water by the irradiation at lower energy than the Cerenkov-light threshold [1]. Contrary to this scientific consensus, we found that the luminescence of water can be imaged during proton or carbon-ion irradiations using a cooled charge-coupled device (CCD) camera at lower energy than the Cerenkov-light threshold [2, 3]. We also reported that the luminescence imaging of water was possible even for alpha particles and x-ray at lower energy than the Cerenkov-light threshold [4, 5]. The luminescence of water was applicable to the range estimation [2, 3] as well as beam width estimations [6] of particle therapies and will be promising for dose estimations. However the source of the light emitted from water during radiation irradiations at lower energy than the Cerenkov-light threshold was not yet obvious.

Measurements of light spectra of the luminescence are possible method to estimate the sources of the light emitted from water during irradiation at lower energy than the Cerenkov-light threshold. Spectra measurements were used for scintillation of air from the nitrogen gas for the detection and imaging of alpha particles [7–9] and x-ray [10]. Possible light sources of the discovered luminescence of water are electromagnetic pulse produced by the dipole displacement inside the water molecules which has the light spectrum proportional to $\lambda^{-2}$ where $\lambda$ is the wavelength of the light [11], bremsstrahlung in optical region which has the light spectrum proportional to $\lambda^{-1}$ [11], and radicals produced in water which may have some peaks in the
spectrum. Also scintillation phenomenon may be occurred similar to the light production of acrylic resin [12]. If the source of the luminescence of water becomes clear, it will have a great impact and advance in high energy physics, applied physics, radiation chemistry, medical physics and other many research fields. Consequently we measured the light spectra of the luminescence of water during carbon-ion beam irradiations with the energy slightly higher than the Cerenkov-light threshold. With the carbon-ion energy, since Cerenkov-light was not included in Bragg peak but it is included in the shallow part of water, we could obtain significant information to estimate the sources of the luminescence of water.

2. Methods

2.1. Experimental setup for light spectra measurements during carbon-ion irradiation

Figure 1 shows a schematic drawing of our experimental setup for the spectra measurements of water during carbon-ion irradiation. These are basically same setup as the luminescence imaging of water during proton or carbon-ion beam experiments [2, 3], except one of the optical filters was set in front of the lens of the CCD camera. In addition, ultraviolet (UV) sensitive CCD camera and UV transparent lens were used to enable the measure of the shorter wavelength of the luminescence more precisely.

We placed a black box on a table of a carbon-ion therapy system (Mitsubishi electric Corporation, Japan) in which the water phantom and the CCD camera were installed. We used a UV sensitive cooled-CCD camera (BITRAN BU-56DUV, Japan) with a C-mount F-2.8 UV transparent lens (Universe Optical Industries Co, UV0928CM2, Japan) set ~40 cm from the phantom surface. We used a UV transparent liquid container made of 5 mm thick acrylic plate (Kuraray PARAGLAS UV00, Japan) with outer dimensions of 20 cm (horizontal) × 20 cm (vertical) × 10 cm (depth) in which distilled water was contained. The sensitivity profile of the CCD camera, transmission properties of lens and acrylic container used for the experiments are shown in figure 2 which has the sensitivity or transparency between 250 nm to more than 700 nm. The water phantom was set in the black box and luminescence imaging was conducted.

For the measurements of the spectra of the luminescence of water, we set one of the long-pass optical filters in front of the lens of the CCD camera. We measured each image with one of the filters during irradiation of carbon-ion beam. The optical filters used were long-pass type which pass longer than 350 nm (Asahi Spectra, LU0350), 450 nm (Asahi Spectra, LV0450), 550 nm (Asahi Spectra, LV0550), 650 nm (Asahi Spectra, LV0650), and 750 nm (Asahi Spectra, LI0750).

The subtracted images were calculated between these images to produce bandpass images and profiles of shorter than 350 nm, 350 nm to 450 nm, 450 nm to 550 nm, 550 nm to 650 nm, and 650 nm to 750 nm. The intensities of these profiles were corrected for sensitivity of the CCD camera and transmission properties of the lens and acrylic container.

2.2. Spectra measurements of water during irradiation of carbon-ion beam

A 241 MeV/u carbon-ion beam was irradiated to the water phantom from the upper side of the phantom. The CCD camera imaging was started before the carbon-ion beam irradiation. We did not apply the triggering in the measurements of the CCD images. During imaging of the CCD camera, the carbon-ion beam was irradiated to the phantom for 30 s (total number of particles: 5.6 × 10¹⁰). We measured the images of the luminescence of
water without optical filter and with 5 types of long pass filters. We also measured an image without irradiation for background and uniformity correction of the images.

### 2.3. Image processing

The acquired images were processed using public domain software (ImageJ). First, the noise spots due to the direct detection of radiation by the CCD image sensor were eliminated using the high-intensity characteristics and the small pixels of these noises using a function of the ImageJ software. Then the background level and the non-uniformity of the images by the CCD camera were corrected by subtracting the image measured without irradiation. To evaluate the absolute size of the images, optical photo of the phantom was used. We converted the distances to absolute values using the absolute pixel size obtained from the optical image. We also measured the depth profiles of the images. The widths of the profiles were 30 pixels (25 mm). Depth profile was obtained by setting a profile line on the beam part in each luminescence image in the depth direction (vertical direction) from the water surface, and the intensity distribution for the profile line was calculated.

### 2.4. Calculation of the light spectrum of Cerenkov-light of water

We calculated the light spectrum of Cerenkov-light of water taking into account of the wavelength dependency of the refractive index because we noticed that the refractive index of water is changed with the wavelength. Since Cerenkov-light threshold is changed with the wavelength of the light, the shape of the spectrum of Cerenkov-light intensity is slightly different from the conventional Cerenkov-light spectrum which is proportional to $\lambda^{-2}$. We used the reported refractive index of water as a function of wavelength of light \[13\] and calculated produced number of Cerenkov-light photons ($N$) for a fixed length of water ($l$) within the wavelength between $\lambda_1$ to $\lambda_2$ by the following equation reported by Jelley \[11\].

$$ N = 2\pi a(l/\lambda_1 - 1/\lambda_2)(1 - 1/\beta^2/n^2) $$  \hspace{1cm} (1)

where $a$: fine structure constant ($1/137$)

$\beta$: ratio of the speed of electron (v) in water to that of speed of light in vacuum (c) ($v/c$)

$n$: refractive index of water

$\beta$ was calculated by the following equation for electron.

$$ \beta = \sqrt{1 - [511/(E + 511)]^2} $$  \hspace{1cm} (2)

where $E$: average energy of electrons in keV produced by carbon-ion higher than Cerenkov-light threshold

The energy spectrum of the secondary electrons produced by carbon-ion was calculated by Monte Carlo simulation and average energy of electrons produced by carbon-ion higher than Cerenkov-light threshold was calculated by the energy spectrum of the secondary electrons.

We calculate the number of produced Cerenkov-light photons for 1 mm length with 10 nm wavelength width from 200 nm to 700 nm and plotted as a function of wavelength. The plotted curve was fitted with a power function approximation.
3. Results

3.1. Spectra measurements of water during irradiation of carbon-ion beam

Figure 3 shows luminescence images of water without and with 5 different optical long pass filters during irradiation of 241 MeV/u energy carbon-ion beam. As the wavelength of the optical long pass filter increased, the intensity of the images was decreased.

The depth profiles of the images of the water phantom without and with 5 different optical long pass filters are shown in figure 4(A). We could clearly observe the Bragg peaks of the carbon-ion beams in all profiles. Also we could observe the higher intensities in the shallow region (smaller depth) by the Cerenkov-light in the depth profiles.

We calculated the bandpass luminescence images of water with 100 nm wavelength widths made from the subtracted images. For example, the image of 400 nm (350 nm–450 nm) was made by subtracting the luminescence image of water with 450 nm long pass filter from that with 350 nm long pass filter. The subtracted images were corrected for CCD camera sensitivity, lens and acrylic container transmission properties. The background signal of the CCD camera in the image was corrected by the subtraction process of the images because both images contained the same background signals. We show depth profiles obtained from the bandpass images of 100 nm wavelength widths in figure 5. We could observe the differences of the shapes of the depth profiles with the wavelengths.
Light spectra of luminescence of water during irradiation of carbon-ion beam obtained from the bandpass profiles are shown in figure 6. Figure 6(A) shows the spectrum of water during irradiation of carbon-ion beam at shallow part (10 mm to 40 mm of the depth profiles) where Cerenkov-light is included in the luminescence [1, 3]. The spectrum was corrected for the CCD camera sensitivity and transmission property of the lens. The distribution decreased rapidly as the wavelength increased. The fitting curve showed the spectrum at the shallow part was proportional to $\sim \lambda^{-2.6}$. We also show the spectrum of water near Bragg peak position (100 mm to 130 mm of the depth profiles) during carbon-ion beam irradiation in figure 6(B) where the fraction of Cerenkov-light in the luminescence was almost zero because the energy of carbon-ion and produced secondary electrons are below the Cerenkov-light threshold [1, 3]. The spectrum was also corrected for the CCD camera sensitivity and transmission property of the lens. The intensity distribution decreased more slowly as the wavelength increased. The fitting curve showed the spectrum at the Bragg peak was proportional to $\sim \lambda^{-2.0}$.

### 3.2. Calculated light spectrum of Cerenkov-light of water by carbon-ion irradiation

We show refractive index of water plotted from the data in the table of ref [13] in figure 7(A). The refractive index of water increased as the wavelength of light decreased. Figure 7(B) shows the calculated Cerenkov-light threshold energy for electrons as a function of wavelength. The Cerenkov-light threshold level is lowered as the wavelength decreased.

We show the produced number of light photons as a function of wavelength for various average electron energy in figure 8(A). The Cerenkov-light photons increased as the average electron energy increased. We show...
the comparison of shapes of these curves in figure 8(B). All the curves are steeper than that of $\lambda^{-2}$ and the curves are steeper for lower average electron energies.

Monte Carlo simulation result of the secondary electron energy spectrum in 10 mm to 40 mm depth of water with 241 MeV/u carbon-ion irradiation is shown in figure 9(A). Most of the produced secondary electrons are distributed lower energy than the Cerenkov-light threshold and the average electron energy higher than the Cerenkov-light threshold was 330 keV. We show the calculated produced number of light photons as a function of wavelength for average electron energy of 330 keV in figure 9(B). The produced number of light photons are higher than the curve of $\lambda^{-2}$ and the fitting curve showed the spectrum was proportional to $\sim \lambda^{-2.5}$.

4. Discussion

We could successfully obtain the light spectra of luminescence of water during irradiations of carbon-ion beams by the use of the UV sensitive CCD camera and optical filters. The spectra of luminescence of water showed difference between that in the shallow part and near the Bragg peak as shown in figures 6(A) and (B). Because the carbon-ion beam used for the measurements (241 MeV/u) was slightly higher than the energy of carbon-ion
(120 MeV/u for carbon-ion beam) to produce the induced secondary electrons which produce Cerenkov-light (260 keV for electrons for ~500 nm wavelength of light) [1], luminescence of water in the shallow part contained Cerenkov-light in addition to the luminescence of water.

Because the spectrum of luminescence of water at Bragg peak region showed the distribution proportional to \( \lambda^{-2.0} \), the luminescence of water can be attributed to the electromagnetic pulse produced by the dipole displacement inside the water molecules which has spectrum of \( \lambda^{-2.0} \) [11]. Although Cerenkov-light also shows the spectrum proportional to \( \lambda^{-2} \) without considering the refraction index changes of water with wavelength, the source of the luminescence at Bragg peak region could be the electromagnetic pulse produced by the dipole displacement inside the water molecules since the luminescence at the Bragg peak region does not contain Cerenkov-light. The light produced by the electromagnetic pulse becomes Cerenkov-light when the energy of electrons is higher than the Cerenkov-light threshold. Thus the observed luminescence of water is expressed as the original source of light that could not be Cerenkov-light because of the insufficient energy of the secondary electrons.

The spectrum of luminescence of water at shallow region showed the distribution proportional to \( \lambda^{-2.6} \), steeper than that at the Bragg peak region. Although both the spectra of Cerenkov-light and electromagnetic pulse produced by the dipole displacement inside the water molecules are known to have the distributions proportional to \( \lambda^{-2} \), the distribution at shallow region had steeper distribution than \( \lambda^{-2} \). This is because Cerenkov-light spectrum proportional to \( \lambda^{-2} \) is not always true when the energy of the produced secondary electrons from carbon-ion beam is slightly higher than the Cerenkov-light threshold as was shown in the calculated result in figures 8(B) and 9(B). Since the number of the Cerenkov-light photon production is the function of refractive index of water which is higher for shorter wavelength for water as shown in figure 7(A) [13], the water produces higher Cerenkov-light photons in shorter wavelength than the curve proportional to \( \lambda^{-2} \) when the precise refractive index was used for calculation. This phenomenon is more significantly observed when the electron energy is only slightly higher than the Cerenkov-light threshold as shown in figure 8(B). In fact, the shape of the spectrum of the measured luminescence of water at shallow region shown in figure 6(A) \( (\lambda^{-2.6}) \) is similar to the calculated shape of the spectrum shown in figure 9(B) \( (\lambda^{-2.5}) \).

From the analogy of the sources of the luminescence of acrylic resin [12], we can also expect the scintillation phenomenon in water during irradiation of radiation. Although water molecules, hydrogen and oxygen atoms, or other produced molecules such as OH radicals by the interactions between water molecules and radiations, have their own exited states which can emit light in visible optical regions [14, 15], we could not so far observe obvious distribution from these phenomena in the experiments. One of the reasons was the small number of optical filters limited the wavelength resolution of the measured light spectrum of luminescence of water. By using a high sensitivity optical spectrometer to obtain higher wavelength resolution of the measured light spectrum of luminescence of water, we may be able to measure the contribution of the light from the exited states of water molecules or OH radicals.

Figure 9. Electron energy distribution of 10 mm to 40 mm depth of carbon-ion beam (A) and produced number of light photons as function of wavelength of light for average electron energy of 330 keV (B).

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\( \lambda \) denotes the wavelength of light.
5. Conclusions

The luminescence of water during irradiations of carbon-ion beams in the Bragg peak region is attributed to the electromagnetic pulse produced by the dipole displacement inside the water molecules. The steeper distribution of the spectrum than the theoretical distribution of Cerenkov-light ($\lambda^{-2}$) at shallow part attributed to the lower Cerenkov-light threshold of water for shorter wavelength of light due to the refractive index changes with the wavelength of light. The discovery of the source of the luminescence of lower energy than the Cerenkov-light threshold will impact many research fields, such as nuclear physics experiments [16, 17], molecular imaging [18–20], or medical physics [21–24] because all these research assumed the detected optical signals originated from only Cerenkov-light. Most of the detected optical signals might not come from the Cerenkov-light but from our discovered luminescence lower energy than the Cerenkov-light threshold that was from electromagnetic pulse produced by the dipole displacement inside the water molecules.

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