Auger effects on bromo-deoxyuridine-monophosphate irradiated with monochromatic X-rays around bromine K-absorption edge

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Summary. In order to study the enhanced effect by Auger cascade, samples of bromo-deoxyuridine-monophosphate (Br-dUMP) in aqueous solutions were irradiated with monochromatic X-rays at 13.49 keV and 13.43 keV, just above and below the K-absorption edge of bromine, using synchrotron radiation as a source. Radiolytic products such as deoxyuridine-monophosphate (dUMP), uracil and bromo-uracil (Br-uracil) were isolated using high performance liquid chromatography. Their amounts were quantitatively analysed as a function of the absorbed dose in the solutions containing Br-dUMP for the energy of the X-rays. G values for these products were calculated on the basis of the absorbed energy. As the results, the ratios of G values of radiolytic products from Br-dUMP between X-rays of 13.49 keV and 13.43 keV were 2.2 for dUMP, 1.02 for Br-uracil and 1.23 for uracil, suggesting clearly the energy dependent enhancement. On the other hand, little significant difference between X-rays of 13.49 keV and 13.43 keV was observed for the G values of uracil released from dUMP irradiated in aqueous solutions. It seemed to confirm that the Auger electrons from K-shell of bromine atoms might play the main role for energy-dependent enhancement at induction of these radiolytic products.

Introduction
In recent years the possibility of a new method of radiotherapy for cancer, namely, photon activation therapy, has been proposed on the basis of the idea that the Auger effect enhances the lethal effect of the radiation on cells (Fairchild et al. 1982; Halpern 1982). Several studies on the biological effects associated with Auger phenomena have been carried out using Auger electron emitters (\(^{125}\)I, \(^{77}\)Br) incorporated in DNA in various types of cell (Booz and Smit 1977; Burki et al. 1977; Feige 1977; Krisch et al. 1977). For X-ray-induced Auger cascade, an enhancement in biological effectiveness has also been reported (Halpern and Mütze 1977; Shinohara et al.)
Fig. 1a and b. The irradiation cell. It is made of plastic of acrylic resin, and the front is covered by a thin film of polyimido (62.5 μm thickness) which is highly transparent for X-rays at 13.49 keV and 13.43 keV. The coefficient of transparency is 0.992.

1985; Maezawa et al. 1988a, b). In molecular system very few studies have been made concerning Auger effect (Diefallah et al. 1970; Halpern and Stöcklin 1974).

In this report to induce Auger enhancement effectively a strong and monochromatic (ΔE/E ~ 0.001) X-rays in the soft X-ray region (Kobayashi et al. 1987) from a synchrotron radiation has been used. Br-dUMP in aqueous solution were irradiated with monochromatic X-rays, by using synchrotron radiation selected below and above the K-edge of bromine (13.43 and 13.49 keV).

Materials and methods

Preparation of sample solutions

5-Bromo-2'-deoxyuridine-5'-monophosphate (Br-dUMP) and 2'-deoxyuridine-5'-monophosphate (dUMP), purchased from Sigma Co. Ltd., were dissolved in triply-distilled water at concentration of 200 mg/cm³. Then 50 μl of solution was irradiated under aerobic condition in a specially designed cell made by Mr. Furusawa of Tokai University (shown in Fig. 1) having an irradiation area of 2 x 25 mm and a thickness of 1 mm, which is much thicker than the range (4 μm, Cole 1969) of photoelectrons with the maximum possible energy of 13 keV.

Monochromatic X-ray irradiation

Synchrotron radiation (from the Photon Factory, National Laboratory for High Energy Physics, Tsukuba) was used as the source of X-rays. Details of the irradiation system, the characteristics of the X-rays and their performance were described elsewhere (Kobayashi et al. 1987). Monochromatic X-rays were obtained with an 111-channel-cut silicon crystal monochromator. The calibration of energy was carried out using the K-absorption edge of Br and Ni atoms. The size of the monochromatic X-ray beam was about 3 mm vertically and 33 mm horizontally. A free air parallel-plane ionization chamber (ion collection-repeller distance, 4 cm; ion collector length, 3 cm; ion collection volume, 0.247 cm³) was used for measurements of exposure rate. The chamber was placed in front of the sample position. The X-ray intensities per 100 mA ring current were about 6.45 x 10⁻¹ C·kg⁻¹·min⁻¹ at 13.49 keV and 13.43 keV. X-irradiation for sample solution were carried out at the ring current, ranging from 100 mA to 200 mA. The total dose