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CALORIMETRIC MEASUREMENT OF THE ABSORBED DOSE IN IRRADIATION WITH AN ELECTRON OR PHOTON BEAM FROM A 60 MeV ELECTRON ACCELERATOR

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A simple calorimetric dosimeter has been contrived to measure the radiation dose absorbed by di(2-ethylhexyl)phosphoric acid in irradiation with an electron or photon beam from a 60 MeV electron accelerator. The absorbed dose rate is related to the beam current measured with a beam-position monitor in the accelerator. The beam position monitor is usable for dosimetry in calibration with a calorimeter.

Keywords: Calorimetry, Dosimetry, Dose Rate, 60 MeV Electron, Photon Beams, Di(2-ethylhexyl)phosphoric Acid, Electron Accelerator, Beam Position Monitor, Beam Current, Absorbed Dose

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60 MeV電子線加速器からの電子あるいは制動X線照射におけるカロリメトリーによる吸収線量の測定

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超プルトニウム元素等の分離に用いる抽出剤 di（2－ethylhexyl）phosphoric acid（DEHPA）の放射線分解を調べる研究の一環として，60 MeV 電子線および制動X線による照射実験を行った。試料の吸収線量を求めるために熱量計を検討した。電子加速器のビーム位置モニターでビーム電流を測定しながら熱量計の応答を記録した。試料の温度上昇速度と吸収線量率との関係を^{60}Co γ線により求め，加速器のビーム電流と線量率との関係を検討した。その結果，ビーム位置モニターの電流値から照射試料の吸収線量が推定できた。

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1. Introduction

Di(2-ethylhexyl)phosphoric acid(DEHPA) is a very useful extractant in the treatment of the waste solution from nuclear reprocessing. However, its chemical behavior in solvent extraction under high radiation dose rate is not known. In the course of the study of the separation of fission product nuclides and transplutonium elements from the reprocessing waste, the author has been studying the effects of radiation degradation of DEHPA on the extraction efficiency. Besides the study with γ rays from ^{60}Co, the use of radiation from a 60 MeV electron linear accelerator of Tohoku University has been undertaken to discriminate various chemical effects which may depend on the nature of the radiation used.

The study of radiolysis requires an accurate measurement of the absorbed energy, and the Fricke dosimeter has been widely used for such a purpose. However, G values for a continuous energy spectrum of electrons and x rays produced by an electron accelerator have hardly been studied so far.

On the other hand, a calorimetric dosimeter measures the rise of the temperature of the medium, which is inherently related to the energy imparted to the absorbing medium,1) and makes it possible to know the absorbed dose from a complicated radiation. The use of calorimetry in ion-beam studies has been reported by a number of authors.2)∿6)

During the operation of a linear accelerator, the position and intensity of the beam are liable to fluctuate. Yagi and Kondo7) installed a beam monitor on the center axis of the beam in the electron linear accelerator of Tohoku University so that the intensity and the position of the electron beam could be controlled. The beam monitor measures the electron current of the beam incident on the sample. If the energy absorbed by the sample is corelated to the electric current of the beam measured by the beam monitor, the latter can be used in place of the real dosimeter.

The present work includes two subjects, one of which is to determine the absorbed dose in DEHPA under irradiation with electron or photon beams, and the other is to find a relationship between the beam current measured with the beam position monitor and the absorbed energy of the sample material measured with a simple calorimeter.

2. Experimental

2.1 Calorimeter Assembly and Temperature Measurement System

The apparatus used for the calorimetric dosimetry is schematically shown in Fig. 1. The dosimeter consists of a cylindrical absorber cell(C) with a thermistor(A) and double walled vessel(E) surrounded with an ice-water jacket. The cell, having an approximate capacity of 7 ml, contains the substance for the irradiation, which is introduced through a guide tube(F).

In addition to DEHPA, H_2O was also irradiated for comparison. The vessel is evacuated before irradiation through the stop cock. The Takara TE 30-0.5 glass bead thermistor, having a bead diameter of 2.3 mm and a resistance of 29.64 kΩ at 0°C and 1.015 kΩ at 100°C (temperature coef. β: -4.4%), is inserted to the center of the cell through the glass capillary(B) and is used as the temperature-sensor of the calorimeter. It is incorporated into a Wheatstone bridge so as to form one side of the bridge. Although it has an exponential temperature/resistance response, the response can be considered linear in a small temperature interval ($\Delta t_{max} \simeq 2°C$). A constant voltage of 4 V is supplied to the bridge from a dry cell and the unbalanced voltage of the bridge due to the change of the temperature of the thermistor is recorded through a D.C. amplifier. The level of noise of the amplifier and the recorder is equivalent to 1.0×10^{-4} °C. The calorimeter was standardized with a ^{60}Co γ ray source (approximately 45,000 Ci), of which the spatial distribution of radiation dose rate, $1 \sim 15 \times 10^5$ R/hr, was determined by the Fricke dosimeter.

2.2 Irradiation

The irradiation set-up is shown schematically in Fig. 2. The accelerator was operated with a peak current up to 200 mA and a repetition rate of 300 Hz with a pulse duration of 3 μs producing 60 MeV electrons. The electron beam was converted to bremsstrahlung x rays of maximum energy of 60 MeV by impacting a Pt converter having a thickness of 2 mm placed behind the beam position monitor. When necessary, a sweep magnet was used to eliminate unconverted electrons from the x ray beam. The beam position monitor, a Pt plate having a diameter of 3 mm and a thickness of 0.1 mm, was set exactly on the center axis of the wave guide tube and used to determine "beam intensity" of the electron beam incident on the monitor.

The beam intensity is expressed in count per minute(cpm) and one count is equal to 1.0×10^{-6} Coulomb. The calorimeter was placed behind the beam monitor so that the center of the absorber cell lies on the center axis of the beam.

2.3 Calorimetric Theory

In the constant-temperature environment calorimetry, the energy lost or gained by the calorimetry absorber can be dealt with the laws governing energy flow. Heat transfer rate can be expressed according to Newton's law of cooling$^{8)}$,

$$\frac{dE}{dt} = -A \sum_i h_i (T_a - T_j) \tag{1}$$

where E is the energy transfered and $\sum_i h_i$ is the summation of the heat transfer coefficients for all the modes, T_a and T_j denote the absorber and jacket temperatures and A is a constant. In a semi-adiabatic method ($T_a \simeq T_j$), the heat leakage can be neglected for sufficiently small temperature rises, and the temperature rise of the absorber can be regarded as linear against time. Thus we can finally get the following relation

$$C \cdot \frac{dT_a}{dt} \propto \frac{dE}{dt} \tag{2}$$

where C is the thermal capacity of the absorber. This relation means that the absorbed dose rate can be obtained, provided C is known, by measuring the rate of the temperature rise of the absorber.

3. Results and Discussion

3.1 Calibration of the Calorimeter by ^{60}Co γ ray Source

Figure 3 shows the calibration curves of the calorimeter containing DEHPA or water as absorber. The rate of the rise in the output voltage is plotted against the dose rate of ^{60}Co γ rays for both absorbers. In the range of the dose rate examined, the response of the calorimeter showed a good proportionality to the dose rate within a standard deviation of ±2.5%. The response of the calorimeter with DEHPA and H_2O were 303 mV/Mrad and 141 mV/Mrad respectively.

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3.2 Absorbed Doses of DEHPA and H_2O under the Irradiation of Electron or Photon Beams

Results obtained by the beam experiments are summarized in Table 1. A typical recording of the temperature rise in DEHPA, which was exposed to the pulsed beam for 2 minutes, is shown in Fig. 4. The intensity of the beam in this irradiation was 99.0 cpm. After the thermal responses, measured in mV/min, had been converted to the dose rate, rad/hr, according to the curves in Fig. 3, the dose rates for DEHPA and water were plotted against the beam intensity for the pure x rays and electrons-bremsstrahlung x rays in Fig. 5.

As most results in Table 1 exceed the value obtained in the calibration with ^{60}Co, the curves in Fig. 5 were plotted on the premises that the proportionality of the rate of the temperature rise with dose rate would be valid up to the maximum temperature increase in Table 1, e.g. 84 mV/min ≈ 1°C/min, and that the contribution of the glass wall to the rise of temperature of liquid would be equal degree for ^{60}Co γ rays and 60 MeV radiations. The former assumption was sustained by the linear increase of temperature with beam current as shown in Fig. 4, which implies that there is no significant energy escape from the absorber during the course of the irradiation. Since the thermistor measures the temperature of the center of the liquid, the influence of the wall which causes the heterogeneous distribution of temperature within the absorbing body must be considered especially in different radiation fields. However, as the temperature response was constant for at least about 1 min after the irradiation as seen in Fig. 4, the temperature differences within the liquid might be small.

The most probable straight line was calculated by the least square method for the results with both DEHPA and H_2O. The doses in DEHPA and H_2O are approximately equal for the irradiation with both the electron-x rays and pure x rays. Moreover Fig. 5 shows that the absorbed dose of the materials exposed to the electrons-x rays is approximately 7.9 times greater than that for the pure x rays; this means that the absorbed energy in the absorber materials from the x ray component in the electron-x rays is about 13% of the total absorbed energy. The contributions of photonuclear reactions, that is, (γ,n), $(\gamma,2n)$, (γ,p), (γ,pn) etc. or the radiations from the nuclei produced by these reactions, and other radiation-chemical reactions were estimated several percent.[9]

3.3 Feasibility of the Beam Position Monitor for Dosimetry

The slope of the line for the electrons-x rays in Fig. 5 was 0.779 ± 0.064 and that for the x ray was 0.783 ± 0.078. The agreement of these values means that the dependency of the absorbed energy upon the beam intensity is equal in the irradiation with both pure x rays and electrons and that the absorption mechanism for photons is substantially similar to that for electrons in the energy region around 60 MeV. The fact that these values are not equal to unity implies that the dose rate is not proportional to beam intensity. This might be attributed to various factors, such as the energy distribution of the beam, the boundary radiation effects and perhaps the heat loss.

4. Conclusion

A simple calorimetric method is found to be useful for the determination of the absorbed dose of DEHPA or water irradiated with the electrons or photons from an accelerator. The doses absorbed in DEHPA and H_2O are approximately equal in the irradiation with both the electrons-photons and pure x rays, and the dose imparted to both absorbers from the electrons-photons is approximately 7.9 times greater than that from the pure x rays.

Although the addiational experiments concerning the photonuclear reactions and radiation-chemical reactions, wall effect, the heat loss and the dose uniformity through the sample will be necessary for more precise results, the beam position monitor attached to the accelerator is applicable to dosimetry by the calibration with the calorimeter.

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Table 1 Thermal responses of H_2O and DEHPA by the beam irradiations at various intensities

Absorber : H_2O		Absorber : DEHPA	
Beam intensity(cpm)	Response(mV/min)	Beam intensity(cpm)	Response(mV/min)
Electron + x ray		Electron + x ray	
11.5	8.2 ± 0.25	18.0	24.0 ± 0.72
24.5	14.1 ± 0.42	41.0	45.0 ± 1.35
50.0	30.0 ± 0.90	65.0	57.0 ± 1.71
56.5	31.0 ± 0.93	99.0	84.0 ± 2.52
77.5	40.2 ± 1.21		
x ray		x ray	
55.0	4.3 ± 0.13	31.5	4.5 ± 0.14
65.0	4.0 ± 0.12	59.5	7.7 ± 0.23
116.0	6.4 ± 0.19	77.4	10.4 ± 0.31
130.0	7.1 ± 0.21	98.0	10.8 ± 0.32
133.0	6.4 ± 0.19	112.5	11.4 ± 0.34
171.0	9.2 ± 0.28		
180.0	9.0 ± 0.27		

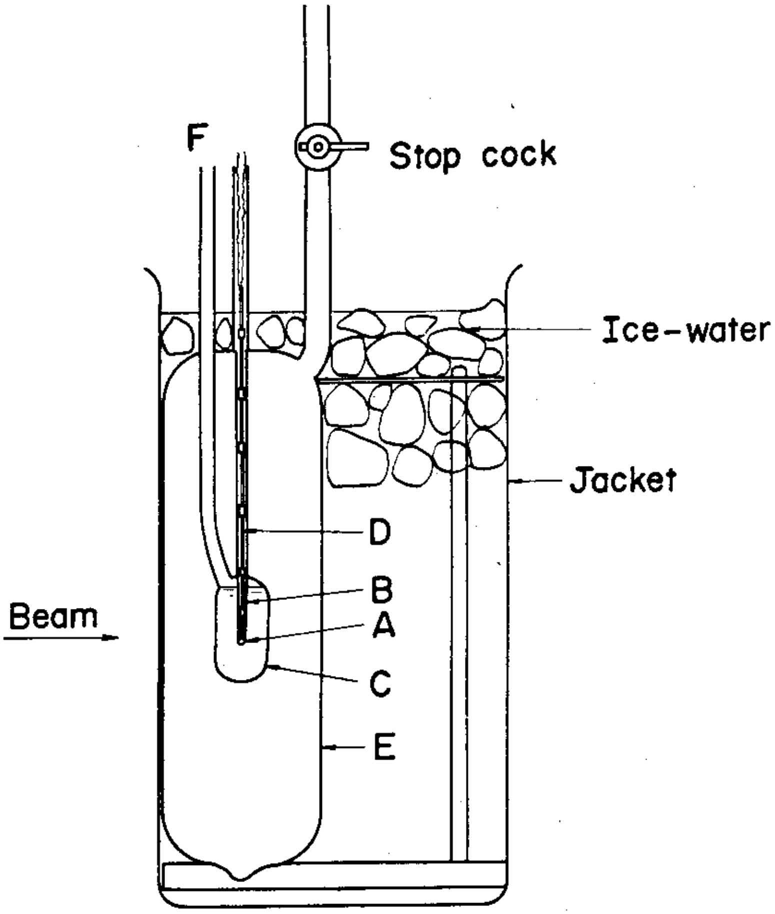

Fig. 1 Calorimetric dosimeter
A: Thermistor B: Capillary C: Absorber cell
D: Leading wire E: Vessel F: Guide tube

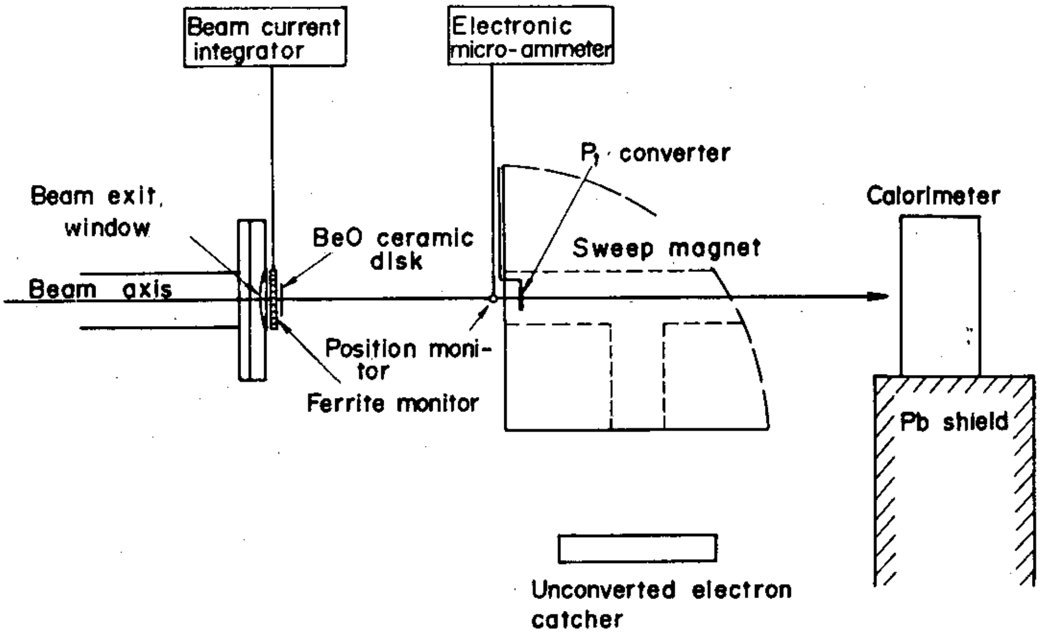

Fig. 2 Irradiation set-up

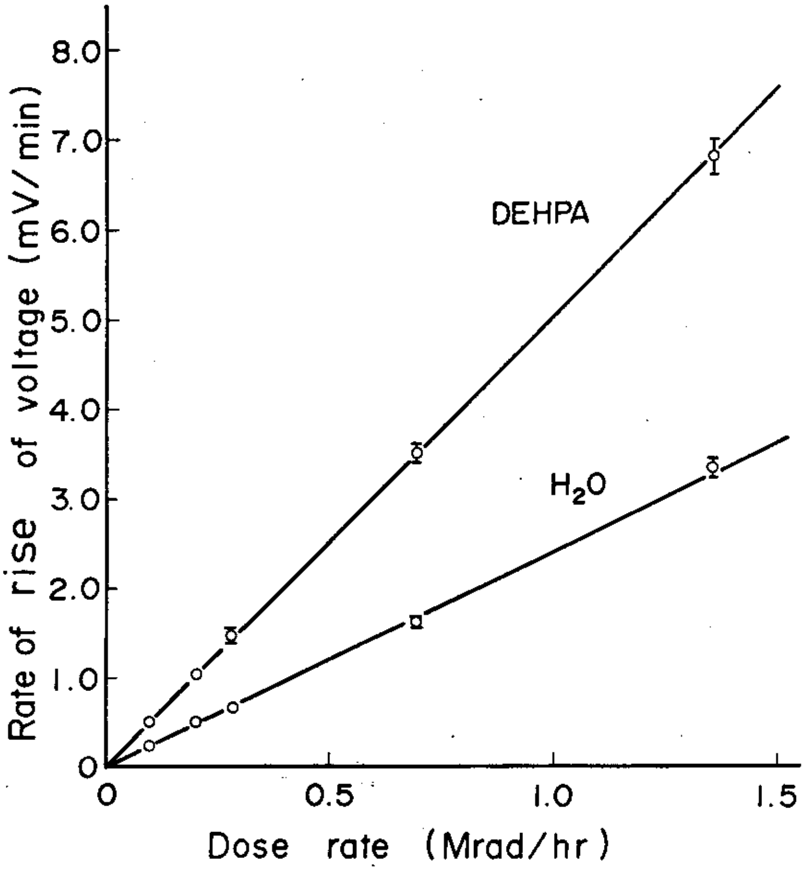

Fig. 3 Response of the calorimeter as a function of dose rate for ^{60}Co γ rays

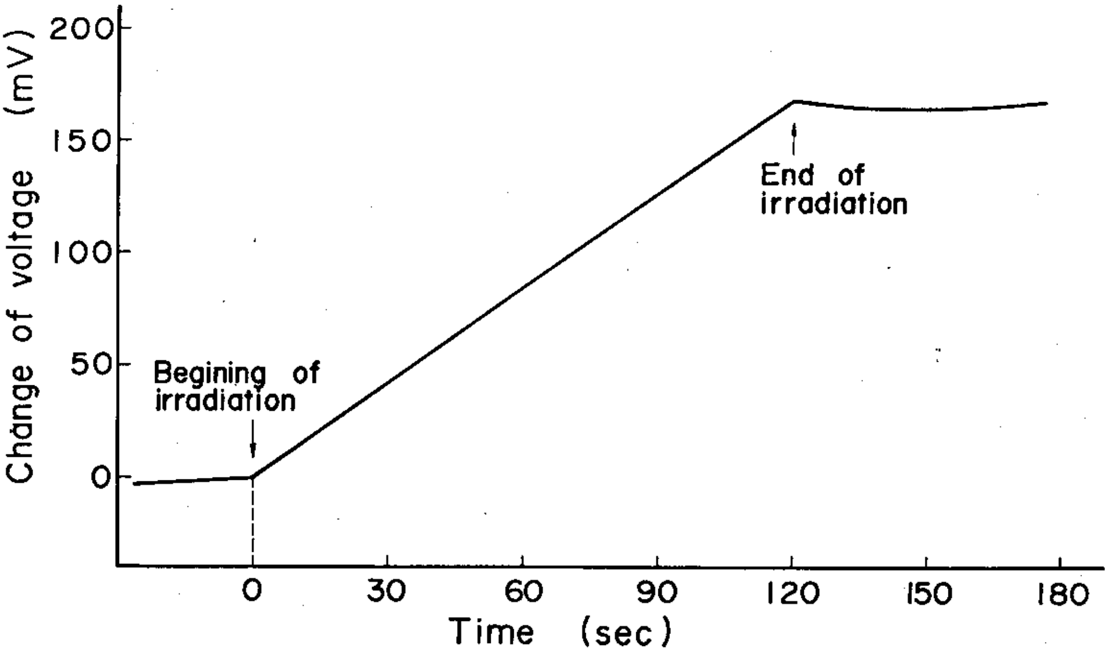

Fig. 4 A typical recording of the temperature increase in DEHPA by 2 minutes exposure of the electrons-x rays

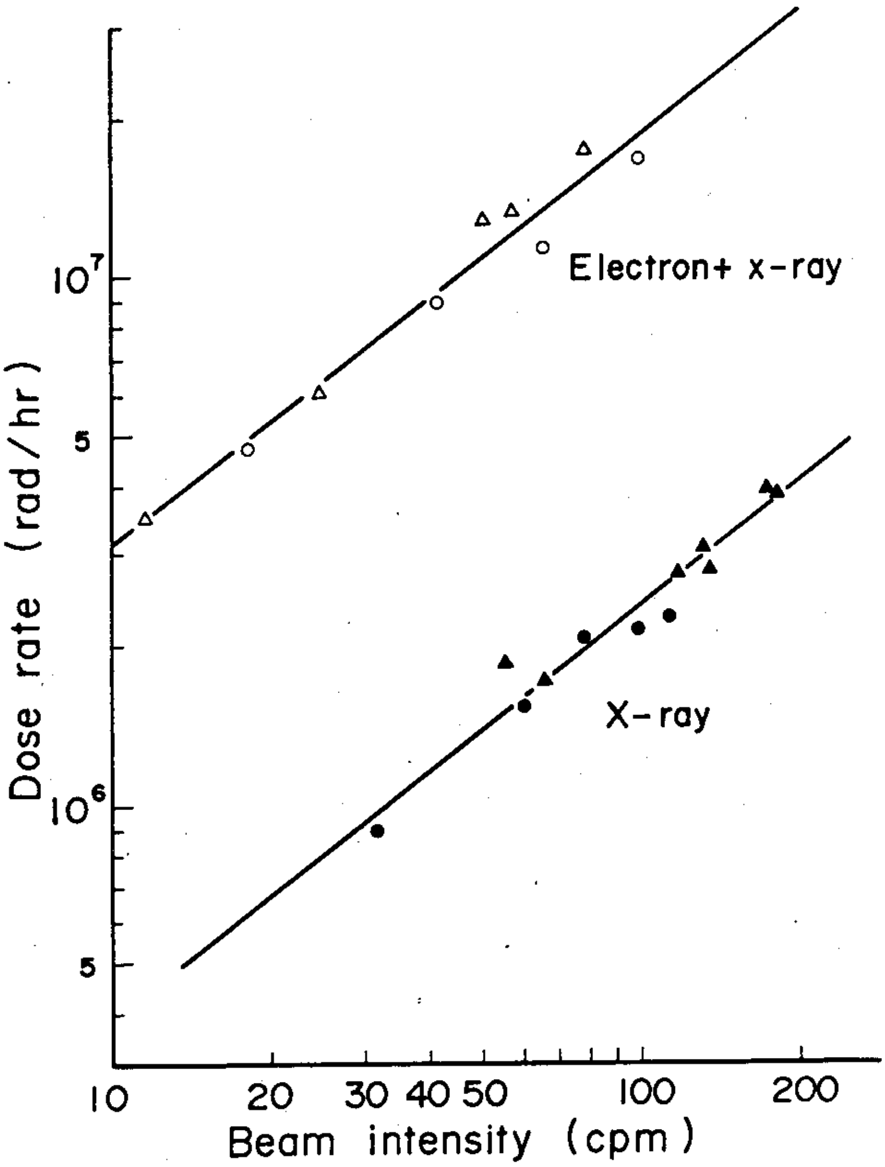

Fig. 5 Relation between dose rate and beam intensity measured with beam position monitor

○,● : DEHPA △,▲: H_2O