ABSOLUTE AND SECONDARY DOSIMETRY AT THE CYCLOTRON ION BEAM RADIATION EXPERIMENTS*

XA9949727

Z. STUGLIK
Laboratory for Measurement of Technological Doses,
Institute of Nuclear Chemistry and Technology,
Warsaw, Poland

Abstract

One of the characteristic features of ion beam radiation experiments is that the absolute methods of dosimetry are more convenient than secondary ones. In this paper the absolute method used in the course of the radiation experiments performed on the U-400 Dubna cyclotron is presented in detail. Some remarks dealing with the secondary methods are also given.

1. INTRODUCTION

The principle of resonant particle acceleration in a static magnetic field was discovered by Ernest O. Lawrence in 1930 and only two years after the first cyclotron was built at Berkeley. From that time, the technology of particle acceleration has undergone big development giving such constructions as synchrotrons and storage rings both capable to accelerate particles up to TeV. Nevertheless, the cyclotrons still hold an important position. They give an intensive and good quality beams of different ions with energy in the range 1-100 MeV/amu. Compact size, reasonable cost of installation and limited power consumption are positive economical factors of these devices. Actually, more than 200 cyclotrons are in operation over the world. From many years the number of facilities dedicated to applied studies has increased faster than that for the pure physics. Cyclotrons are used in atomic and nuclear physics, material science, biology and medicine. Chemical studies have been done rather seldom. It results not only from still high cost and hard access to the ion beams but also from experimental difficulties, among them – dosimetry. The secondary methods, useful and commonly used in low-LET experiments lose their versatile character in high-LET area. It is connected mainly with LET-dependence of the G-values. As a consequence, "the dosimetric answers" to the same dose are different not only for different ions with the same energy, but also for the identical ions with different energies. The secondary dosimeters may be eventually applied in the experiments using one kind of ions with the same energy. Of course, they should be carefully calibrated with an absolute dosimeter before using them. In such situation, the advantages in using secondary dosimeters are rather low and the absolute methods (despite their time-consuming and complicated character) are recommended for high-LET ion beam radiation experiments.

The absolute methods could be divided in two classes:

a) calorimetric methods,

b) methods based on energy and fluence measurements.

The range of the ions in matter - the third quantity necessary for dose evaluation – can be taken from physical tables [1,2] or calculated by means of computer programs [3,4].

The calorimetric methods can be used only for the light particles with relatively long ranges. However, their applicability is rather doubtful for intermediate energy heavy ions with the ranges of about 10 mg/cm^2 (~ 0.1 mm in water). The next shortcoming is that they cannot be used as on-line methods. The cyclotron ion beams are rarely so stable as electron ones and to achieve reasonable accuracy of the experiments, the ion beam fluences has to be measured directly during the irradiation.

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The methods based on energy and fluence measurements have fewer disadvantages than calorimetry and are commonly used in the cyclotron experiments. One of such methods - used in the course of the radiation experiments at Dubna facility - will be presented below.

2. EXPERIMENTAL CONDITIONS

2.1. U-400 cyclotron and the ion beams used at the chemical radiation experiments

Four-meter U-400 isochronous cyclotron has operated at Flerov Laboratory of Nuclear Reactions, JINR, Dubna, Russia from 1978. During our experiments (1990 – 96) it was equipped with PIG ion source and capable of accelerating ions from Li to Xe with a mass-to-charge ratio from 5 to 12. The maximum energy per nucleon ranged from 18 MeV for lighter ions to 3.8 MeV for ^{129}Xe.[1] The ions were extracted via charge exchange in thin (40-200μg/cm^2) graphite foils. The intensities of external beams were in the range of 10^{13} particles per second (pps) for the light ions and 10^9 pps for the heaviest one. The energy of the ions produced by the U-400 cyclotron depended on the "extraction mode" and could be changed by step of 30-40%. At a fixed extraction mode, the energy could be changed smoothly within ±5% around the value corresponding to the maximum intensity for each charge. The U-400 ion beams had a time structure: there was a train of 1.5 ms pulses separated by 5.1 ms intervals, and each such pulse consisted of a number of much shorter pulses with a duty factor of 10 and a frequency equal to that of the accelerating RF voltage (5-12 MHz). As a consequence, the instantaneous dose rate is ~40 times higher than the average value obtained as a dose divided by time of irradiation. For more experimental details see [6,7].

TABLE I. PHYSICAL PARAMETERS OF THE IONS USED IN THE RADIATION EXPERIMENTS

cyclotron chamber	extracted beams	energy in vacuum, E_0 (MeV)	specific energy in vacuum (MeV/amu)	energy on the surface of the sample, E_S (MeV)	specific energy on the surface (MeV/amu)	range in air [a] for $E = E_S$ (mm)	range in water [a] for $E = E_S$ (μm)
$^{11}B^{2+}$	$^{11}B^{4+}$	113	10.3	102	9.3	460	462
$^{12}C^{2+}$	$^{12}C^{6+}$	133	11.1	122	10.2	410	433
$^{18}O^{2+}$	$^{18}O^{8+}$	118	6.5	79	4.4	112	108
$^{22}Ne^{3+}$	$^{22}Ne^{9+}$	182	8.3	131	5.9	141	147
$^{24}Mg^{3+}$	$^{24}Mg^{11+}$	186	7.7	138	5.7	106	110
$^{40}Ca^{5+}$	$^{40}Ca^{18+}$	299	7.5	177	4.4	62	64
$^{52}Cr^{6+}$	$^{52}Cr^{21+}$	326	6.3	168	3.2	47	48
$^{56}Fe^{6+}$	$^{56}Fe^{22+}$	317	5.7	108	1.9	29	30
$^{59}Co^{7+}$	$^{59}Co^{25+}$	397	6.7	203	3.4	50	51

[a] The ranges were calculated by means of TRIM programme [3]

In the radiation chemical experiments we used defocused external ion beams which characteristics are shown in Table I. The flux was commonly attenuated to 10^8 - 10^9 pps and the fluence was usually in the range 10^9 - 10^{11} ions/cm^2. As it is seen from Table I the ranges of the ions are directly proportional to the specific energies and inversely proportional to masses. The first dependence is much stronger than the second one. The ranges in water varied from some hundreds of micrometers for the lightest ions to some tens of micrometers for the heaviest one. The arrangement of the experiment has been fitted to the ion beam characteristics, especially penetration.

[1] Some years ago it was updated (ECR source, axial injection system) and its current possibilities are much higher [5].

2.2. KHIPTI facility and irradiation conditions

The chemical laboratory KHIPTI (Russian abbreviation of the name Chemical Investigation on Heavy Ion Beams) has been dedicated mainly to radiochemical studies of super-heavy elements but also to high-LET radiation experiments. The laboratory is situated at the basement near the vertical channel of cyclotron (Fig.1). The beam extracted from U-400 chamber passes through ~8-meter long horizontal beam-pipe and then is turned 90° down by a magnet into the vertical ~10-meter part of the channel. Above mentioned magnet, two pair of magnetic lenses, a pair of steering magnets and three diagnostic loops allow to focus and guide the beam. The third diagnostic loop, situated just in chemical room was equipped with the shutter (to cut-off the ion beam if necessary), a Faraday cup (for fluence measurements), a BeO-luminofor (for visual beam profile control) and a scattering foil (0.2 μm Au) for energy measurement. The last one was driven by an electric drive, the other one pneumatically. The accelerated ions were extracted into atmosphere through thin (~ 10-μm Al or 4-μm Ti) vacuum windows. Due to the small thickness of the exit windows and rather low ion fluxes no cooling system was necessary.

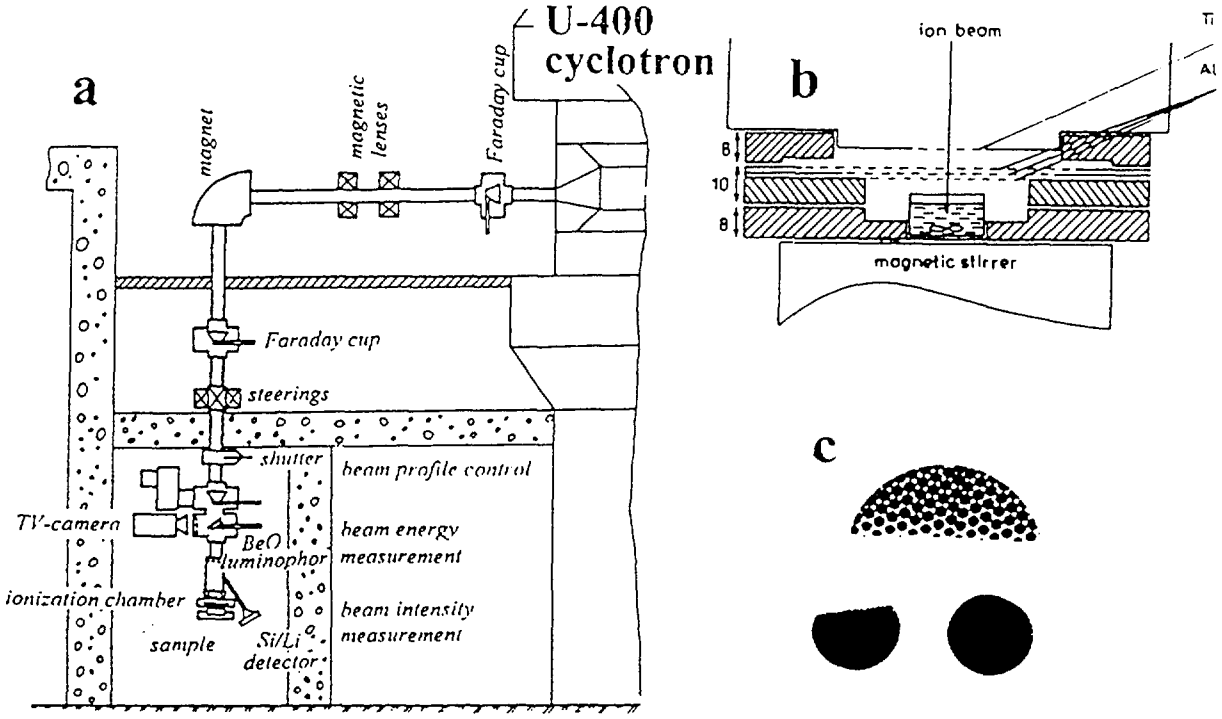

FIG.1. a) KHIPTI facility for radiochemical and radiation experiments,
b) an arrangement for the chemical ion-beam radiation experiments,
c) PVC foils used as a beam profile indicators at the time of ion beam tracing.

Unlike many other works, the samples were bombarded with the ion beams perpendicular to the air/sample surface. Such geometry has been very convenient for powder and liquid samples.

The irradiation set-up is shown on the Fig.1b. It consists of polystyrene support, thin ionization chamber, the vessel and the vessel's holder. All the elements are carefully aligned and fastened to the end of the pipe. The magnetic stirrer allows to stir liquids, if necessary.

The samples were irradiated in two positions. For the light ions, the distance between the exit window and the sample was 30-60 mm, while it was reduced to 14 mm for the heavy ones. In the last case the vessel with the sample was placed in a cavity made in the jacket of the ionization chamber. The diameter of the vacuum window was much smaller than that of the ionization chamber electrodes and also 2 mm smaller than the diameter of irradiation vessel. At such arrangement all the ions going through the exit window were registered by the ionization chamber before they were stopped in the sample.

3. ABSOLUTE DOSE MEASUREMENTS

In the intermediate energy region (few MeV/nucleon) the radiative losses are negligible and, as a rule, the ions are stopped in the samples. In such conditions the overall energy imparted to the sample, energy input, E_{in}, can be calculated according to Eq. (1)

$$E_{in} = E_s \, S \, \Phi \tag{1}$$

where, E_s : is the ion energy at the surface (eV),
S : is the beam spot (cm^2), and
Φ : is the fluence (cm^{-2}).

and the mean dose, D_{av}, according to Eq. (2)

$$D_{av} = E_s \, \Phi \, (r)^{-1} \, (\rho)^{-1} \tag{2}$$

where, r : is the range of the ion (cm), and
ρ : is the density of the sample (g cm^{-3}).

So, to determine the mean dose we had to measure fluence and energy on the surface of the sample. Ranges and densities were taken from literature.

3.1. Energy measurement

According to the technical properties of U-400 cyclotron, the particle energy corresponding to the maximum intensity of the ion beams is strictly defined (for the set parameters) and the half width of the peak is about of 1% of the energy value. However, there are some sources of uncertainty, also. First of all the tuning parameters allow to change the energy (at the same mode) within the range ±5%. Further, ion beam extraction performed via charge exchange in stripping foils results in 2 or 3 ion beams with a little different (2–5 MeV) energies and different charges. Theoretically it is possible to calculate energy in vacuum, E_o, but it is better to measure it. An additional benefit from energy measurements is an assurance that we are really working on the selected ion.[2]

The simple and attractive idea of energy control by the measurement of the magnetic field inside the bending magnet had to be rejected because of technical reasons. Instead, a semiconductor detector (situated in the arm of the vacuum pipe) has been used. It measured the energy of the ions scattered in the Au-foil. The signals produced in the detector were pre-amplified and sent across the biological shielding to an amplifier, a multi-channel analyzer and computer. The detectors (Si surface-barrier or Si/Li) were calibrated by α-particles from ^{212}Bi (E_α = 6.090 MeV, $t_{1/2}$ = 60.6 min) and ^{212}Po (E_α = 8.78 MeV, $t_{1/2}$ = 0.3 ms). The source was prepared just before the experiment. Scattering angle and energy losses were taken into account when evaluating the ion energy in the beam pipe.

[2] The ions with the same rigidity are accelerated together and may be difficult to judge between for instance $^{16}O^{2+}$ and $^{24}Mg^{3+}$ without the energy measurement.

The energy on the surface of the sample, E_s, was calculated as E_o minus energy losses in the target system. The losses were calculated on the base of stopping power data taken from Refs [1, 2] or computer programs [3, 4]. As it is seen from Table I the energy losses for the ions depend very much on the specific energy and on the mass of the ion. For nearly the same target system (Ti exit window, air, electrodes of ionization chamber, thin layer of air) energy losses changed from 11 MeV for 113 MeV ^{11}B up to more than 200 MeV for 397 MeV ^{59}Co. Thus, the overall accuracy of E_s determination was different for different ions. Two factors, the accuracy of the stopping power data and the decrease of energy on the way from vacuum to the sample, had to be taken into account.

For the light ions, all sources of the stopping power data gave practically the same results. This and the small energy loss on the way to the sample led us to the conclusion that the energy of the light ions on the surface of the sample can be determined with the accuracy ±2 to 3 %. For heavy ions, the situation was worse. The discrepancies between the stopping power data (especially for air) reached 8-10% and the energy losses in the targets reached 50%. In such conditions the overall accuracy of the energy measurements was estimated to be ±5 to 10 %.

3.2. Fluence measurement

There are several of methods of fluence measurements. For low fluences ($<10^8$ ions per cm^2) and not very high fluxes ($<10^8$ pps) it is possible to use solid state track detectors, SSTD. For lower fluxes (10^6 pps or less) one can use thin ionization chambers working in the particle counting regime. For fluxes higher than 10^8 pps and fluences higher than 10^9 ions per cm^2 (ordinary conditions for chemical experiments) one can choose between the ion beam current collection [8-10] and fluence measurement with ionization chamber working in current registration regime.

The ion beam current collection method is more difficult than in the case of electron beams. It is because a charge of the ion changes from the initial value (vacuum conditions) up to zero. Nevertheless, this method was successfully used in many experiments [8-10]. Generally, it is more applicable to cyclotrons with electrostatic beam extraction system because only in such conditions the initial charge of the extracted ions is sufficiently known.

In our facility the fluence was measured by means of an air ionization chamber working in the current registration regime. The number of the ion pairs was proportional to the number of projectiles and their energy losses and inversely proportional to energy of ion pair formation in air, w_{air}. The chamber consisted of three parallel flat electrodes made of 5.6 μm thick Al-foil with a 0.48 mm air space, f, between them. A collar support sealed the electrodes and fixed the distance between them. The diameter of the smallest electrode was 6 mm larger than the diameter of the largest circular vacuum window. The chamber worked at ambient pressure. The potential of the outer electrodes with respect to the inner, collecting one was +250V, giving electric field strength of about 2.7 kV/cm. Such conditions ensured the full charge collection. The charge from the collecting electrode was measured using digital electrometer (P-100, P-Firm, Poland) with the accuracy of 0.5%.

The number of ions that passed through the ionization chamber and stopped in the sample was calculated from Eq. (3)

$$\Phi = w_{air}\, Q\, (\Delta E)^{-1}\, (S)^{-1}\, (e^-)^{-1} \qquad (3)$$

where, Φ : is the fluence (cm^{-2}),
Q : is the charge collected from signal electrode (C),
ΔE : is the energy deposited between the electrodes of ionization chamber (eV),
S : is the beam spot (cm^2) and
e^- : is the electron charge (C).

And, ΔE was calculated from Eq. (4)

$$\Delta E = \rho_{p,T} \; [(dE/dm)_1 + (dE/dm)_2] \; f \qquad (4)$$

where, $\rho_{p,T}$: is the pressure and temperature dependent air density (g cm^{-3}),
f : is the thickness of the air gap between two electrodes (cm),
$(dE/dm)_1$: is the mass stopping power in the first gap (eV g^{-1} cm^2) and
$(dE/dm)_2$: is the mass stopping power in the second gap (eV g^{-1} cm^2).

It should be mentioned that the accuracy of the fluence determination in this method is doubly dependent on the accuracy of the mass stopping power data: apparently (Eq. 4) and non-apparently (calculation of energy of the ions reaching the ionization chamber). According to the considerations in the section 3.2 the overall accuracy of fluence determination will be much higher for the light ions than for the heavy ones.

The second source of uncertainty is w_{air} value. It is well known for electrons [11] and sufficiently for protons [11], alpha particles [11] and carbon ions [12]. For heavy ions the experimental data are scarce and scattered. Taking into account state of affairs we decided to calibrate our ionization chamber directly on the solid state track detectors. Thin polyethylene terephtalate foils have been used as SSTD. The foils were situated at the place of the sample and irradiated with uniform, low intensity (10^7–10^8 pps) ion beam. The charge collected from ionization chamber was measured simultaneously. Then, the irradiated foils were etched in 6M NaOH and analyzed on JSM-840, JEOL electron microscope. From such experiment we could calculate the coefficient k equal to the charge generated inside the ionization chamber by one heavy ion.

$$k = Q \; (N)^{-1} \; (S)^{-1} \qquad (5)$$

where, N : is the number of tracks per cm^2 (cm^{-2}).

Using this calibration, no data about w_{air} and stopping power data were necessary for fluence determination. Additionally, the k coefficients enabled us to calculate unknown w_{air} – values of heavy ions if ΔE-values are known from elsewhere. The reasonable values of w_{air} obtained for ^{40}Ca and ^{59}Co [13] confirmed the dosimetry technique described in this work. Another confirmation of our ion beam dosimetry system was presented in [14]. Dose and energy dependence of the integral yield of Fe^{3+} formation in Fricke solution bombarded with ^{12}C ion beam was measured and compared with the data from three other laboratories. An agreement was satisfactory.

4. SECONDARY DOSIMETERS

We checked four dosimetry systems: a) Fricke dosimeter, b) ethanol solution of nitryl of malachite green, c) L-α-alanine, and d) standard bone powder. Transparent and dyed PVC foils were successfully used as a beam profile indicators at the time of tracing of the beam (Fig.1c).

Each of the investigated dosimetry system has its own advantages and shortcomings. EPR dosimeters (with alanine or standard bone powder as dose-sensitive material) can be used successfully for the total fluences in the range 10^8-10^{10} ions per cm^2. For the higher fluences, the signal-to-dose curve saturates because of overlapping of the latent tracks and the radiation destruction of previously created radicals [15]. The interesting feature of alanine is its sensibility to the LET of ionizing radiation. It reveals in the characteristic changes of the shape of EPR signal, especially on the borders [16] but also in power saturation characteristics of the central line [17].

On the base of literature data [8-10] and our own experience we consider that Fricke dosimeter can be useful for high-LET dosimetry. NaCl supplement is necessary. Without it, the contamination of

Fricke solution with organics will give a substantial increase in $G(Fe^{3+})$ values in stirred solutions but a decrease or only slight increase in non-stirred ones [18].

Ethanol solutions of malachite green can be used without stirring (which is very convenient) and for high doses. Additionally, they immediately deliver an initial information about the dose from the colour of irradiated sample. The main disadvantages of this dosimeter are sensitivity to UV light and volatility of solvent.

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