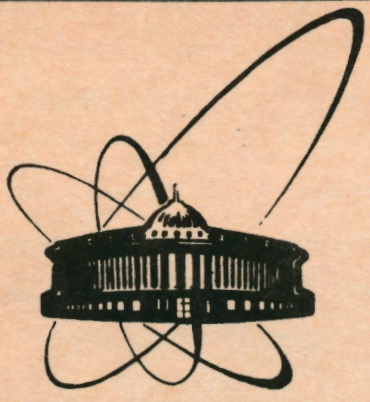

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A FACILITY FOR LIQUID-PHASE RADIATION EXPERIMENTS ON HEAVY ION BEAMS

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1. Introduction

The structure of the tracks produced by heavy charged particles appreciably differs from that of the electron tracks. Despite our knowledge of the general aspects of this structure (Allen 1961, Pikayev 1986), much more experimental work is needed to understand the high-LET radiation phenomena deeply enough. The results might be of interest not only for the field of radiation chemistry but also for some topics in technology, radiobiology, and nuclear and cosmic medicine.

Heavy ion accelerators are most prospective radiation sources for such studies as they provide particles of various masses and energies, hence, of very different LETs. Although the first radiation works on ion beams were published already in the 50-ties (Schuler 1957, Garrison 1957, Schwarz 1959) their total number is not large; see (LaVerne 1981,1988) for bibliography. It is so, first of all, because of a limited access to appropriate accelerators, which are expensive and dedicated mostly to physical research. Last years the situation is seen to be changing. Some technological problems have stimulated the investigation of ion beam induced radiation processes in polymers (Kouchi 1989, Apel 1990,1991, Schnabel 1991, Sasuga 1991, Balik 1987, Ingemarsson 1989, Vannikov 1990), and the use of high-LET radiations for the cancer therapy revived interest in the radiation chemistry of liquids, especially of aqueous solutions.

It has been manifested by a series of works dealing with the radiation phenomena near the Bragg peak (La Verne 1983,1985a,1987a,1989) and in the intermediate energy region (Jayko 1976, LaVerne 1985b, 1986ab,1987b), with ion beam pulse radiolysis (Sauer Jr. 1977, 1978ab) and with the effects of GeV-energy ion beams (Christman 1981, Appleby 1885,1986, Schimmerling 1976). Theoretical works have been performed in parallel (Magee 1980, Chatterjee 1980, Mitierev 1987, Waligorsky 1986, Katz 1986, Miller 1989). The problems connected with the non-absolute dosimetry of high-LET radiations have also been considered (Hansen 1984,1986, Olsen 1984).

Recently we started liquid-phase radiation experiments at Dubna. It was made possible since one of the beam lines of the 4-meter U-400 cyclotron was dedicated to chemical research. The vertical direction of this beam pipe provides favorable conditions for the irradiation of liquids. The cyclotron is working in the intermediate energy region, in which only few radiation works have been done to date. In the present work we describe our facility and give some preliminary results on the radiolysis of two liquid systems by three different ion beams.

2. Ion beam characteristics

The U-400 isochronous cyclotron is capable of accelerating the ions with mass to charge ratio from 5 to 12. Presently some 30 different particles from ^{7}Li to ^{129}Xe are accelerated (Gikal 1989,1991). The maximum energy per nucleon ranges from 18 MeV for lighter ions to 3.8 MeV for ^{129}Xe. A high-current internal

PIG-type source allows to obtain intense ion beams from both gaseous and solid feed materials. The ion beams are extracted via charge exchange in thin (40-200 $\mu g/cm^2$) graphite foils. The charge of the accelerated ions is relatively low, from +1 to +12, but after passing the extraction foil it increases 2.5 to 4.5 times. This way the lighter ions can be obtained completely stripped. The intensity of the external beams reaches 10^{13} pps for the light ions and 10^{9} pps for the heaviest ones. It can be easily attenuated to a required value.

The energy of the ions produced by the U-400 cyclotron depends on the "extraction mode" and can be changed by steps (Gikal 1990). As it is seen from Fig.1, switching from one mode to another changes the energy by 30-40%. At a fixed extraction mode, the energy can be changed smoothly within ±5% around the value corresponding to the maximum intensity for each charge. The energy of the extracted beams can be reduced by metallic foils. This, of course, broadens the energy spectrum; see Fig.1, left upper insert. In not degraded beams the FWHM of the peak is about 1% of the total energy value. The U-400 ion beams have a time structure: there is a train of 1.5 ms pulses separated by 5.1 ms intervals, and each such pulse consists 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 true dose rate is ≈40 times higher than the total dose divided by the time. On the other hand, the high intensity of the U-400 beams and the time structure might provide good conditions for employing the pulsed radiation techniques.

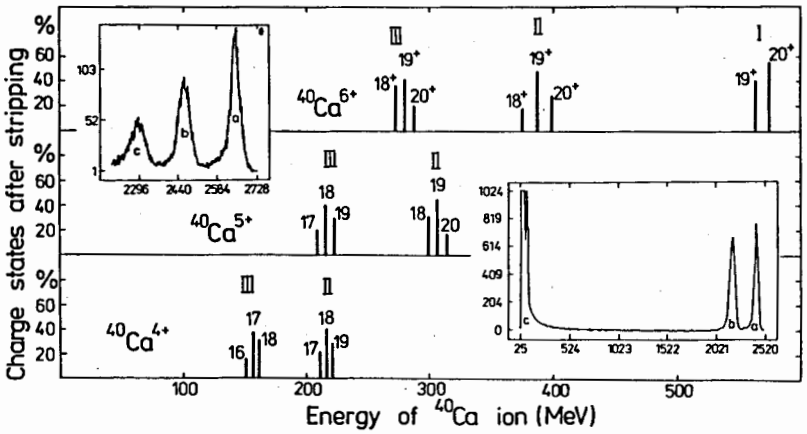

Fig. 1. Calculated energies of ^{40}Ca ion beams in U-400 cyclotron; from (Gikal 1990).

The lines show the energy for which the maximum beam intensity is expected. Charges of the ions being accelerated are indicated. Roman numerals denote the extraction mode.

Left insert: Energy straggling of the primary ^{40}Ca ion beam (a) after passing through 8.5 μm Al-foil (b) and through 17 μm Al-foil (c).

Right insert: Demonstration of the purity level of the beam: (a) - initial ^{40}Ca ion beam, (b) - after passing 8.5 μm Al-foil (support for alpha source), (c) - calibration peaks. No peaks are seen between those in (a) or (b) and in (c).

3. The facility for radiation experiments

U-400 is equipped with 12 beam lines positioned at three levels. The beam post of the chemical channel and laboratory rooms are situated at the basement; see Fig.2. The horizontal part of the pipe is about 8 m long, the vertical one - about 10 m. Two pairs of magnetic lenses, a 90° magnet, a pair of steering magnets, and three diagnostic loops allow to focus and guide the beam. The third diagnostic loop, situated right in the chemical room, is equipped with a shutter (to cut-off the chemical equipment installed on the channel), a Faraday cup (for flux measurements), a BeO-luminophor (for visual beam profile control), and a scattering foil (for energy measurement). The latter foil (0.2 μm Au) is introduced by an electric drive, the other devices - pneumatically.

In radiation experiments we usually work on wide, defocussed beams to ensure uniform intensity over all the exit window area. Four types of the vacuum-tight windows have been used:

- a 2×50 mm^2 slit-window with 8.5 μm Al-foil to uniformly irradiate large objects passing under the slit on a conveyer;
- a round, ø = 40mm window with 8.5μm Al-foil, backed by a perforated (ø = 2 mm) support with 75% transparency;
- a round, ø = 8 mm, 4.3 μm Ti-window;
- a round, ø = 18 mm, 4.3 μm Ti-window.

Because of the small thickness of the exit windows and rather

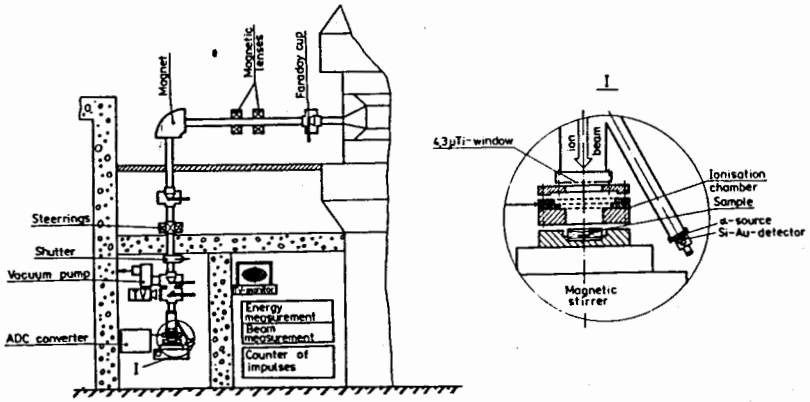

Fig. 2. Overall arrangement of the irradiation post.

Table 1. Experimental conditions

Ion	E_c	E_c/A	E_o	E_o/A	Range	LET_{init}	Beam intensity × 10^{-8}
	MeV	MeV/amu	MeV	MeV/nucl.	mm	eV/A	ion $cm^{-2}s^{-1}$
^{11}B	110	10.0	96	8.7	0.53	12	2 - 3
^{24}Mg	181	7.5	138	5.7	0.10	86	2 - 5
^{40}Ca	299	7.5	177	4.4	0.06	213	0.2 - 0.9

E_c - energy of the ion in cyclotron, E_o - energy of the ion at thesurface of the solution, A - mass number of the ion. Ranges and LET_{init} are taken from the N-S data tables for water.

low fluencies in experiments (cf. Table 1), cooling of the system has been unnecessary.

The vacuum in the cyclotron chamber is 1×10^{-4}Pa - 6×10^{-5}Pa (10^{-6}- 5×10^{-7} Torr), and about 1×10^{-3}Pa (10^{-5} Torr) in the pipe. After the vacuum window, a thin-walled ionization chamber is placed as the fluence monitor. It is isolated from the pipe by a plexiglass mount.

4. Irradiation conditions

The solutions were bombarded in flat cylindrical vessels made of glass or teflon. It was carefully verified that the materials do not influence the results. The diameters of the vessels were, as a rule, by 2 mm larger than the diameters of the vacuum windows. In all the cases the window, the ionization chamber, and the vessel were strictly centered. The volume of the liquid, v, was 5 or 3 ml and the thickness of its layer, d, was 2-9 mm. This ensures the complete stopping of ions in the solution and permits using a magnetic stirrer for thicker layers. Most of the experiments were done using the 18 mm Ti-window, with v = 3 ml and d = 9 mm. In contrast to other radiation works on ion beams, the liquid samples were bombarded from the liquid surface side, which was exposed to air. This preserves a high oxygen concentration in the zone under irradiation. The samples were irradiated in two different positions. In the first one, the vessel was placed into a cavity made in the jacket of the ionization chamber, as close as possible to the vacuum window.

The air and heat flows were hindered a little. The solutions were air-saturated; they were not stirred. In the second position, the vessel was put on the plate of a magnetic stirrer (see Fig.2, insert), and the distance between the ionization chamber and the solution surface was 20-30 mm. The solutions were irradiated with and without stirring.

A single experiment typically lasted from several dozen of seconds to a few minutes. Some 50 - 80 of such bombardments, with a few minute intervals between them, were usually performed in series ("a run").

5. Dosimetry

Under our experimental conditions, ions are completely stopped in the solution and the radiative losses are negligible. If such, the energy absorbed in the object, the energy input, E_{in}, can be calculated as the product of the ion energy at the surface of the solution, E_o, and the total fluence, ϕ_{tot}. Each of the factors was measured separately.

5.1. Energy measurement

As mentioned above, the particle energy corresponding to the maximum intensity of the ion beam is strictly defined. Nevertheless, it is possible, at the expense of the intensity, to change the energy in some range. To known the actual, accurate value of the energy, this is always measured by a semiconductor

detector situated in an arm of the pipe; see Fig.2, insert. The signals produced in the detector by the ions scattered in the Au-foil (see above) are preamplified and sent across the biological shielding to the laboratory room where an amplifier, a multichannel analyzer, and a computer are operated. According to the type of the ion and its energy, a Si surface-barrier or a Si/Li detector have been used.The calibration of the detectors has been performed by α-particles from ^{212}Bi(E_α=6.090 MeV, $t_{1/2}$=60.6 min) and from ^{212}Po(E_α= 8.78 MeV, $t_{1/2}$=0.3 ms), which was in equilibrium with the parent ^{212}Pb ($t_{1/2}$= 10.64 h). The source was prepared just before the experiment. The scattering angle and the energy losses in the Au-foil were taken into account when evaluating the ion energy in the beam pipe, E_c. Reproducibility of the energy measurement was 1% or better, but the final accuracy, at the moment, is estimated as 2-3%. The energy value was measured several times during a run; we have not observed fluctuations larger than 1-2%.

The energy E_o, was calculated as E_c minus the energy losses in the target system: in the vacuum window (1.94 mg/cm^2 Ti or 2.30 mg/cm^2 Al), in the ionization chamber (1.51 mg/cm^2 Al, 0.48 mm of air, 1.51 mg/cm^2 Al, 0.48 mm of air, 1.51 mg/cm^2 Al), and in 30 mm of air above the solution. The influence of atmospheric conditions (p, T) on the air density was taken into account.

5.2. Total fluence measurement

The most frequently used method is based on the beam current

collection (Schuler 1955). However, this is better applicable at the accelerators with electrostatic beam extraction systems. In our arrangement we measured the total fluence by using an air ionization chamber working in the current regime. This technique has been used for many years in radiobiological experiments conducted on JINR cyclotrons (Tcherevatenko 1989). There are three parallel flat electrodes from 5.6 μm Al-foil with 0.48 mm air space, *l*, between them. A collar support allows to seal the electrodes and to strictly fix the distance between them. The diameter of the smallest electrode is 46 mm, *i.e.* by 6 mm larger than the diameter of the largest round vacuum window. The chamber works at the ambient pressure. The outer electrodes are at a potential of +250 V vs. the inner, collecting one. It gives an electric field strength of about 2.7 kV/cm, what ensures complete charge collection. The current from the collecting electrode is fed to an analog-to-digital converter (Zinov 1971) with the coefficient k=1.00 nC/pulse ; this device is placed some 50 cm from the chamber. The digital signal is sent across the biological shielding to a counter equipped with a timer. Taking into account the literature values of the stopping power in air, dE/dm, and of the energy of ion pair formation in air, W_{air}, we calculated the total number of ions which passed through the ionization chamber and stopped in the solution from the formula

$$\phi_{tot} = \frac{knW_{air}}{\rho_{p,T} \cdot 2l \cdot dE/dm} \quad , \qquad (1)$$

where n is the number of pulses from the A/D converter, and $\rho_{p,T}$ is the pressure and temperature dependent air density. Since

the energy loss in the ionization chamber is calculated basing on the stopping power of the ions under operation, the purity of the ion beam is very important. The high enough mass resolution of the U-400 cyclotron (Gulbekian 1987) and the variety of the ion extraction modes (Gikal 1990) allow to make a good option. The method of energy measurement offers an independent test of the ion beam purity; see Fig.1,right insert.

Besides its advantages, the employed method of the total fluence measurement has also a disadvantage. It is a loss of ion energy in the chamber, especially large for projectiles with high atomic numbers and lower energies. However, our vacuum window is thinner than the vacuum windows in other works, and no vessel's entrance window is needed. So, finally, the incoming energy at the liquid surface is comparable with that in the works of other authors.

6. Results and discussion

To test the performance of our facility, we made experiments with three ion beams very different in physical properties. Their characteristics and the conditions of experiments are given in Table 1.

6.1. The investigated systems and raw experimental data

Two liquid dosimetric systems were irradiated: the Fricke solution (1×10^{-2}M Fe^{2+}, 4×10^{-1}M H_2SO_4, air saturated H_2O) and

malachite green (MGCN) radiochromic dosimeter [1×10^{-3}M 4.4'-bis (N,N'-dimethylamino)-triphenylmethane cyanide, 1×10^{-3}M HCl, air saturated 95% ethanol]. The value of $G(Fe^{3+})$ and the radiation yield of the malachite green cation (MG^+) formation have been evaluated from optical density measurements at 302 nm (ε_{302}=2200 $M^{-1}cm^{-1}$ at 25°C) and 622 nm ($\varepsilon_{622}=1.04\times10^5$ $M^{-1}cm^{-1}$), respectively. The influence of temperature (-0.69%/°C) on the molar absorption coefficient of Fe^{3+} was taken into account. The two systems were investigated in parallel, *i.e.* in each run both of the solutions were irradiated and analyzed.

The Fricke solution is the system recommended many years ago as a secondary dosimeter for low-LET radiations. There are also data about its radiolysis by high-LET radiations (LaVerne 1987 and references therein), among them by light ion beams (Schuler 1957,1967, Imamura 1970, Matsui 1970, Jayko 1976, LaVerne 1983,1987). Radiolysis of the Fricke solution leads to the oxidation of ferrous ions to the 3+ state according to the well known radiation scheme (Allen 1961, Pikayev 1986). *G*-value of Fe^{3+} formation in air or O_2-saturated solutions can be expressed as:

$$G(Fe^{3+}) = 3G_H + G_{OH} + 2G_{H_2O_2} + 3G_{HO_2} \qquad (2)$$

where G_H, G_{OH}, $G_{H_2O_2}$, G_{HO_2} are the yields of H, OH, H_2O_2, and HO_2 species which escape from the track. In high-LET radiation tracks the probability of radical-radical reactions strongly increases, while the probability of Fe^{2+} oxidation decreases. So, the observed lower $G(Fe^{3+})$ values for high-LET radiations give

us some insight into early time radiation processes in dense tracks in water matrix.

The radiolysis of alcoholic solutions of malachite green is not known so well, nevertheless, many aspects of it have been revealed lately (Bobrowski 1985, Stuglik 1986a,b, Grodkowski 1989,1990,1992). It has been reported that in the solutions irradiated by electrons, MG^+ formation strongly depends on O_2 concentration. In deoxygenated solutions, MG^+ does not appear as a stable product. Small concentrations of oxygen (10^{-6}-10^{-5}M) initiate a mechanism of MG^+ formation through $MG^{\cdot}$ oxidation lasting hundreds of microseconds (Grodkowski 1992). In aerated solutions this mechanism does not operate, and MG^+ is generated in much slower processes, probably in the reactions between MGCN and peroxy-radicals from the alcohol. $G(MG^+)$ is rather low, about 0.1; it is dose-proportional up to hundreds kGy. To our best knowledge, the malachite green has not been investigated on ion beams or with other high-LET radiations, but there are some data on ion beam irradiated radiochromic films containing a related thriphenylmethane dye (Olsen 1984, Hansen 1984).

The results of ion beam radiolysis of the investigated solutions are presented in Figs. 3 & 4. In the case of Fricke solutions, we pay attention to that, in contrast to the previous works (Schuler 1957,1967, Matsui 1970), stirring the solutions does not influence significantly Fe^{3+} production. In all the cases the effect is not higher than 5 - 15%. The discrepancy is probably due to different irradiation conditions. All the previous works were performed on horizontal ion beams, and ions

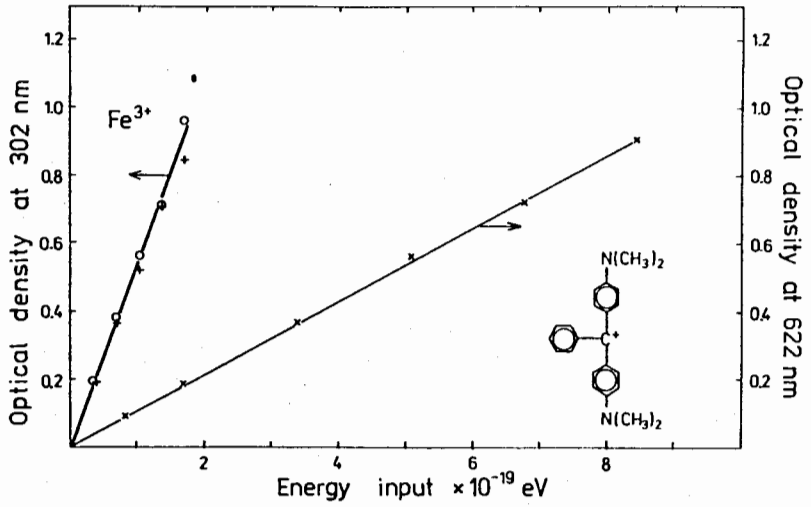

Fig. 3. Dose dependence of Fe^{3+} and MG^{+} generation in solutions irradiated by 96 MeV ^{11}B ion beam.
o - with stirring, + - without stirring, × - with and without stirring (no difference observed).

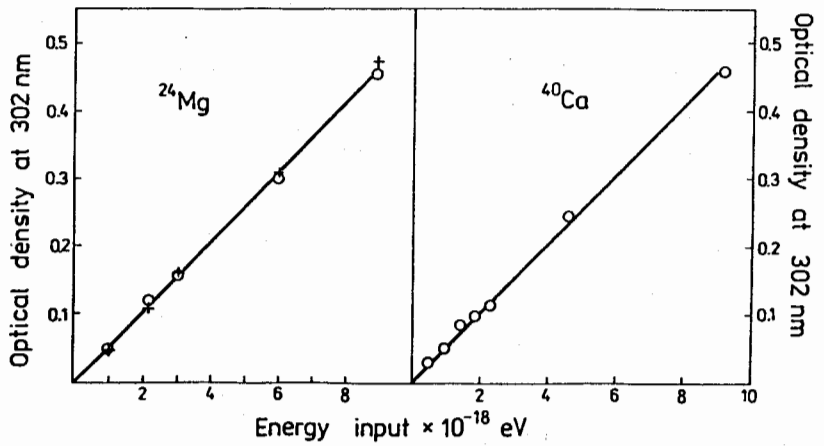

Fig. 4. Dose dependence of Fe^{3+} generation in unstirred Fricke solutions irradiated by ^{24}Mg and ^{40}Ca ion beams.

were injected into the solution from the wall-liquid interface. Moreover, the beams were usually focussed on a small spot. Both the factors, together with short ranges of the ions in the solution, obviously caused prompt depletion of the O_2 concentration in the irradiation zone adjacent to the vessel entrance window. Correct and reproducible results could be achieved in those works only with vigorously stirred solutions.

In our set-up, solutions are bombarded from the gas-liquid interface. So the irradiated zone is in contact with the air-saturated solution from the bottom and with air from the top. Because of that and due to low fluences, the reduction of O_2 concentration in the irradiated zone is rather small. As a consequence, our $G(Fe^{3+})$ values are very similar in stirred and unstirred solutions.

Another feature observed in the Fricke system is a significantly broader dose range in which the effect is dose-proportional. This probably results from small $G(Fe^{3+})$ value specific to high-LET radiations and from the easy supply of oxygen to the irradiated zone as discussed above. MG^{+} production proportionally increased up to at least 16 kGy, and no influence of stirring has been observed.

6.2. Energy input uncertainties

When trying to evaluate G-values, we encountered considerable difficulties because of the lack of accurate pertinent data on stopping powers and mean energies of ion pair formation, while both quantities are necessary for dose determination.

Stopping powers

There have been published several calculational programs and tables of stopping powers, which cover broad ranges of the atomic numbers, Z, of projectiles and targets, and of the bombarding particles energies. Generally, the data for projectiles with low Z are in agreement, but for heavier ions increasingly larger differences are found. For instance, the widely used Northcliffe and Schilling (N-S) tables (Northcliffe 1970) agree quite well with the recent data of Hubert *et al.* (Hubert 1990) for projectiles up to Z = 10, but already for Z = 20 the discrepancy is as high as 10-15% for light targets. Our experimental results on ^{40}Ca ions stopping powers in Al (to be published separately) are in better agreement with the Hubert *et al.* tables than with the N-S ones. Unfortunately, the Hubert *et al.* tables are limited to solid stopping media, while for our aims the stopping power of air is also required. The program STOPPOW (Henninger 1988) gives stopping power values between those of N-S and Hubert *et al.*, approaching the latter data in a higher energy region. So in the case of ^{40}Ca and ^{24}Mg we decided to use the Hubert *et al.* data for aluminium and titanium, and the program STOPPOW to calculate air stopping powers. The data for ^{11}B have been taken from the N-S tables as for these ions the program STOPPOW and both the mentioned tables give essentially the same results.

W_{air}-values

At present, W_{air} values are accurately known only for high energy electrons (Boutillon 1987), for 1.8 MeV protons (ICRU 1979), and for 5.3 MeV alphas (ICRU 1979). These energies (in eV) are 33.97±0.05, 35.1, and 35.18, respectively; the ICRU data are the recommended values. To our best knowledge, there has been only one work (Liesem 1981) with heavy ions to obtain W_{air}=37.9±2.0 eV for 3.2 MeV/nucleon ^{208}Pb and W_{air}=37.3±2.5 eV for 4.2 MeV/nucleon ^{238}U, which is close to W_{N_2}=38.05 eV reported by Varma and Baum (Varma 1982) for 2.6 MeV/nucleon ^{16}O ions and to the values calculated from a semi-empirical formula proposed by Zielczynski (Zielczynski 1988). But the situation is not clear as Liesem based his calculations on the N-S stopping power tables, which are probably incorrect for high-Z projectiles.

The data obtained for the stopping media other than air show that W values are LET dependent - they increase with it (ICRU 1979). Taking into account that the LET for 10 MeV/nucleon ^{11}B ions is nearly the same as that for 5.3 MeV α-particles, we took W_{air}=35 eV in dose calculations for ^{11}B ions. the LET of ^{24}Mg ions is higher (see Table 1) but still much lower than the LET of the projectiles investigated by Liesem; so we chose W_{air}=35.5 eV for ^{24}Mg. In the case of ^{40}Ca ion beams, the fluence was measured directly by calibrating the ionization chamber with the use of solid state track detectors (Fleischer 1975) for counting the number of ions which passed through the chamber. So no assumption about W_{air} was needed.

6.3. Evaluation of G

The energy input (in eV) obtained with the options discussed above was then utilized for G_o-value calculation according to the formula

$$G_o(P) = \frac{OD_P \cdot vN_A \cdot 100}{E_{in}\,\varepsilon_P} \quad \text{molecule / 100 eV,} \qquad (3)$$

where $G_o(P)$ is the integral value of the radiation yield of the product "P" (Fe^{3+}or MG^{+}) for the energy region from 0 to E_o, OD is the optical density of the product in the solution absorption spectra, ε_P is the molar absorption coefficient (see sect. 6.1.), and N_A is Avogadro number.

$G_o(MG^{+})$ for 95 MeV ^{11}B ions was calculated to be 0.019±0.001, which is about 4 times lower than that observed for 250 kV X-rays (Stuglik 1988, unpublished). $G_o(Fe^{3+})$ values for ^{11}B, ^{24}Mg, and ^{40}Ca ion beams are given in Table 2. The comparison of our and some literature data is presented in Fig.5. There seems to be a reasonably good agreement in the energy range (up to 100 MeV) where the comparison is possible. The uncertainties in the G-values, including the possible systematic errors, are estimated to be 5% for ^{11}B and ^{40}Ca ion beams, and 10% for ^{24}Mg. A considerable contribution to these error bars results from the lack of reliable data on W_{air} and on stopping powers (see above). To obtain more accurate values, we plan to work with beams of higher energies and to make the air space

between the vacuum window and the surface of the solution as small as possible. We also intend to employ the calibration of the ionization chamber by track detectors as a routine method.

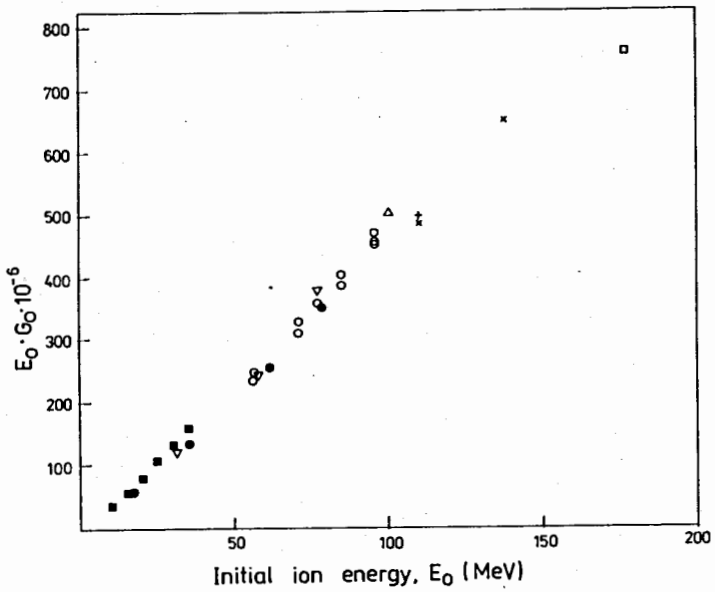

Fig. 5. Energy dependence of $E_o G_o(Fe^{3+})$ values for Fricke solution irradiated by ion beams
o..^{11}B (this work, stirred), ▪..^{11}B (La Verne1987), Δ..^{12}C (Schuler 1967), ∇..^{12}C (Imamura 1970), ●..^{14}N (Imamura 1970),
×...^{24}Mg (this work, stirred), +..^{24}Mg (this work, unstirred), ▫....^{40}Ca (this work, unstirred).

Table 2. G-values of Fe^{3+} formation in ion-beam irradiated Fricke solution

Ion	E (MeV)	$G(Fe^{3+})$ molecule/100 eV
^{11}B	96	4.8 ± 0.1
	85	4.6 ± 0.1
	71	4.4 ± 0.1
	56	4.2 ± 0.1
^{24}Mg	138	4.7 ± 0.2*
	131	4.5 ± 0.2*
	110	4.4 ± 0.2
^{40}Ca	177	4.6 ± 0.1*

* without stirring

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Установка для жидкофазных радиационно-химических экспериментов на пучках тяжелых ионов

Описана установка для радиационно-химических экспериментов на вертикальном отводе 4-метрового циклотрона У-400 в Лаборатории ядерных реакций им. Г.Н.Флерова, ОИЯИ, Дубна. Ускоритель производит пучки ионов средних энергий (около 10 МэВ/нуклон) от ионов Li до Xe. Представлены первые результаты по радиолизу двух жидких систем — раствора Фрике и раствора малахитового зеленого в этаноле под пучками ионов ^{11}B, ^{24}Mg и ^{40}Ca. Обсуждаются экспериментальные проблемы и неопределенности, возникшие при количественной оценке экспериментальных данных.

Работа выполнена в Лаборатории ядерных реакций ОИЯИ.

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A Facility for Liquid-Phase Radiation Experiments on Heavy Ion Beams

The title facility is described, installed on a beam line of the 4-meter U-400 cyclotron in the Flerov Laboratory of Nuclear Reactions, JINR, Dubna. The accelerator provides intermediate energy (some 10 MeV/nucleon) beams of ions from Li to Xe. Preliminary results on the radiolysis of two liqyid systems — Fricke solution and malachite green in ethanol — by ^{11}B, ^{24}Mg, and ^{40}Ca ions are presented. Some experimental problems and uncertainties faced at the quantitative evaluation of the data are discussed.

The investigation has been performed at the Laboraty of Nuclear Reactions,JINR.

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