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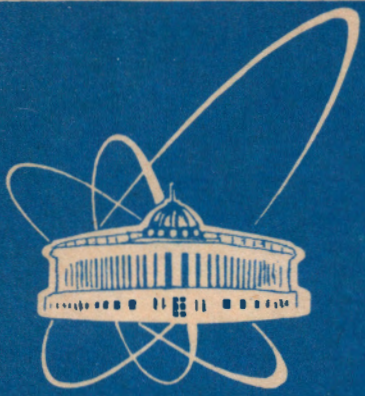

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RADIATION FACILITY AT THE JINR U-400 CYCLOTRON CHECKED BY FRICKE DOSIMETER MEASUREMENTS

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Introduction.

Ion beam radiation experiments with liquids, in spite of their scientific importance, have been done rather seldom (see [1, 2] for bibliography) mainly because of experimental difficulties and limited access to appropriate accelerator facilities. Three years ago such experiments were started [3] at the 4-meter isochronous cyclotron U-400 at the Flerov Laboratory of Nuclear Reactions, Joint Institute for Nuclear Research, Dubna using the KHIPTI facility. The latter consists of two parts, dedicated to radiochemical or radiation research. Unlike most other radiation facilities, ours has been performing at a vertical ion-beam pipe, which allows to irradiate liquid and powder samples in open vessels [3-6]. Above all, it gives possibility to investigate high-LET radiation processes running in thin layers of liquids exposed to air [4].

It is well-known that the ion-beam radiation experiments are not only more difficult than the low-LET ones, but also more susceptible to systematic errors. Having this in mind, we decided to test our facility and techniques by repeating some experiments reported in literature and comparing the results. Quite a few experimental data are available for acidic 0.01 M ferrous sulfate aqueous solutions saturated with air or O_2 and irradiated by ^{12}C ion beams [7 - 11]; this is why we have chosen this system for study.

Experimental.

The U-400 cyclotron is capable of accelerating ions with mass to charge ratios of 5 ÷12 to energies of 3 ÷15 MeV per nucleon. Presently more than 30 particles ranging from ^{7}Li to ^{129}Xe are available. The beams are extracted via charge exchange in thin graphite foils. Relatively low charges of the ions being accelerated increase 2.5 to 4.5 times after passing through the extraction foil [12]. For each kind of particles, several values of energy are attainable directly from the cyclotron [13]. Other energies can be obtained by degrading the beams in metallic foils of known thickness.

The beams have a time structure: there is a train of 1 ms pulses separated by 5.1 ms intervals, while each pulse consists of a number of much shorter pulses with a duty factor of 10 and the frequency equal to that of the accelerating RF voltage (5÷12 MHz). In consequence, the dose rate for the millisecond pulses is 7 times, and for nanosecond ones about 40 times higher than that calculated as the total dose over the time.

In this work, we made experiments with carbon ions accelerated as $^{12}C^{2+}$ and extracted as $^{12}C^{6+}$. Aluminum foils were used for energy degradation. The Fricke solution (0.01 M Fe^{2+}, 0.4 M H_2SO_4, air saturated H_2O) was prepared from triple distilled water (second distillation over potassium permanganate) and from the ammonium iron(II) sulfate hexahydrate recrystallized twice from acidified aqueous solutions bubbled by argon. The last operation reduced the Fe^{3+} concentration below the detection limit of a spectrophotometric method. Samples of the dosimetric solution were irradiated in flat cylindrical dishes of glass or PTFE. It was checked that the vessel's material does not influence on the results. During irradiation the samples were vigorously stirred by a magnetic stirrer. Optical absorption density was measured at λ=304 nm with the Specord 40M UV/VIS spectrophotometer, and the ferric ion concentration was calculated from the Lambert-Beer's law. The molar absorption

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coefficient ε_{304} = 2200 M^{-1} cm^{-1} (T = 298 K) and its temperature coefficient k = 0.0069/K were used in calculations. Irradiated solutions were measured against the non-irradiated ones. After bombardment but prior to analysis, the samples were kept in small closed bottles. The total energy imparted to the solution, the energy input, was calculated as a product of ion energy at the surface of the solution, E_S, and the total number of ions stopped in the sample, N. This was justified by the fact that all ions completely stuck in the sample and radiative losses at the indicated initial energy were negligible. The E_S-value was calculated as the ion energy in the channel, E_C, minus the energy losses in the target assembly: exit window (Ti, 2 mg/cm^2), 8 mm layer of air, ionization chamber (Al, 1.5 mg/cm^2; air, 0.48 mm; Al, 1.5 mg/cm^2; air, 0.48 mm; Al, 1.5 mg/cm^2) and 28 mm layer of air. The E_C-value was defined by accelerator parameters but it was also measured by a Si/Li semiconductor detector calibrated with α-particles of ^{212}Bi and ^{212}Po. The value of N was measured using an air ionization chamber working in a current regime. This technique has been used successfully for many years in radiobiological experiments conducted at JINR cyclotrons [14] and it seemed suitable also for our case. The chamber has three parallel electrodes with air gaps, l = 0.48 mm, between them. A collar support made of one piece of Lucite and also some springs allow to seal the electrodes and to fix strictly the distance between them. The chamber worked at ambient pressure. The outer electrodes were at a potential of + 250V vs. the inner, collecting one. The charge from the collecting electrode was measured using the P-100 (P-Firm, Poland) digital electrometer with an accuracy of 0.5%. For additional information on experimental conditions see Table 1.

Results and Discussion.

It is well established that irradiation of acidic ferrous sulfate solution leads to oxidation of ferrous ions to the Fe^{3+} state, and that the reaction pattern does not depend on the kind of radiation [15]. The ferric ion concentration increases linearly with dose in a wide dose range, and $G(Fe^{3+})$-value, the number of Fe^{3+} ions generated by 100eV of energy imparted, depends on the yield of primary products of water radiolysis as follows:

$$(1) \qquad G(Fe^{3+}) = G_{OH} + 3G_{H} + 3G_{HO_2} + 2G_{H_2O_2}$$

A maximum value of $G(Fe^{3+})$ equal to 15.6 ±0.1 is found in low-LET fields. In high-LET ones, $G(Fe^{3+})$-values are much lower. It results mainly from radical recombination in dense ionization regions leading to reconstruction of H_2O molecules or to synthesis of some other non-oxidizing species.

One of the peculiarities of high-LET radiolysis is a dependence of G-values upon energy which arises according to LET variation along the particle path (Fig.1, lower insert). So, two kinds of G-values, the differential, G', (instantaneous, thin target or G_i) and the integral, G_E , (thick target, G_0 or G) are used to describe radiation effects. They are related by the equation:

$$(2) \quad G'(E) = d/dE(G_E E_S).$$

For liquid samples bombarded with intermediate energy ions (penetration range < 1mm) in steady-state experiments, the only measurable value is G_E. In the present work, we measured $G_E(Fe^{3+})$-values for 0.01M ferrous sulfate solutions irradiated by ^{12}C ion beams having E_C = 133 MeV and E_S in the range from 120 to 36 MeV. As it is seen from Table 2, this energy range partly overlaps some of the ranges explored before. At the same E_S, optical absorption density at 304 nm, OD_{304}, increased linearly with the dose (Fig 1, upper insert) that pointed to stability of the $G_E(Fe^{3+})$-value in the explored range of energy inputs. Radiation yields of Fe^{3+} generation were evaluated using the formula:

$$(3) \qquad G_E(Fe^{3+}) = \frac{OD_{304} \cdot v \cdot N_A \cdot 100 \cdot e \cdot \Delta E_{2l}}{Q \cdot E_S \cdot w_{air} \cdot \varepsilon_{304}} .$$

where v is the volume of irradiated solution, N_A is the Avogadro number, e$^-$ is the charge unit, ΔE_{2l} is the energy deposited by ^{12}C ion in two air gaps between the electrodes of ionizing chamber, Q is the total charge collected by the electrometer during irradiation of the sample, w_{air} is the differential energy of ion pair formation in air irradiated by 10 MeV/amu ^{12}C ion beams, and ε_{304} is the molar absorption coefficient of the Fe^{3+} ion at 304 nm.

The results are presented in Table 2. Each $G_E(Fe^{3+})$-value was measured twice (the results agreed within 1 - 3%) and the mean values are given. Standard deviations determined earlier from 7 independent measurements for Fricke solution irradiated with ^{11}B and ^{25}Mg ion beams were 1.5% and 2%, respectively. The overall uncertainty of $G_E(Fe^{3+})$-value at the 95% confidence level, U, was calculated according to the IAEA recommendation for dosimetric systems [22]:

$$(4) \qquad U = \sqrt{z_i^2 + 1.13 z_j^2} ,$$

where z_i are the random errors and z_j are the systematic errors.

In eq. (3) there are three parameters, N_A , e , and ε_{304}, which are known accurately enough and three parameters, OD_{304} , v , and Q, which are measured with an accuracy of about 0.5%. However, for still three parameters involved , E_S, ΔE_{2l} and w_{air} , the situation is more complicated and is discussed below in detail.

E_C was measured with a precision of about 1%, while the uncertainties in energy losses depend on stopping power and target thickness errors and are proportional to the relative decrease in the ion beam energy $(E_C - E_S)/E_C$. Though we used a thin vacuum window and no cell window, the ^{12}C ion beam energy was reduced by about 10%, mainly in the ionization chamber mounted between the vacuum window and the sample. However, because stopping power values for light projectiles with intermediate energies are known with good accuracy and the energy loss is not very large, possible error of E_S determination seems to be negligible. The situation changes when the particle are additionally degraded in Al-foils and straggling strongly broadens their energy spectrum. In such cases, the experimentally measured $G_E(Fe^{3+})$-values present some averages for rather poorly defined energy ranges. If we ascribe

them to the peak energy in the spectrum , an overestimate takes place. It arises from $G'(Fe^{3+})$-value increasing with the energy. This situation is clearly seen from our data in Table 1. The $G_E(Fe^{3+})$-values pass through a minimum at ~50% energy degradation and then grow up; this, on the base of present knowledge, should be rather considered as an artifact. Because of that, only the G-values for two highest energies (initial energy degradation equal 10% and 21%) are recognized as correct.

The next parameter which should be discussed is the energy deposited by a ^{12}C ion in air, ΔE_{2l} , calculated as follows:

(5) $\Delta E_{2l} = dE/dm \cdot \rho_{p,T} \cdot 2l,$

where dE/dm is the ^{12}C ion beam stopping power in air, $\rho_{p,T}$ is the pressure and temperature dependant air density, and l is the distance between two adjacent electrodes. As said before, the stopping power values for ^{12}C ion beams seem to be accurate enough. The Northcliffe and Schilling's data tables [17], commonly used in the previous works, and the Stoppow-82 program [16] used in this work yield results which agree within 0.1%. Temperature and pressure were measured and, consequently, $\rho_{p,T}$ was known with an accuracy better than 1%. The $2l$ - distance uncertainty was ± 2%. The overall uncertainty of ΔE_{2l}-values was about 3 %.

The last parameter which should be discussed is w_{air}. It is accurately known only for high energy electrons [18], for 1.8MeV protons [19] and 5.3 MeV alphas [19]: (33.97±0.05) eV, 35.1 eV and 35.18 eV, respectively. To our best knowledge, there have been only two works [20, 21] in which differential energies of ion pair formation in air irradiated by heavy ions were determined experimentally. According to [19], the w_{air}-value for 6.7 MeV/amu ^{12}C ions is 36.2±1.0 eV and for 129.4 MeV/amu -- 33.7±0.9 eV. Based on this work, we took w_{air} for 10 MeV/amu as (36.1±1.0) eV, but because previously we used (35± 1) eV, we present the calculations for this value as well. As a result, $G_E(Fe^{3+})$-value were determined with U of about 5%.

Since $G_E(Fe^{3+})$ is a non-linear function of energy, it is better to compare ours and the previous results in a graph (Fig.1), in the form commonly used in such cases [11]. Here the data from Table 2 are presented except this of Jayko et al. for a solution saturated not with air but oxygen, and except our results for the lower energies (the reasons were discussed above). One can see that the experimental points follow a smooth line which can be interpreted as a satisfactory agreement among all the data. Our data seem to fit better with w_{air} = 35 eV, however, this cannot be considered as a strong support of validity of the value.

From Table 1 it is seen that in vigorously stirred solutions the beam intensity in the range $(1 - 40)\cdot 10^9$ ions$\cdot$cm$^{-2}\cdot$s^{-1} does not influence on $G_E(Fe^{3+})$. The same can be said about 0.001M NaCl additive (Table 2). Saturation by oxygen probably increases the ferric ion radiation yield by some percent.

On the basis of the results obtained for 0.01 M ferrous sulfate solutions, we have concluded that the facility functios correctly. We are aware that the difficulties and uncertainties will increase with the atomic mass of the projectiles. But it is a general problem, not only with this particular installation. More accurate stopping power data for heavy projectiles, and more numerous and accurate data on differential w-values for heavy ions in air are needed.

Table 1.
Experimental conditions.

Ion	E_S MeV	Vacuum window (mg/cm^2)	Cell window (mg/cm^2)	Beam spot cm^2	Beam intensity $\times 10^{-9}$ cm^{-2}s^{-1}	Energy input$\times 10^{-18}$ eV	Volume ml	Beam direction	Ref.
$^{12}C^{6+}$	102			0.3	10	1 - 6	5	hor.	[7]
$^{12}C^{4+}$	77	Al (2.7)	mica* (2.9)	0.2	0.8 - 20				[8]
$^{12}C^{6+}$	102	Al (5.1)		28.3	~1				[9]
$^{12}C^{6+}$	20	Ti (4)	mica* (4)	0.3	3	1-10	30	hor.	[10]
$^{12}C^{6+}$	35	Ti (4)	mica* (4)	0.3	3	1-5	30	hor.	[11]
$^{12}C^{6+}$	121	Ti (2)	no	2.5	1 (7-40**)	1-13	3	ver.	this work

* - 2.9 g/cm^3 was taken as the mica density;
** - fluence rate in micro- and nanosecond pulses, respectively;
hor. = horizontal;
ver. = vertical.

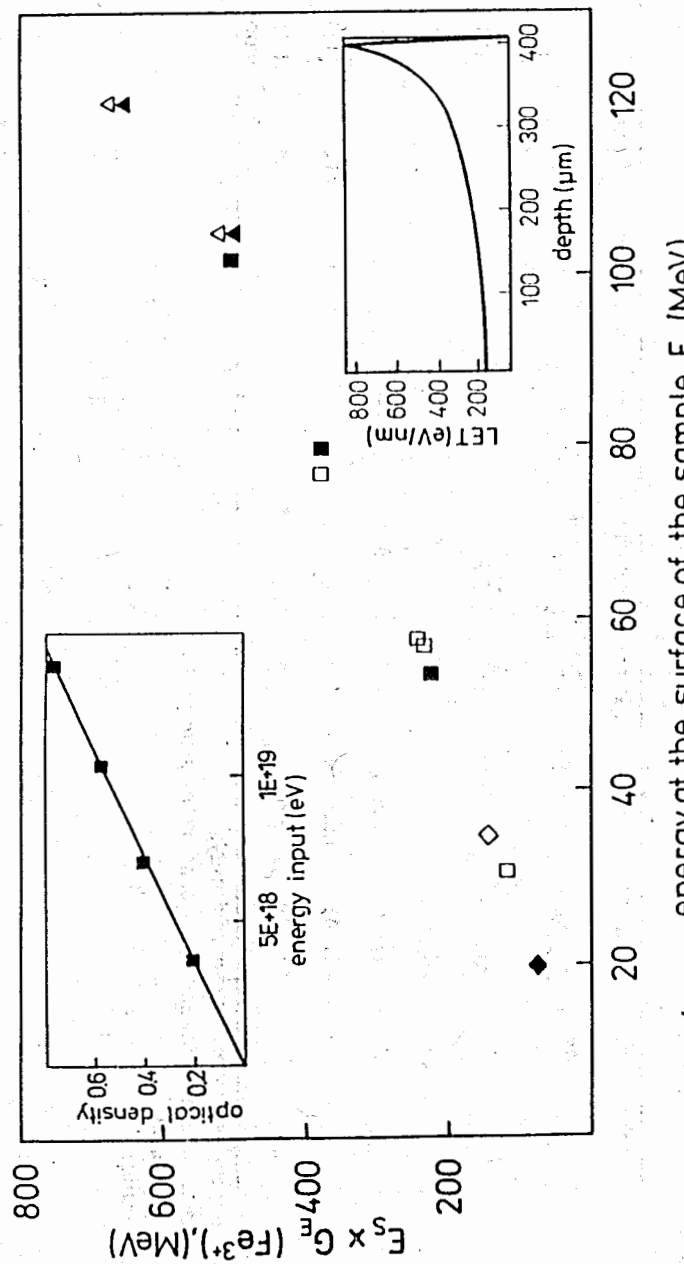

Figure. Energy dependence of E_S·G_E(Fe^{3+})-value for Fricke solutions irradiated with carbon ions. (■) [7], (□) [8], (◆) [10], (◇) [11], (▲) this work, w_{air}= 36.1 eV; (Δ) this work, w_{air}= 35 eV. Upper insert: optical density at 304 nm versus energy input. Lower insert: LET-depth dependence for ^{12}C ion beams with initial energy of 120 MeV penetrating the Fricke solution (according to [16]).

Table 2.
G_E(Fe^{3+})-values for stirred 0.01M Fricke solutions irradiated by ^{12}C ions. .

E_C MeV	E_S MeV	$E_C/(E_C-E_S)$ %	G_E(Fe^{3+})**	Remarks	Ref.
120	102	15	4.94		[7]
120	80	33	4.7*		[7]
120	54	55	4.2*		[7]
91.5	77	16	4.91	0.001M NaCl	[8]
74.2	58	22	4.2	0.001M NaCl	[8]
73.5	57	22	4.11	0.001M NaCl	[8]
54.1	31	43	3.83	0.001M NaCl	[8]
	102		5.2	O_2 saturated	[9]
	20		3.73		[10]
	35		4.1*		[11]
133	120	10	4.89±0.22 (5.08)		this work
133	105	21	4.83±0.20 (4.98)		this work
133	86	35	4.48 (4.62)		this work
133	66	50	4.45 (4.59)		this work
133	36	73	4.78 (4.94)		this work

* - obtained from the Figure

** - G-values in parenthesis were obtained assuming that w_{air} = 35 eV.

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Проверка функционирования установки
для радиационных исследований на циклотроне У-400 ОИЯИ
с помощью дозиметра Фрике

Несколько лет назад на вертикальном канале пучков четырехметрового циклотрона тяжелых ионов в Дубне была создана специальная установка для радиохимических и радиационных исследований (проект ХИПТИ), которая обеспечивает очень хорошие условия для облучения жидкостей и порошков. В настоящей работе сообщается о практическом испытании радиационной части установки. Подкисленные водные растворы аммоний сернокислого железа (II) облучались ионами углерода-12 с энергией 10 МэВ/а.е.м. Результаты измерений зависимости интегрального выхода образования Fe^{+3} от дозы и энергии ионов достаточно хорошо согласуются с данными трех других лабораторий. Обсуждаются некоторые технические проблемы радиационных экспериментов с тяжелыми ионами.

Работа выполнена в Лаборатории ядерных реакций им.Г.Н.Флерова ОИЯИ.

Препринт Объединенного института ядерных исследований. Дубна, 1995

Stuglik Z. E12-95-109

Radiation Facility at the JINR U-400 Cyclotron Checked
by Fricke Dosimeter Measurements

A few years ago, a vertical beam line of the 4-meter heavy ion cyclotron in Dubna was dedicated to radiochemical and radiation research (KHIPTI facility) providing excellent conditions for irradiation of liquids and powders. Practical examination of the radiation part of this facility is reported on. Acidic aqueous ferrous sulfate solutions (Fricke dosimeter) were bombarded with 10 MeV/amu carbon-12 ions. Dose and ion energy dependence of the integral yield of Fe^{3+} formation was measured and compared with the data from three other laboratories to find a satisfactory agreement. Some technical problems of heavy ion experiments are discussed.

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

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