

FAST NEUTRON FACILITIES FOR RADIOBIOLOGICAL IRRADIATIONS IN THE »RUĐER BOŠKOVIĆ« INSTITUTE

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The use of a neutron generator and a cyclotron as neutron facilities for radiobiological irradiations is presented. The neutron fields are mapped and the gamma components determined. Different kinds of ionization chamber, a chemical dosimeter, a proton-recoil detector, the associated alpha particle measurement and a Geiger-Müller counter are used to determine the dose. So far the facilities are used to irradiate cell cultures and mice.

1. Introduction

Radiosensitivity of some tumorous cells to fast neutron bombardment exemplified through the relative biological effectiveness — *RBE*, the oxygen enhancement ratio — *OER*, and the differences in the cell survival curves has stirred great interest for neutron therapy of some cancers and concurrently an interest for radio-

biological research of the effects of neutron irradiation. The fast neutrons are also in the focus of interest in some other fields, like the generation of mutations in the breeding of new plants with better characteristics.

To meet the growing need for fast neutron irradiation facilities we have tested the possibilities of existing accelerators at the »Ruđer Bošković« Institute namely a 300 kV Cockcroft-Walton accelerator and a 14 MeV deuteron cyclotron.

Besides the development of the irradiation facilities we have also developed the necessary accompanying dosimetry. We have tested a number of independent dosimetric arrangements minimizing thus the possibility of systematic errors in the absorbed dose determination.

In the present paper we discuss the characteristics of the fields achieved at both accelerators, their intensities, uniformity and spatial distribution. The basic demand on the accelerator capabilities and the geometries used was to accomplish dose rates necessary for radiobiological studies, i.e. not less than 5×10^{-4} Gys $^{-1}$.

2. Neutron generator

2.1. Characteristics of the facility

The 14 MeV neutron facility consists of a Texas Nuclear 300 kV accelerator in conjunction with a rotating, water cooled tritium target (Multivolt Ltd). The un-analyzed deuteron beam at the accelerating voltage of 240 kV was used to produce neutrons by the $d + t \rightarrow {}^4\text{He} + n$ reaction. The beam was typically 700—900 μA . The beam spot on the target was approximately 6 mm in diameter. The tritium target activity was 2.3×10^{10} Bq/cm 2 . With the present setup neutron yields of $2 - 4 \times 10^{10}$ s $^{-1}$ have been achieved. The half life of the target in the described regime is approximately 50 hours.

2.2. Neutron monitoring

The associated alpha particle method was used for neutron monitoring. To detect alpha particles a surface barrier silicon detector was placed 50.6 cm from the tritium target at an angle of 90° with respect to the incident deuteron beam. A diaphragm of 0.358 mm 2 area determines a solid angle of 1.4×10^{-6} sr. The spectrum of alpha particles registered by the detector has been recorded by a multichannel analyzer. The background from (n, α) and (n, p) reactions in the detector material as well as that one originating from the interaction of deuterons with deuterons and/or ${}^3\text{He}$ in the target has been subtracted. The neutron yield has been determined with a precision of 2.5% by integrating the yield of associated alpha particle peak pertaining to the $T(d, n){}^4\text{He}$ reaction.

2.3. Dosimetry

To measure the absorbed dose and dose rate in the field around a 14 MeV neutron generator one can use several approaches:

— conversion of the measured neutron fluence into dose rate via the kerma factors which are well known in this energy region $^{1)}$. One should however be careful tha

the neutron fluence is actually modified by: a) scattering and absorption of neutrons in the target housing and cooling layer, b) scattering from surrounding massive objects (floor, magnet etc.).

— measurement of the absorbed dose with an ionization chamber. In our measurements we have used a tissue equivalent (*T. E.*) chamber produced by CEN Fontenay aux Roses²⁾ in conjunction with a Keithley model 602 electrometer. As the distance between the chamber and the electrometer was approximately 20 m, parasite capacities of such long cables forced us to use the electrometer in the 10^{-7} Coulomb range. The chamber was filled with tissue equivalent gas, based on methane, at atmospheric pressure.

It is a well known fact that a neutron source produces a mixed neutron and gamma radiation field. The gamma rays are produced in the target holder and its immediate surrounding by various neutron induced reactions (e. g. (n, n') , $(n, 2n)$ etc.). Because of the difference in the radiobiological efficiency (*RBE*) and oxygen enhancement ratio (*OER*) of neutrons and photons the exact knowledge of the gamma component is important for each facility used for radiobiological purposes.

For the evaluation of the separate absorbed doses of neutrons and photons in a mixed field we used the «two chamber method» which implies two dosimeters. One device (*T*) usually has approximately the same sensitivity to neutrons and to gamma rays, while the second instrument (*U*) has lower sensitivity to neutrons than to gamma rays.

As the first instrument the *T. E.* chamber was used, whereas for the neutron insensitive one we used a Geiger-Müller (*G. M.*) counter (Philips model ZP 1100). Both instruments were calibrated in a known gamma field of ^{60}Co used for radiotherapy purposes. The following calibration factors were obtained: *T. E.* chamber field with *T. E.* gas: $(1.58 \pm 0.07) \times 10^7 \text{ GyC}^{-1}$, *G. M.* counter: $(2.3 \pm 0.001) \times 10^{-9} \text{ Gy per count}$. The dead time of $(28.5 \pm 0.7) \mu\text{s}$ for the *G. M.* counter was determined using two gamma ^{60}Co sources.

For the mixed field one can write the following system of equations³⁾:

$$\begin{aligned} R_T &= k_T D_N + h_T D_G \\ R_U &= k_U D_N + h_U D_G. \end{aligned} \tag{1}$$

The quantities $R_{T,U}$ are determined using the ratio of the charge collected by the dosimeters ($Q_{T,U}$) and the gamma ray dose $\left(\left(\frac{Q_C}{D_C}\right)_{T,U}\right)$ obtained in the calibration run, in the following way:

$$R_{T,U} = Q_{T,U} \left(\frac{D_C}{Q_C}\right)_{T,U} \frac{P_C}{T_C} \frac{T}{P}. \tag{2}$$

$\frac{P_C}{T_C} \frac{T}{P}$ is the correction for different masses of gas in the sensitive volume of the dosimeters for calibration and measuring conditions.

In the system of equations (1) D_N and D_G represent absorbed doses of neutrons and photons respectively, for the tissue in the mixed field; k_T and k_U are sensitivities of each dosimeter to neutrons relative to its sensitivity to the gamma rays used for calibration and h_T and h_U are sensitivities of each dosimeter to the photons in the mixed field relative to its sensitivity to the gamma rays used for calibration. It can be shown that:

$$k_T = \frac{\overline{W}_C (S_{m,g})_C (K_t/K_m)_C}{\overline{W}_N (S_{m,g})_N (K_t/K_m)_N} \quad (3)$$

$$h_T = \frac{\overline{W}_C (S_{m,g})_C (K_t/K_m)_C}{\overline{W}_G (S_{m,g})_G (K_t/K_m)_G} \quad (3)$$

where \overline{W} is the average energy expended to create an ion pair; $S_{m,g}$ is the ratio of the average mass stopping power of the wall relative to the gas and K_t/K_m is the ratio of the kerma in tissue to that in the dosimeter material. The subscript C denotes values applicable to the calibration situation.

To determine neutron and gamma absorbed doses in the mixed field of the 14 MeV facility we used the following parameters for the system of equations (1):

$$\begin{aligned} \overline{W}_C/\overline{W}_N &= 0.95; \quad (S_{m,g})_C/(S_{m,g})_N = 1; \quad (K_t/K_m)_C = 1; \\ (K_t/K_m)_N &= 0.959; \quad k_U = 0.004; \quad k_T = 0.99; \quad h_T = h_U = 1 \end{aligned}$$

Kerma factors for *ICRU* muscle (K_t) and A-150 plastic (K_m) were taken from the Caswell tables¹⁾ for the neutron energy of 14.5 MeV, while the other factors are recommended in *ICRU* 26³⁾. It has to be notified that all these factors are subject to some systematic errors which lead to a total systematic error of about 8% in the dose determination. The same discussion also holds for cyclotron produced neutrons described later. These errors can not be avoided at the moment and they are not declared in the dosimetric results. However, intercomparison measurements were done, both for neutron generator and cyclotron produced neutron fields, to minimize systematic errors. For further discussion about errors and intercomparison measurements see Ref. 4.

Using the above method the neutron and gamma dose rates obtained for two distances from the neutron source and normalized to a neutron yield of 10^{10} s^{-1} are shown in Table 1.

TABLE 1.

source distance (cm)	P_N (Gys ⁻¹)	P_G (Gys ⁻¹)	$\frac{D_G}{D_{N+G}}$
11	4.87×10^{-4}	0.31×10^{-4}	0.06
20	1.51×10^{-4}	0.13×10^{-4}	0.08

Neutron and gamma dose rates at 14 MeV neutron generator for two distances from the neutron source, normalized to a neutron yield of 10^{10} s^{-1} .

The neutrons produced by the $d + t$ reaction using 240 keV deuterons are nearly monoenergetic and isotropic. Up to that extent the isodose curves are concentric circles around the source and the neutron absorbed dose decreases with the distance as $1/r^2$.

2.4. Possibilities and limitations of the neutron generator for use in radiobiological applications

Due to the relatively low neutron yield obtainable with the described facility the usefulness of the neutron field is limited to the immediate proximity of the source. The required dose rates for radiobiological experiments of approximately 10^{-3} Gys^{-1} can be obtained at the distance of about 10 cm from the neutron source.

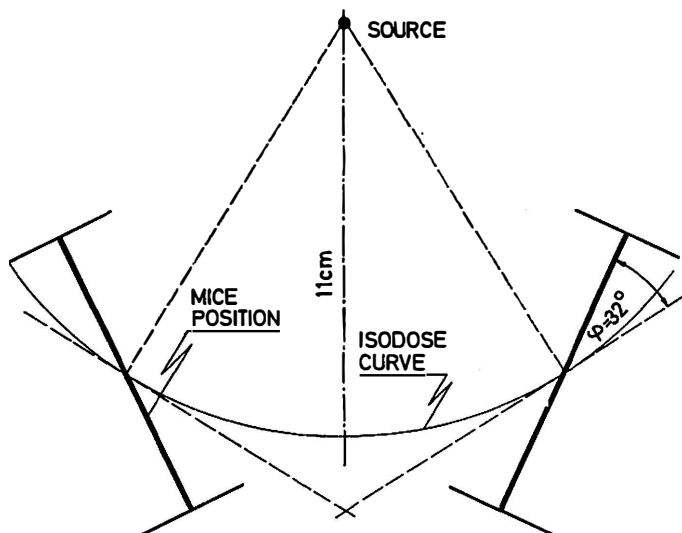


Fig. 1. Experimental setup for the mice irradiation in the 14 MeV neutron generator irradiation field.

The neutron generator has been used for irradiations of cell cultures⁵⁾ and mice. The physical dimensions of the mice introduce at such small source to skin distances a very strong geometrical factor of inhomogeneity of the radiation field. To remedy that fact we have made a stand where 4 mice could be irradiated simultaneously taking care to interrupt the irradiation when half of the dose has been delivered and to rotate the mice in the position by 180°. The arrangement is shown in Fig. 1. Two mice are positioned as shown in Fig. 1, and the other two are placed symmetrically with regard to the horizontal plane through the source. Using this method the homogeneity of the radiation of up to about 3% can be achieved for small animals⁴⁾. Individual irradiations were monitored by the associated alpha-particle counting.

3. Cyclotron

3.1. Characteristic of the facility

An internal beam of 14 MeV deuterons, available in the cyclotron of the »Ruder Bošković« Institute, has been used for neutron production via the $^{27}\text{Al}(d, n)$ reaction. The maximum obtainable beam intensity is $400 \mu\text{A}$. However, in the present arrangement, the water — cooled Al target can withstand long term irradiation with a typical current of $200 \mu\text{A}$.

The setup of the facility is shown in Fig. 2. The target position and yoke dimensions do not allow to approach the neutron source closer than 1 m.

In contrast to the case of the $\text{T}(d, n)$ reaction at low deuteron energies the $^{27}\text{Al}(d, n)$ reaction is highly anisotropic. Therefore the measurement of the spatial distribution of the neutron yield is necessary. We have used a modified acti-

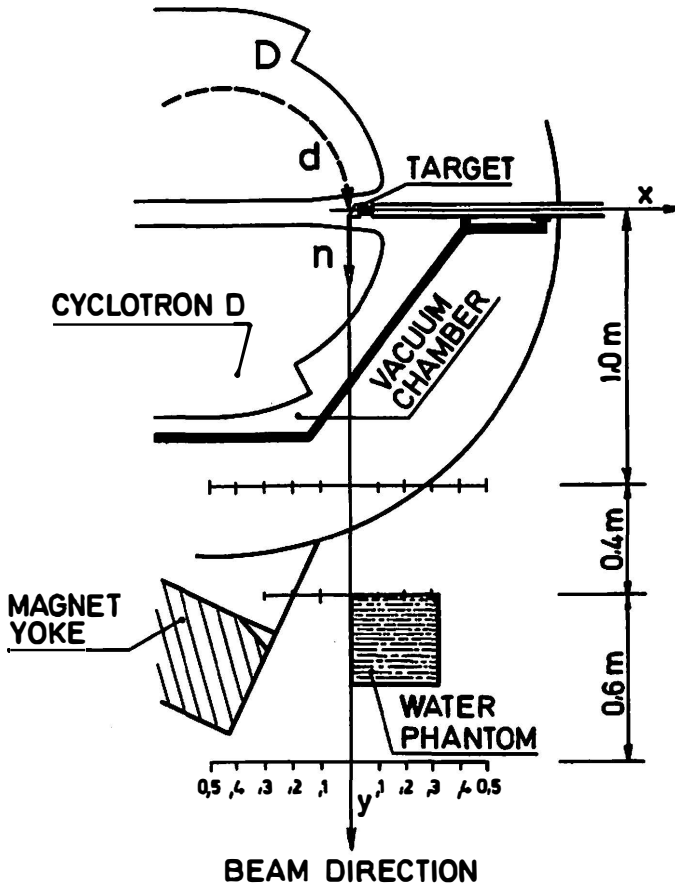


Fig. 2. Layout of the cyclotron produced neutron irradiation field.

vation technique: an aluminium foil (100 cm × 35 cm) was placed 1 m far from the target, perpendicular to the beam direction, and was irradiated for about 2 hours. Immediately after irradiation the foil was covered by three (30 cm × 35 cm) radiographic films. After about 12 hours films were removed, developed and analyzed for optical density. The obtained spatial distribution of the optical density is shown in Fig. 3. The main contributor to the neutron activation of the aluminium is the $^{27}\text{Al}(n, \alpha)^{24}\text{Na}$ reaction ($Q = -3.14$ MeV). Since this reaction has a threshold of 3.5 MeV, the spatial distribution obtained by this method does not reflect the distribution of neutrons with energies below 3.5 MeV.

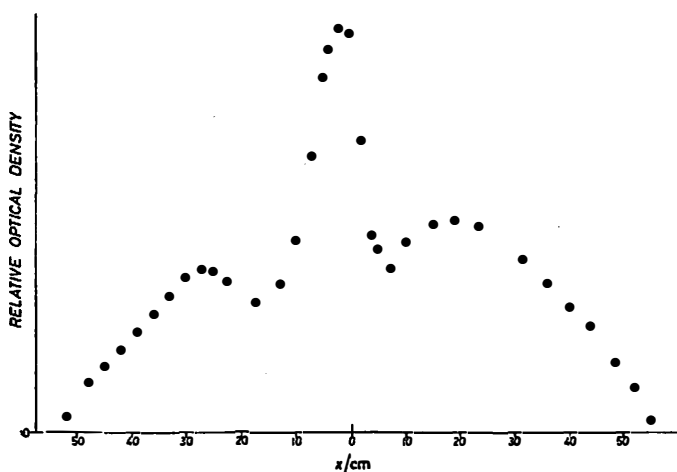


Fig. 3. Neutron field profile obtained at the cyclotron using the ^{27}Al activation method. Neutron source — Al foil distance is 1 m.

The spatial dose distribution was also measured with pencil shaped, 10 cm long chemical dosimeters⁶⁾ and checked with an ionization chamber. The response of the dosimeters is equal for gamma rays and neutrons, therefore the readings give the total dose with a mean error of $\pm 3\%$ at dose of about 3 Gy. Dose distributions obtained with chemical dosimeters for 1 m and 1.4 m distances from the Al target are presented in Fig. 4. The radiation field profiles similar to that depicted in Fig. 3 were obtained. A reproducibility better than $\pm 7\%$ was obtained by checking thus obtained profiles several times during one year.

In view of these results we use the flat portion of the radiation field on the right side from the beam direction, i.e. from $x = 15$ cm to $x = 50$ cm, as the working field. The vertical dependence of the field within the given coordinates has been also measured with the same chemical dosimeters. The field was found to be homogeneous up to ± 10 cm from the median plane. Thus, radiation fields of $15\text{ cm} \times 15\text{ cm}$ at $y = 1.4$ m could be specified with inhomogeneity not larger than 5% and with the dose rates of 10^{-3} Gys⁻¹.

The neutron energy spectrum has been measured using two methods:

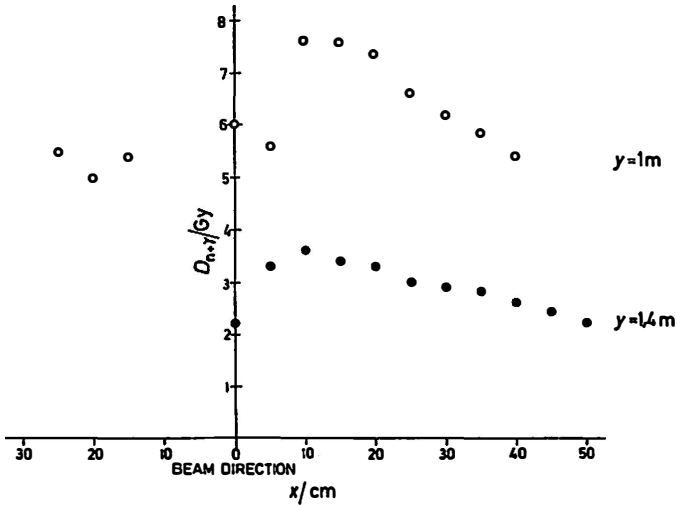


Fig. 4. Total dose profiles in the median plane at 1 m and 1.4 m distances from the Al target measured with chemical dosimeters.

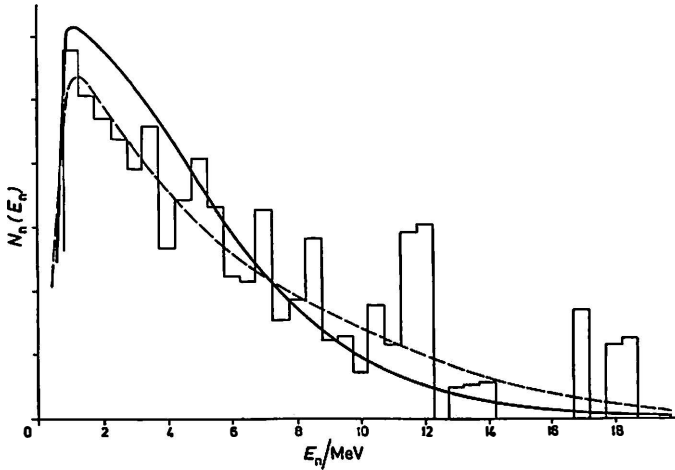


Fig. 5. Cyclotron produced neutron energy spectra obtained a) with nuclear emulsion method (hystogram and/or dashed curve), and b) with proton recoil detector (solid curve).

i) Nuclear emulsions — measuring the recoil proton tracks in a specified forward pyramide. Nuclear emulsions have been exposed to a 10 cm × 10 cm collimated neutron beam at 0° direction ($x = 0$) and the distance of 2.4 m from the Al target. Fig. 5 shows the neutron spectrum determined from the measurement of recoil protons within a pyramide defined by the angles $\alpha = \pm 30^\circ$, $\beta = \pm 15^\circ$ corresponding to the horizontal and vertical openings, respectively.

ii) A thin semiconductor detector measuring the deposited energy of recoil charged particles (p or d) from CH_2 and CD_2 radiators^{7,8)}. For non-monoenergetic neutrons the energy deposited in a thin detector is a complex image of the recoil particle energy spectrum and thus of the incident neutron spectrum. However, using radiators of different thicknesses and of different kinds, one can disentangle the contributions of recoil charged particles with ranges less than the detector thickness from those losing only a fraction of their energies in the detector. Thus, at least in principle, one can determine the neutron energy spectrum. Measurement of the cyclotron neutron spectrum has been done using a semiconductor detector with four different CH_2 radiators (2.7, 5.4, 10.8 and 32.4 mg cm^{-2} thick) and one CD_2 radiators (18.1 mg cm^{-2}). The geometry used was similar to the one used with nuclear emulsions. Assuming several different neutron spectra, it was found that the best fit to the measured deposited energy spectra for all radiators is obtained for the spectrum shown in Fig. 5 by a solid curve.

3.2. Dosimetry

In the case of the neutron field obtained by the cyclotron it is almost impossible to determine the exact number of neutrons in the low energy region (less than 0.5 MeV). Therefore the evaluation of the dose using the known kerma factors, as explained in the case of the 14 MeV neutrons, is not readily available.

The absorbed dose was measured using the two chamber method as explained in the section 2.3. For the neutron detection two chambers were alternatively used — the *T. E.* ionization chamber described in section 2.3. and a CH/acetylene chamber. The gamma rays were detected using a chamber with magnesium walls, filled with argon. The CH/acetylene and Mg/Ar chambers were manufactured by the »Jožef Stefan« Institute in Ljubljana, Yugoslavia according to the specifications of the IAEA Technical Report No 76. Chambers are of cylindrical shape with diameter of 14.5 mm and 55 mm long. They were calibrated in the same ^{60}Co gamma field as the *T. E.* chamber and the following calibration factors were obtained:

$$\text{CH/acetylene: } (7.9 \pm 0.1) \times 10^6 \text{ GyC}^{-1}$$

$$\text{Mg/Ar: } (5.77 \pm 0.04) \times 10^6 \text{ GyC}^{-1}.$$

The G. M. counter was not found satisfactory for measurements at the cyclotron, because of the large dead time correction which had to be applied.

The parameters for the determination of the separated neutron and gamma absorbed doses, according to the system of equation (1), are presented in Table 2. Kerma factors were taken from the Ref. 1 using a modal energy of 5 MeV, and the other factors from Ref. 3.

The absorbed doses derived from the *T. E.* and CH/acetylene chamber differed by 5%, the higher value being the one of the CH/acetylene chamber. This may be due to the difference in the chamber shape and in the $\overline{W}_c/\overline{W}_N$ value used for CH/acetylene chamber.

The neutron doses and gamma fractions, measured with the described setup, at some points in the cyclotron radiation field, are presented in Table 3. All the values are normalized to the incident deuteron beam current integral of $6 \times 10^5 \mu\text{A s}$.

TABLE 2.

Chamber	$\frac{\bar{W}_C}{\bar{W}_N}$	$\left(\frac{K_t}{K_m}\right)_N$	$\left(\frac{K_t}{K_m}\right)_C$	$k_{T,U}$
T. E./T. E.	0.95	0.96	1	0.99
CH/acetylene	0.95	1.23	1.01	0.78
Mg/Ar				0.02

Parameters for the determination of the separated neutron and gamma absorbed doses at the cyclotron.

TABLE 3.

Position		D_N (Gy)	D_G (Gy)	$\frac{D_G}{D_{N+G}}$
y(m)	x-right (cm)			
1	15	7.5	1.9	0.20
1.4	15	3.2	0.7	0.18
1.4	50	2	0.35	0.15
2	15	2	0.27	0.12

Neutron doses and gamma fractions at some points in the cyclotron normalized to the incident deuteron beam current integral of $6 \times 10^5 \mu\text{A s}$

Since the gamma dose decreases as the distance from the Al target increases, one can conclude that most of the gamma rays came from interactions in the target or in the vicinity of the target.

3.3. Possibilities and limitations of the I.R.B. cyclotron for use in radiobiology

At a distance of 1.4 m from the target, in the region of maximum field homogeneity, neutron dose rates of 10^{-3}Gys^{-1} were achieved with deuteron currents of 200 μA . At this location the accompanying gamma component is larger than in the case of the neutron generator and amounts to 12—20% of the total dose.

So far, the cells and mice were irradiated in an open geometry i.e. without shielding and/or collimation between the source and irradiation region. It is conceivable that an effective shielding could improve the neutron/gamma dose ratio.

A radiation field greater than 15 cm \times 15 cm could be achieved in open geometry choosing special positions with respect to the beam as shown in Fig. 6a where the decrease of the neutron yield is compensated through shorter distance to the source. The homogeneity of a 100 cm \times 35 cm radiation field was checked

with the chemical dosimeters positioned as shown in Fig. 6b. The results are presented in Fig. 6c. We found that the field of 60 cm × 35 cm can be obtained with a homogeneity of ± 10%. This gives the possibility of irradiating larger biological samples. To simulate such larger biological sample a 30 cm × 30 cm water phan-

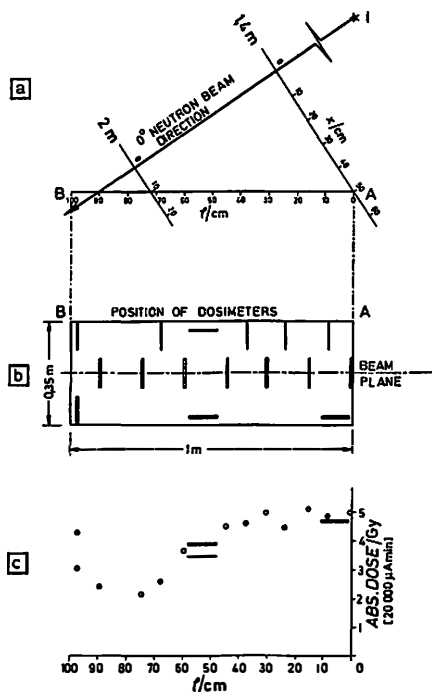


Fig. 6. Homogeneity of a 100 cm × 35 cm cyclotron radiation field: a) position of the field in the horizontal plane of the cyclotron with regard to 0° neutron beam direction is denoted by line AB, b) arrangement of chemical dosimeters in the vertical plane of 100 × 35 cm field, c) doses measured with chemical dosimeters. Solid circles correspond to measurements with chemical dosimeters above the beam plane, while open circles refer to positions in the beam plane or below.

tom was used. In order to get some preliminary results the depth dose distributions in the phantom were measured separately for neutrons and gamma rays using *T. E.* + Mg/Ar pair of chambers. The position of the phantom is shown in Fig. 2 and the obtained depth dose distributions are given in Fig. 7.

4. Conclusion

Two facilities producing neutrons suitable for radiobiological applications have been developed at the »Ruder Bošković« Institute, as well as the accompanying dosimetry which allows to determine the total dose or neutron and gamma doses separately, with a precision of about 5%.

The main characteristics of the facilities are summarized in Table 4.

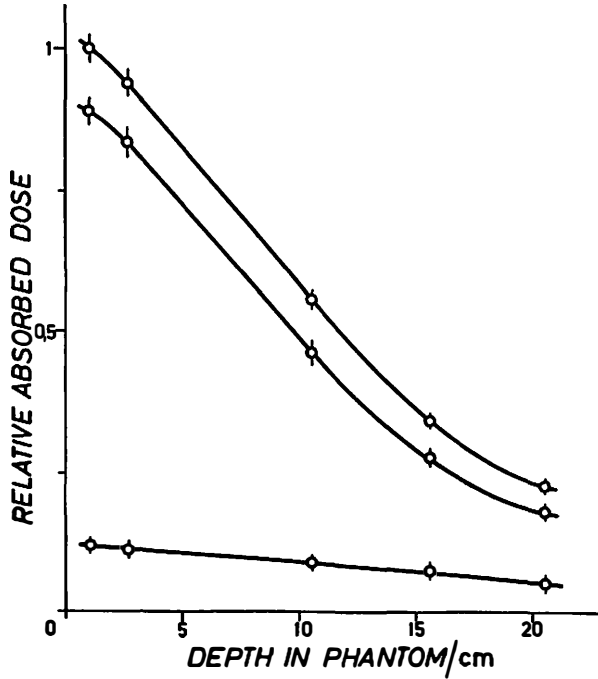


Fig. 7. Depth dose distributions in a water phantom in the neutron field of the Institute »Ruder Bošković« cyclotron.

TABLE 4.

	neutron generator	cyclotron
neutron producing reactions	$d + t \rightarrow n + \alpha + 17.6 \text{ MeV}$	$d + {}^{27}\text{Al} \rightarrow {}^{28}\text{Si} + n + 9.35 \text{ MeV}$
maximum deuteron energy	300 keV	16 MeV
average beam current	400–900 μA	200 μA
angular distribution	isotropic	anisotropic — forward peaked
energy spectrum	monoenergetic 14.5 MeV	mean energy 2.5 MeV
γ -contamination	6–8%	12–20%
P_{tot}^* 0.11 m	$(0.7 - 1.7) \times 10^{-3} \text{ Gys}^{-1}$	$2.5 \times 10^{-3} \text{ Gys}^{-1}$
1 m	$(0.7 - 1.7) \times 10^{-5} \text{ Gys}^{-1}$	$1.07 \times 10^{-3} \text{ Gys}^{-1}$
1.4 m		
possibilities for the irradiations of biological samples	12 samples of cellcultures or 4 mice with $\sim 10^{-3} \text{ Gys}^{-1}$	several fields 15 cm \times 15 cm with inhomogeneity up to 5% and dose rates up to $2 \times 10^{-3} \text{ Gys}^{-1}$ could be defined. For 10^{-3} Gys^{-1} there is a field 35 cm \times 60 cm with inhomogeneity of $\pm 10\%$

* P_{tot} = Total ($n + \gamma$) absorbed dose rate for tissue in air at the specified target distances

Fast neutron facilities for radiobiological irradiation in the »Ruder Bošković« Institute.

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MOGUĆNOSTI RADIOBIOLOŠKIH OZRAČAVANJA NEUTRONIMA NA AKCELERATORIMA INSTITUTA »RUDER BOŠKOVIĆ«

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Neutronske generator i ciklotron ispitani su u svrhu primjene neutronske polja u radiobiologiji. Snimljena su polja te su određene γ -komponente. Za određivanje doze upotrebljene su različite ionizacijske komore, kemijski dozimetar, Geiger-Müller brojač, detekcija raspršenih protona poluvodičkim detektorom, te metoda pridruženih α -čestica. Do sada su na tim akceleratorima vršena ozračivanja miševa i kultura stanica.