New applications of the recombination method in the dosimetry of mixed radiation fields

N. Golnik[1] and M. Zielczy□ski[2]
[1] Institute of Precision and Biomedical Engineering, Warsaw University of Technology, Chodkiewicza 8, 02-525 Warsaw, Poland
[2] Institute of Atomic Energy, 05-400 Otwock-Swierk, Poland

INTRODUCTION

In order to define the recombination methods, one should distinguish between two main processes of recombination of ions in gases - *initial recombination*, which occurs within a track of a single charged particle and *volume recombination*, which occurs when ions from tracks of different particles may recombine. It is essential that the initial recombination does not depend of the dose rate but strongly depends on local ion density, i.e. it can be correlated with LET. Such correlation was used in 1962 (1, 2) for the first designs of, so called, recombination chambers, which were invented with the aim of building a detector having a response that could simulate the dependence of the radiation quality factor on LET.

Most generally, the recombination chambers are ionization chambers operated in unsaturated mode, under conditions of initial recombination of ions. Usually, such chambers contain several parallel-plate tissue-equivalent electrodes spaced by few millimeter gaps. Mostly, the chambers are filled with tissue-equivalent gas mixtures up to the pressure of some hundreds of kilopascals. The electrical charge created between the electrodes is proportional to the absorbed dose, while the shape of the saturation curve of the chamber (example in Figure 1) provides information on radiation quality (3-6).

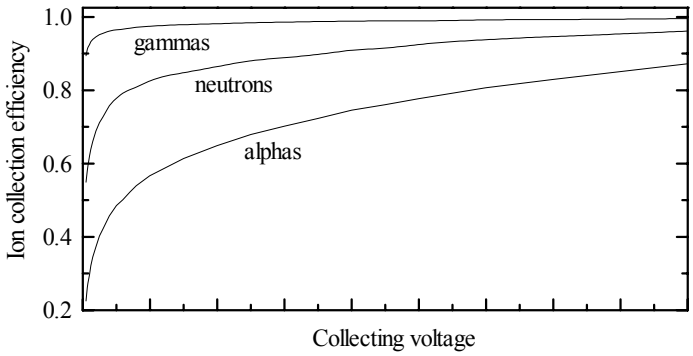

Figure 1. Saturation curves of the recombination chamber of REM-2 type (see Figure 5) for gamma (^{137}Cs), neutron (^{241}Am-Be) and $\alpha + \beta$ (^{222}Rn + daughters) radiations.

Since that time both the detector design and methods of measurements have been considerably improved. Recombination chambers have often been used in radiation protection dosimetry as a practical instrument for determining the quality factor and dose equivalent in mixed radiation fields. In many series of experiments the chambers have proved to work as reliable instruments which can be used even in complex radiation fields with poorly known composition and broad energy range (5, 7-9). Tissue-equivalent chambers can be also used for determination of the photon and neutron components of the absorbed dose in mixed neutron-gamma fields and for estimation of the dose distribution versus LET.

This kind of applications of the recombination methods will be illustrated in this paper by the results of recent measurements performed in fields of nearly monoenergetic neutrons with peak energies of about 44 MeV and 66 MeV. The recombination chamber was there used for the determination of the absorbed dose and recombination index of radiation quality (4).

Two other applications presented in this paper are quite different from the classical recombination approach. First of them is determination of the photon kerma in the above mentioned neutron fields by a high pressure C-CO_2 ionization chamber. The chamber was operated at low voltages, i.e. under conditions of strong initial recombination in the tracks of the secondary high-LET charged particles, while almost all ions from low-LET tracks are collected. A new method of measurements was developed in order to extend the applicability of the graphite chamber as a „neutron insensitive“ detector to an energy range where the relative neutron sensitivity of the chamber, k_U, was not well known.

The second new application presented here is a thin-wall recombination chamber for the dosimetry of weakly penetrating radiation. In contrary to other designs, the chamber was filled with butadiene (C_4H_6) under atmospheric pressure. Almost all earlier studies of initial recombination concerned pressurized gases, therefore, special series of experiments were performed for this work, in order to show that the proper conditions of initial

recombination can be achieved also under atmospheric gas pressure, provided that heavy gases are used. The chamber can be used for direct determination of radiation protection operational quantities H'(0.07) and H'(3) in mixed radiation fields containing alpha particles or heavy ions. It can be also applied for some special measurements, e.g. for determination of contamination of the beams of neutrons and higher energy photons by low energy photons or for determination of radiation field close to the body or phantom surface.

IONIZATION CHAMBERS

Four different ionization chambers were used. All of them were designed in the Institute of Atomic Energy.

The measurements of H*(10) in fields of neutrons energies of about 44 MeV and 66 MeV were performed with the tissue-equivalent, in-phantom chamber of F-1 type (Figure 2). This is a 3.8 cm^3 parallel-plate chamber, filled with methane up to a pressure of 1.1 MPa. The chamber has three ∅ 34 mm electrodes, the wall thickness is 0.6 g/cm^2 and the distance between electrodes is 1.75 mm (6).

The chamber is designed in such a way that it can be placed inside a water phantom and directly used for measurements at high dose rates. Therefore, it can be applied for the determination of dosimetric parameters in different points in the phantom. Broad operational dose rate range of the chamber - from 10^{-5} up to 100 Gy/min makes it useful for characterization of both the radiotherapy fields and the fields used for calibration of radiation protection instrument.

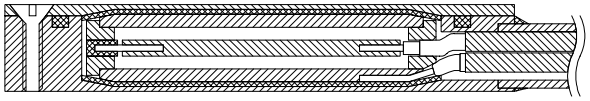

Figure 2. Cross section of the in-phantom chamber of F1 type

A high-pressure graphite ionization chamber of G-5 type (Figure 3) was used in the same 44 MeV and 66 MeV neutron fields . The cylindrical chamber is 115 mm in length and 18 mm in diameter. The distance between the electrodes is equal to 2 mm. The chamber is enclosed in a 0.3 mm thick aluminum container and can be filled with CO_2 gas up to a pressure of several MPa (10). Two gas pressures of 3.4 MPa and 2.1 MPa were used for this work. The chamber was operated with low collecting voltages (between 5 and 100 V) in order to provide the conditions of strong initial recombination of ions in the high-LET particle tracks, because such conditions strongly reduce the relative neutron sensitivity of the chamber. The chamber can be used for measurements of photon dose rates from 0.5 mGy/h up to 500 Gy/h.

Basic physical studies of initial recombination in different gases at atmospheric gas pressure were performed using the chamber of REM-2 type (11). It is a cylindrical, parallel-plate ionization chamber with 25 tissue-equivalent electrodes, the volume of 2000 cm^3, mass of 6 kg and effective wall thickness of about 2 g/cm^2 (Figure 4).

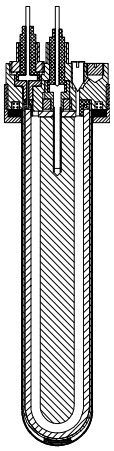

Figure 3. Cross-section of the chamber G-5.

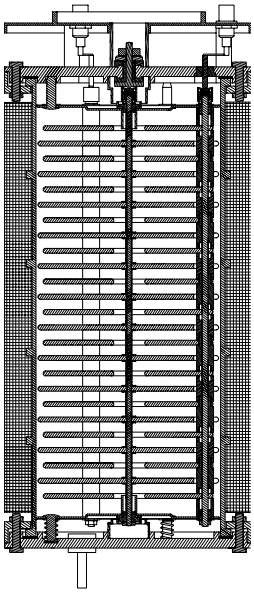

Figure 4. Cross-section of the recombination chamber REM-2.

The thin-walled recombination chamber KR-16 (Figure 5) was specially designed for the dosimetry of weakly penetrating radiation. The KR-16 is a planar, tissue-equivalent ionization chamber, with the gas cavity under a thin absorber and backed by a thick layer of tissue-equivalent plastic having similar backscattering characteristics as tissue. Almost all the details of the chamber are made of tissue-equivalent material. A thin foil stretched on a solid isolating ring serves as the polarizing electrode. The collecting electrode and the guard ring of the chamber form a common flat surface ensuring the uniform electrical field strength and well defined active volume of the chamber.

An additional guarding foil (electrically connected to the guard ring) was mounted at the opposite side of the isolating ring. In effect, the polarizing electrode was placed in the middle between two surfaces being at the same electrical potential. Test measurements, performed without the guarding foil, showed that the attraction of the electrodes at high voltages reduced the active volume and caused the reduction of the measured ionization current by over 2% at highest voltages (above 500 V). Both, the polarizing electrode and the guarding foil are made of 1.6 mg/cm^2 thick Mylar covered with 0.1 mg/cm^2 of aluminum.

A specific property of the KR-16 chamber design is that the voltage insulators are „invisible" from any point of the active volume. Such configuration ensures very good time stability of the electrical field strength in the active volume of the chamber and enables reading of the ionization current in short time after changing the supplying voltage from high (saturation) to low (recombination) voltage and from positive to negative polarity. The chamber is operated with a gas flow system at gas flow rate of about 3cm^3/min.

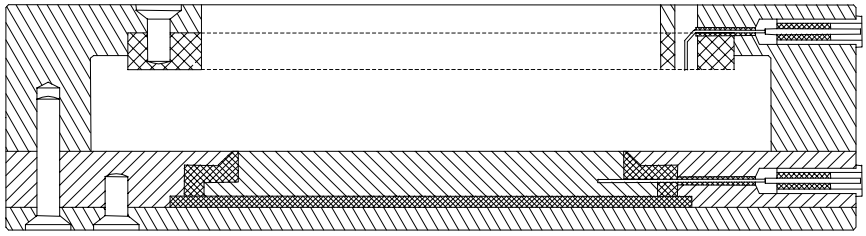

Figure 5. Cross-section of the thin-walled recombination chamber KR-16. Dashed lines show the thin foil entrance window and the voltage electrode, dashed area - TE material, cross-hatched - insulators.

RADIATION FIELDS

The measurements with F-1 and G-5 chambers were performed in two different collimated neutron beams available at the injector cyclotron of the Paul Scherrer Institute (PSI), Villigen, Switzerland (12, 13) and at the accelerator facility of the Université Catholique de Louvain-la-Neuve (UCL), Belgium (14, 15).

At PSI the neutron fields were produced by the p + Be reaction (2 mm thick Be disc) with proton energies of 72.5 MeV and 50.4 MeV, which resulted in the two „peak neutron" energies of 44 and 66 MeV, respectively. The chambers were placed at the distance of 9.11 m from the target under 0 degrees with respect to the direction of the incoming proton beam. The results were related to the reference distance of 7.78 m. The spectral and total neutron fluence of the beam was measured (16), using a NE213 scintillation detector, the activation of silver foils and by a ^{238}U fission chamber, respectively. The neutron beam was monitored by means of the beam charge of the Be target.

At UCL, the neutron fields were produced with the p + Li reaction (5 mm thick Li disc) by bombardment with proton energies of 48.5 MeV, which resulted in a „peak neutron" energy of 45.4 MeV. Two independent neutron monitors were used for all the experiments - a fission chamber with a ^{238}U layer and a 2 mm thin NE211 plastic scintillation detector. The irradiation of the chamber was performed in a peripheral mode behind a thick stack of passive dosimeters, 9.715 m apart from the neutron source under 0 degrees. The total „neutron peak" fluence for these conditions was determined with a proton recoil telescope. The neutron spectral fluence in this point was determined with a NE213 scintillation detector without the stack in the beam, normalised to the „peak neutron" fluence and then extrapolated to low neutron energies (17).

Measurements with KR-16 and REM-2 chambers were performed in reference gamma and neutron radiation fields of isotopic sources in the Institute of Atomic Energy in Swierk, Poland (18).

MEASUREMENTS OF DOSE EQUIVALENT IN FIELDS OF MONOENERGETIC HIGH-ENERGY NEUTRONS.

The use of calibrated A-150 plastic TE ionization chambers with TE gas filling is recommended (19, 20) for radiation-therapy dosimetry as the practical method for determination of the tissue kerma in air and the absorbed dose in a TE phantom. From this point of view, the recombination chambers are not essentially different from the chambers operated under atmospheric gas pressure. The additional advantage of the high-pressure ionization chambers is that they can relatively easy provide some information related to radiation

quality. This may help to ensure the desired conditions of the irradiation.

The simplest approach is to use a quantity called recombination index of radiation quality (RIQ, Q_4). The RIQ is defined (3, 4) as:

$$Q_4 = \frac{1 - f_n(U_R)}{1 - f_\gamma(U_R)} \tag{1}$$

where:

- f(U) is the ion collection efficiency, i.e. the ratio of ionization current measured at voltage U to the saturation current;
- $f_n(U)$ is the ion collection efficiency in the chamber irradiated in mixed radiation field being investigated;
- $f_\gamma(U)$ is the ion collection efficiency of the same chamber when irradiated in a reference field of gamma radiation (^{137}Cs);
- U_R is the "recombination voltage", ensuring $f_\gamma \approx 0.96$, determined during the calibration.

In the past, the RIQ was used as a good approximation of radiation quality factor, like it was defined in the ICRU Report 21 (21). Now, it is considered as an independent parameter, dependent on restricted LET, L_Δ, with energy cut-off Δ of about 500 keV (see Figure 6). It was shown (5, 6) that it can be used for fast and sensitive identification of possible changes of radiation energy or composition due to changes of the facility arrangement, different targets, use of different shields or migration of the proton beam over the target.

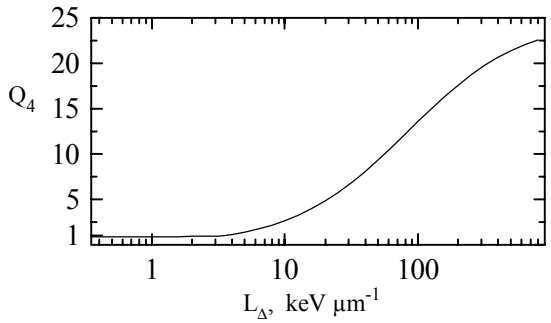

Figure 6. Dependence of recombination index of radiation quality Q_4 on L_Δ.

The radiation quality factor, as given by the Q(L) relationship in the ICRP Report 60 (22), can be determined by the recombination method, based on the mathematical analysis of the saturation curve (5, 23). The measurements and calculations are much more tedious, then in the case of RIQ, because whole saturation curve should be determined (7). Usually, such measurements should be performed only once, for the particular radiation field. Then, the possible changes can be much faster monitored by measuring RIQ, because for small changes of radiation quality, both quantities can be considered as proportional one to another (24).

In last years, some special attention was paid to determination of dosimetric parameters of neutron beams of few tens of MeV. Within this project, the recombination chamber of F-1 type was used at PSI for determination of the absorbed dose and dose equivalent at different depths in tissue.

Saturation curves of the chamber were measured free-in-air or behind some additional layers of A-150, provided by a cap and a block of A-150 plastic with wall thickness of 15 and 28 mm respectively. The values of the absorbed dose (relatively to the charge on the beryllium target) are shown in Table 1, together with the experimentally obtained values of the recombination index of radiation quality. The next columns of the Table present the values of radiation quality factor Q, calculated as:

$$Q = \frac{1}{D}\sum_i D_i Q(L_i) \tag{2}$$

where:

- index i numbers 8 compartments of LET, chosen for determination of the dose versus LET distribution;
- D_i are the fractions of the absorbed dose, associated with the particular compartment i, derived by the analysis of the shape of the saturation curve (5, 23);
- $Q(L_i)$ is the average value of the Q(L) function (22) for the compartment i;
- D is the total absorbed dose.

The value of the dose equivalent can be calculated as the product of the absorbed dose and Q, using the values given in the Table 1. All the presented values are in agreement with the appropriate values measured using a TEPC counter (25).

Table 1. The absorbed dose (in Gy per C of the beam current), RIQ and radiation quality factor determined by recombination method in neutron radiation fields at PSI, generated by protons of energy E_p.

Wall thickness	E_p = 44 MeV			E_p = 71.25 MeV		
(A-150)	D [Gy/C]	RIQ	Q	D [Gy/C]	RIQ	Q
6 mm	1.293	7.24	8.0	1.395	6.96	
21 mm	1.318	7.04	7.65	1.770	6.24	7.2
34 mm	1.104	6.83				
49 mm	1.070	6.59				

As it could be expected, the use of the 15 mm thick build-up cap increased the absorbed dose in the field produced by 71.25 MeV protons by 27% comparing with the irradiation free-in-air. This was accompanied by decrease in RIQ value by about 11%. The effects of build-up in absorbed dose and accompanying changes in radiation quality were much less pronounced in the field produced by 44 MeV protons, where the maximum of the absorbed dose was at lower depth. In this case the use of the 15 mm cap resulted in the increase of the absorbed dose by 3.6% and small decrease in RIQ, by only 1.5%.

The F-1 chamber was previously used (6) for in-phantom measurements in medical beams of high-energy neutrons generated by 660 MeV protons in Joint Institute for Nuclear Research in Dubna (Russia). The results of measurements, presented in this work showed that the chamber could be used for similar purposes also in neutron beams with energy of few tens of MeV.

HIGH-PRESSURE C-CO_2 CHAMBER AS A "NEUTRON-INSENSITIVE" DETECTOR

Measurements of the absorbed dose in mixed neutron-photon fields often involve separate determination of neutron and photon dose components. This approach mainly concerns mixed neutron-photon fields with neutron energies lesser than about 20 MeV. In such fields, the neutron and photon components of the absorbed dose can be determined separately by the twin-detector method (26) using two detectors of different neutron sensitivity. Commonly, two ionization chambers are used, for example, a tissue-equivalent (TE) ionization chamber combined with a C-CO_2, "neutron-insensitive" chamber. Because the C-CO_2 chambers have also some sensitivity to neutrons, the classical method requires information of the neutron spectral fluence of the radiation field, the sensitivity of the detectors as a function of neutron energy, and the kerma factors (fluence-to-kerma conversion coefficients) in materials of both detectors. For neutron energies above 20 MeV such data have relatively high uncertainties.

Usually, C-CO_2 chambers are operated at atmospheric gas pressure and their relative neutron sensitivity k_U - i.e. the ratio of its sensitivity to neutrons to its sensitivity to photons - is below 0.1 up to neutron energies of about 8 MeV. Above this energy, the k_U value increases due to the increasing neutron interaction cross sections of carbon and oxygen and reaches a value of about 0.3 for 15 MeV neutrons (27).

In our method, the high-pressure C-CO_2 chambers are operated at low collecting voltages. Under these conditions, the recombination of the ions for tracks of high linear energy transfer (LET) is considerably greater than for low-LET tracks. As a result, the relative neutron sensitivity k_U of such chambers is much lower than those operated at saturation and depends on the voltage applied. The earlier performed calculations and measurements (10, 5) showed that for chambers filled with CO_2 under the pressure of few MPa and neutron energies of up to 15 MeV, the value of k_U is below 0.03, supposing that the collecting electric field strength in the chamber cavity does not exceed 200 V/cm.

Recently, the method was extended (28) to neutron fields with energies of up to few tens MeV. The developed theory and measurements in the above described neutron fields at PSI and UCL resulted in the values of k_U equal to 0.075 and 0.085 for the neutron beams of 44 MeV and 66 MeV, respectively, for the chamber operating at a gas pressure of 3.4 MPa and collecting voltage of 20 V (Figure 7).

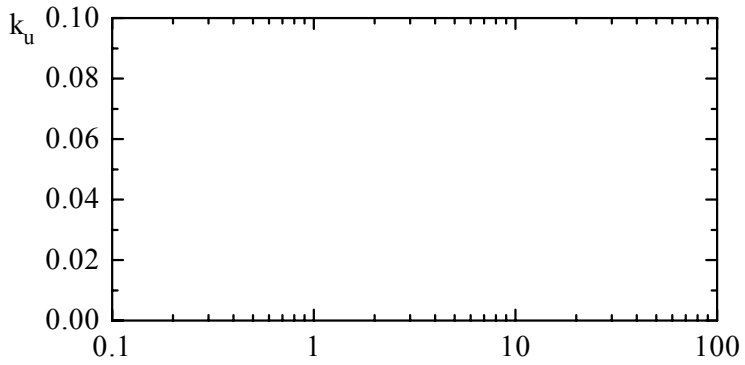

Figure 7. Relative neutron sensitivity k_U of the high-pressure C-CO_2 chamber of G-5 type operated at gas pressure of 3.4 MPa and collecting voltage of 20 V. Solid line - calculated values. Dashed line - guide for eye.

THIN-WALL RECOMBINATION CHAMBER FOR DOSIMETRY OF WEAKLY PENETRATING RADIATION

Ionization chambers operated at saturation are recommended by ICRU (29) and widely used for determination of H'(0.07) and H'(3) in fields of low energy photons and electrons. They, however, cannot provide any information about radiation quality, so their application in fields containing low energy alpha particles or heavy ions is much limited. As mentioned above, the problem was solved by use of a specially designed thin-wall recombination chamber KR-16.

The construction of the chamber was preceded by basic physical studies of the initial recombination in different gases. Recombination chambers are usually filled with methane or so called tissue-equivalent methane-based mixture up to the pressure of several hundreds kilopascals. The wall thickness ensuring the mechanical strength is few millimeters of tissue-equivalent plastic or 1÷2 mm aluminum. Such a wall thickness enables determination of ambient dose equivalent, H*(10) but neither H'(0.07) nor H'(3). The chamber for measurements of these two quantities should be wall-less or the wall surface mass should not exceed 7mg cm^{-2}. Therefore, the chamber has to be filled with a gas under atmospheric pressure. This creates a problem of competition between initial and volume recombination, especially at low collecting voltages, which have to be used for determination of radiation quality factor. This competition is the main factor limiting the range of the dose rate for which the dose equivalent can be determined with good accuracy. The higher is the gas pressure in the chamber the higher electrical field strength can be used and the higher is the upper limit of the dose rate.

Special series of experiments were performed in order to show that the proper conditions of dominating initial recombination could be achieved also under atmospheric gas pressure, provided that heavy gases are used. According to the theory of initial recombination (5), the initial recombination should be higher for gases with larger molecular weight, therefore it was expected that heavy gases should be used at atmospheric pressure. Another criterion for the choice of the filling gas was the atomic composition of the molecule, which should be equivalent to the soft tissue from dosimetric point of view. Usually, the most critical parameter for dosimetry of mixed radiation fields is content of hydrogen, especially when neutrons are involved. Both soft tissue and so called ICRU-tissue, for which the dose equivalent quantities are defined, contain about 10% of hydrogen by weight. The gases normally used in recombination chambers have slightly higher content of hydrogen in order to ensure an equal sensitivity of the chambers to neutron and photon radiation.

Heavy gases, which should be used for the thin-wall chamber, have lower content of hydrogen comparing to tissue. Nevertheless they can be considered as suitable for the determination of H'(0.07) as this quantity concerns only weakly penetrating radiation and is of minor importance for neutrons. Therefore, in this case the proper content of hydrogen is no more the crucial parameter of the gas. Finally, butadiene (C_4H_6) was chosen for the present work, because of its availability, sufficiently high density and reasonable equivalency to tissue.

Studies of initial recombination of ions under atmospheric gas pressure

In order to determine the initial recombination in presence of the volume recombination, the measurements with the REM-2 chamber were performed at different dose rate values. The chamber was placed in radiation field of ^{137}Cs source with dose rate values ranging from 0.05 mGy/h to 25 mGy/h. The results obtained for atmospheric gas pressure are shown in Figure 9 where the upper curve concerns the ideal conditions of initial recombination only. The value of maximum dose rate, $\dot{D}_{max}$ determined from the measurement was equal to about 20 mGy/h, i.e. only minor correction for volume recombination, if any, is needed below this dose

rate. As it was expected the $\dot{D}_{max}$ increases at elevated gas pressures to the value of 40 mGy/h at 127 kPa and to 150 mGy/h at 204 kPa, comparing with $\dot{D}_{max}$ = 200 mGy/h for the same chamber filled with methane up to much higher pressure of 1.2 MPa.

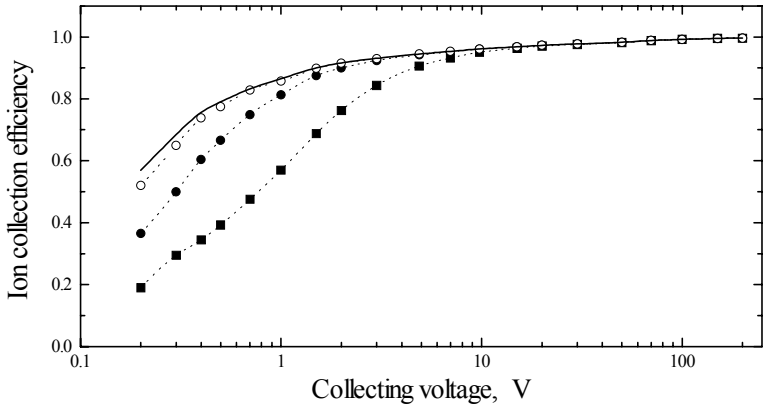

Figure 8. Saturation curves for gamma (^{137}Cs) radiation measured by REM-2 chamber filled with butadiene at the gas pressure of 100 kPa at dose rates of 25 mGyh^{-1} (solid squares), 1.9 mGyh^{-1} (solid circles) and 0.2 mGyh^{-1} (open circles). The upper curve represents the ion collection efficiency for only initial recombination.

The recombination index of radiation quality Q_R was measured in the radiation field of bare ^{239}Pu-Be neutron source at the dose rate of about 1.5 mGy/h. The values of $Q_R = 6.3 \pm 0.7$ and $Q_R = 7 \pm 0.5$ were obtained for the low-pressure chamber filled with butadiene and high-pressure chamber filled with methane, respectively.

The experimental results obtained in this work gave the positive answer to the basic question whether the conditions of dominating initial recombination of ions can be achieved in a gas under a near-atmospheric pressure if the chamber is filled with sufficiently heavy gas. The upper limit of the dose rate appeared to be high enough for radiation protection purposes. The KR-16 chamber filled with butadiene at atmospheric pressure can be used up to dose rate of 30 mGy/h provided that the electrical field strength ensures at least 96% of ion collection in gamma radiation field.

Tests of the thin-wall chamber

The tests of the thin walled chamber were performed mainly in reference radiation field of a ^{137}Cs isotope source. It was proved that the chamber was working correctly and could ensure repeatability of the results within statistical uncertainty of measurements. The dark current of the chamber (without radiation) was below 3 fA. There was no problem to achieve the saturation conditions for gamma radiation. The value of the recombination voltage U_R that ensured 96% of ion collection efficiency in the gamma radiation field was equal to 10 V. The saturation curve of the chamber with an additional guarding foil was almost the same when measured at positive or negative polarities of the collecting voltage. The working conditions are achieved after relatively short time of about 10 minutes after switching on the gas flow through the chamber.

After three months of intensive work with the chamber and irradiation with the dose of about 10 Gy we could observe that some heavy hydrocarbon fraction appeared on inner part of the chamber. The Mylar electrode became degraded and had to be replaced by a new one. The replacement is not difficult in laboratory conditions but inconvenient for routine radiation protection.

Saturation curves of the KR-16 chamber were measured in radiation fields of gamma (^{137}Cs) and alpha (^{238}Pu) radiation. The gamma radiation was used mainly for calibration of the chamber, the alpha radiation was an example of weakly penetrating radiation for which the H'(0.07) value should be determined. Since the range of alpha particles outgoing from the ^{238}Pu source was less than 7 mg/cm^2, therefore, the saturation curves for alpha radiation were measured using the entrance window formed by collecting electrode, i.e. by only one Mylar foil. The necessary information on radiation quality was derived from the shape of the saturation curve.

The value of the recombination index of radiation quality measured by the thin-wall KR-16 chamber for ^{238}Pu alpha particles was equal to Q_R=14.6±1.5. Such a value is in a good agreement with the expected value of the radiation quality factor, like it was defined in ICRP Report 21 (21). The value according the more recent recommendation of ICRP Report 60 (22) can be also calculated from Q_R according to the approach given in Ref. 24. In case of alpha particles used in this work, the calculation procedure results in the value of $Q = Q_R = 15 \pm 3$.

Additionally, the depth distribution of the absorbed dose gamma radiation source was measured for the photon (^{137}Cs) and neutron (^{239}Pu-Be) sources using paper and Plexiglas layers placed in front of the entrance

window of the chamber. The obtained results are shown in Figure 10. As it could be expected, some small increase of the absorbed dose is observed at small depths. Most likely this indicates that the electron equilibrium is not achieved at such depths. The slope of the curve in Figure 10 at depth above 3 mm is in reasonable agreement with the value of the absorption coefficient for gamma radiation of ^{137}Cs source in tissue-equivalent material. Presence of a weakly penetrating component is also evident.

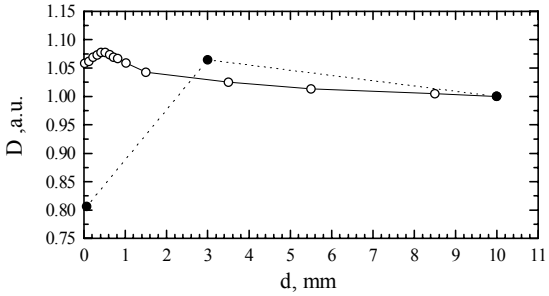

Figure 9. Depth distribution of the absorbed dose for gamma (^{137}Cs - open squares) and mixed neutron-gamma (^{239}Pu-Be - solid circles) radiations. Lines are guides for eye.

CONCLUSIONS

Recombination methods were invented almost contemporaneously with the ICRU/ICRP concept of radiation quality factor expressed in terms of LET. Since then, the recombination chambers and related methodology have been considerably developed following development and new concepts of dosimetry and radiation protection. The overview of the recently developed methods, presented in this work, should support the authors' opinion, that the abilities of the methods based on phenomenon of initial recombination of ions are still far from exhaustion.

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