Radiation Protection Dosimetry (2015), pp. 1–5 doi:10.1093/rpd/ncv293

EQUIVALENCE OF PURE PROPANE AND PROPANE TE GASES FOR MICRODOSIMETRIC MEASUREMENTS

S. Chiriotti[1,2,3,*], D. Moro[3], P. Colautti[3], V. Conte[3] and B. Grosswendt[4]
[1]Belgian Nuclear Research Centre, SCK•CEN, Mol, Belgium
[2]Center of Molecular Imaging, Radiotherapy and Oncology, Institut de Recherche Expérimentale et Clinique, Université catholique de Louvain (UCL), Brussels, Belgium
[3]Laboratori Nazionali di Legnaro, INFN-LNL, Legnaro, Italy
[4]Guest at LNL-INFN, Legnaro, Italy

*Corresponding author: scalvare@sckcen.be

A tissue-equivalent proportional counter (TEPC) simulates micrometric volumes of tissue if the energy deposited in the counter cavity is the same as that in the tissue volume. Nevertheless, a TEPC measures only the ionisations created in the gas, which are later converted into imparted energy. Therefore, the equivalence of the simulated diameter ($D\rho$) in two gases should be based on the equality of the mean number of ions pairs in the gas rather than on the imparted energy. Propane-based tissue-equivalent gas is the most commonly used gas mixture at present, but it has the drawback that its composition may change with time. From this point of view, the use of pure propane offers practical advantages: higher gas gain and longer stability. In this work, microdosimetric measurements performed with pure propane, at site sizes 0.05 mg cm^{-2} ≤ $D\rho$ ≤ 0.3 mg cm^{-2}, demonstrate that the response of a propane-filled detector in gamma and in neutron fields is almost the same if an appropriate gas density is used.

INTRODUCTION

In microdosimetry, tissue-equivalent proportional counters (TEPCs) are commonly assumed to measure the distribution of energy imparted in micrometric volumes of tissue when irradiated by ionising radiation. To achieve this aim, at least the elemental composition of the walls and the filling gas of a TEPC should be as close as possible to that of tissue. In this respect, propane-based tissue-equivalent (C_3H_8–TE) gas[(1)] is generally accepted as a good approximation for the elemental composition of tissue and represents at present the most frequently used filling gas of TEPCs. Particularly in sealed TEPCs, the drawback of using gas mixtures is due to the fact that the composition of the filling gas may change with time due to a different absorption of the gas mixture components with the detector walls. From this point of view, the use of pure propane as the filling gas of a sealed TEPC offers practical advantages: (1) the composition of the filling gas is more stable on long-time operation, and (2) higher gas gains can be reached when compared with the C_3H_8-TE gas mixture. The disadvantage of using pure C_3H_8 gas instead of the C_3H_8-TE gas is related to the different atomic composition of C_3H_8 (mass fractions: 18.3 % H, 81.7 % C) when compared with that of the commonly accepted C_3H_8-TE gas mixture (mass fractions: 10.3 % H, 56.9 % C, 3.5 % N, 29.3 % O).

Sometimes, in particular for measurements in space environment, the use of pure C_3H_8 is preferred. It is often considered to have nearly the same response as C_3H_8-TE. Therefore, it was the aim of the present article to develop an experimental procedure that can be applied in measurements with C_3H_8-filled TEPCs to get a detector response very similar to that of TEPCs filled with the C_3H_8-TE gas mixture. Due to the fact that proportional counters measure the charge distribution caused by ionising radiation instead of the distribution of energy imparted, the attention of this work was focussed more on the ionisation cross sections in the two gases rather than on the corresponding stopping powers.

MATERIAL EQUIVALENCE

In radiation dosimetry, two sites of different materials are considered equivalent if the absorbed dose or the mean imparted energy ε to each site are equal. Applying this principle to the sensitive volume of a TEPC filled either with the C_3H_8-TE gas mixture or with pure C_3H_8 requires that the mean energy imparted, which is caused by a charged particle (e.g. an electron or a proton) when penetrating through the detector volume on one of its diameters D, is independent of the filling gas. If the secondary electrons are completely absorbed inside the detection volume, the material equivalence can be expressed by the following equation:

$$(S/\rho)^{C_3H_8} \cdot (D\rho)^{C_3H_8} = (S/\rho)^{C_3H_8-TE} \cdot (D\rho)^{C_3H_8-TE} \quad (1)$$

Here, $(S/\rho)^{C_3H_8-TE}$ and $(S/\rho)^{C_3H_8}$ are the mass collision stopping powers of the primary particle in the C_3H_8-TE and in pure C_3H_8 gases, respectively; $(D\rho)^{C_3H_8-TE}$ and $(D\rho)^{C_3H_8}$ are the masses per area of

the site diameter in the two different gases and ρ is the corresponding gas density. Equation (1) is valid only for particles that have ranges markedly larger than the target size. If the mass stopping powers are the same in the two gases for all primary particle types and energies, the site sizes of the two filling gases expressed in mass per area would be the same.

In view of the different atomic compositions of the two filling gases, however, the mass stopping power ratio $(S/\rho)^{C_3H_8-TE}/(S/\rho)^{C_3H_8} \neq 1$ and depends on particle type and energy. Therefore, the ratio $(D\rho)^{C_3H_8}/(D\rho)^{C_3H_8-TE}$ varies in the same way.

In addition to these principle problems in the definition of material equivalence, one should also bear in mind that, instead of the distribution of energy imparted, a TEPC measures the distribution of ionisations created in the radiation-sensitive volume of the counter, which are converted later to a distribution of energy imparted using an appropriate calibration factor. In view of this fact, the material equivalence of different filling gases of a TEPC should be based more on the equality of ionisation distributions rather than on the equality of the distributions of imparted energy.

To define material equivalence based on ionisations, assume that single primary particles are penetrating through the detection volume of a TEPC on one of its diameters D. If the range of the primary particles is markedly longer than the diameter of the gas cavity, the mean number of primary ionisations caused by a particle inside the detection volume is given by the ratio $(D\rho)/(\lambda\rho)_{ion}$, where $(D\rho)$ is the mass per area of the particle's track length within the target and $(\lambda\rho)_{ion}$ is the mass per area of the mean free path length with respect to primary ionisation. $(\lambda\rho)_{ion}$ is proportional to the reciprocal of the ionisation cross section of the target material and depends on the type and energy of ionising particles.

TEPCs filled with pure C_3H_8 have the same response function as TEPCs filled with the C_3H_8-TE if the measured ionisation spectra are the same for the same radiation field. This means that, at least, the number of primary ionisations must be the same:

$$\frac{(D\rho)^{C_3H_8}}{(\lambda\rho)_{ion}^{C_3H_8}} = \frac{(D\rho)^{C_3H_8-TE}}{(\lambda\rho)_{ion}^{C_3H_8-TE}} \qquad (2)$$

Here, $(\lambda\rho)_{ion}^{C_3H_8}$ and $(\lambda\rho)_{ion}^{C_3H_8-TE}$ are the masses per area of the mean free ionisation path lengths for C_3H_8 and the C_3H_8-TE gas mixture. Equation (2) shows the same structure as Equation (1) with (S/ρ) exchanged by the reciprocal of $(\lambda\rho)_{ion}$, and the discussion of Equation (1) is also valid for Equation (2). Therefore, the mass per area $(D\rho)^{C_3H_8}$, which is equivalent to $(D\rho)^{C_3H_8-TE}$, varies as a function of energy and particle type like $(\lambda\rho)_{ion}^{C_3H_8}/(\lambda\rho)_{ion}^{C_3H_8-TE}$.

To give an impression of the differences in generating primary ionisations in pure C_3H_8 and in the C_3H_8–TE gas mixture, Figure 1 shows the mean free ionisation path lengths $(\lambda\rho)_{ion}^{C_3H_8}$ and $(\lambda\rho)_{ion}^{C_3H_8-TE}$ for electrons and protons as a function of the particle energy E. For a detailed description of the calculation of ionisation path lengths for electrons and protons in C_3H_8 and the C_3H_8-TE gases, see Refs. [2,3].

At first glance, it can be seen in Figure 1 that for electrons and protons $(\lambda\rho)_{ion}^{C_3H_8}$ is always smaller than $(\lambda\rho)_{ion}^{C_3H_8-TE}$ independently of the particle energy. This means that, exposed to the same irradiation field, the amount of primary ionisations formed in the detection volume of a TEPC is always greater if pure C_3H_8 instead of the TE gas mixture is used. Hence, also a higher gas gain can be reached when using C_3H_8.

To be able to estimate the site size $(D\rho)^{C_3H_8}$ of a TEPC filled with pure C_3H_8, which leads to the same response function as the TEPC filled with the C_3H_8-TE gas mixture according to Equation (2), the conversion factor $(\lambda\rho)_{ion}^{C_3H_8}/(\lambda\rho)_{ion}^{C_3H_8-TE}$ is plotted in Figure 2 for electrons and protons as a function of particle energy E.

As it is obvious from a first glance at Figure 2, the ratio $(\lambda\rho)_{ion}^{C_3H_8}/(\lambda\rho)_{ion}^{C_3H_8-TE}$ as a function of energy is smaller and less dependent on energy than the corresponding ratio of (S/ρ), in particular for the protons.

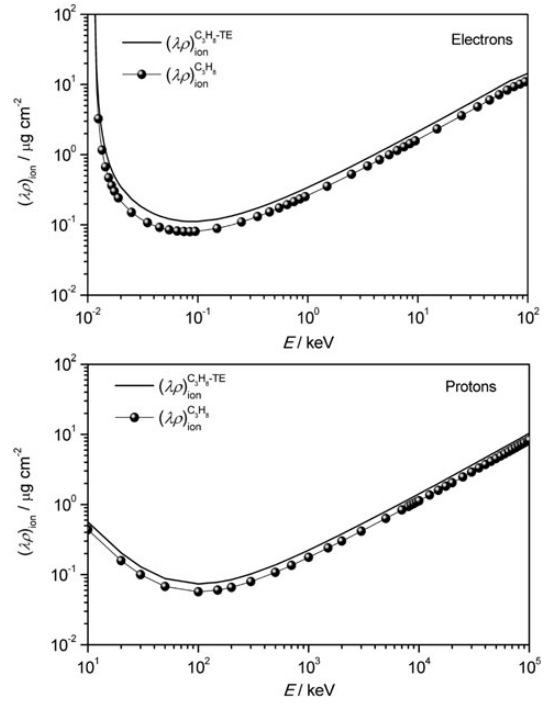

Figure 1. Primary ionisation mean free path of electrons (top) and protons (bottom) in C_3H_8 and C_3H_8-TE gases.

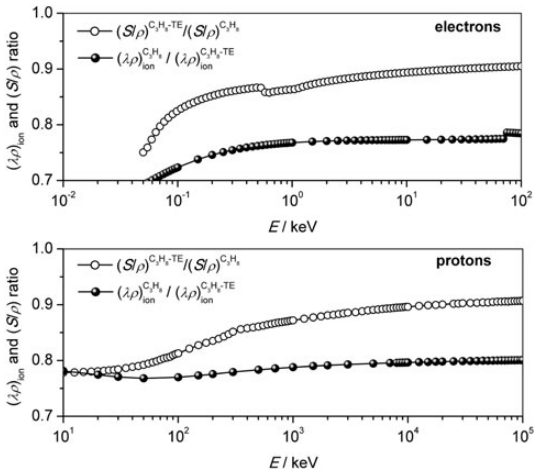

Figure 2. Ratio $(\lambda\rho)_{ion}^{C_3H_8}/(\lambda\rho)_{ion}^{C_3H_8-TE}$ for electrons (top) and protons (bottom). (S/ρ) for protons are taken from Ref. [4] and (S/ρ) for electrons from Ref. [5].

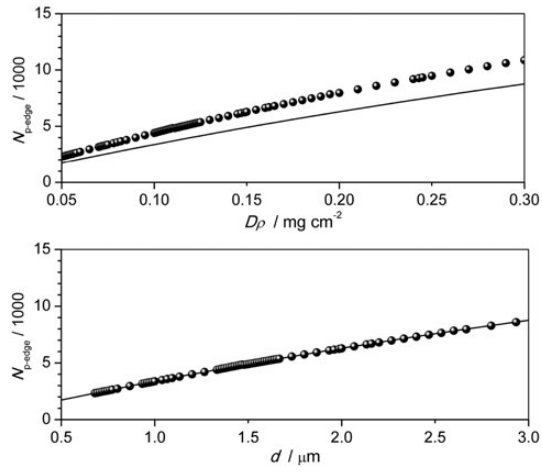

Figure 3. $N_{p\text{-}edge}$ in C_3H_8-TE (line) and C_3H_8 (symbols) as a function of mass per area $D\rho$ (top) and as a function of equivalent site size d at density $\rho = 1\ g\ cm^{-3}$ (bottom).

In addition, it is almost the same for electrons in the energy range between 1 and 10^3 keV and for protons at energies between 10 and 10^5 keV. A similar response of the TEPC is therefore expected if propane is set to a lower gas density with respect to the TE gas, according to the $(\lambda\rho)_{ion}$ ratio, which is between 0.75 and 0.8.

Ideally, the number of measured ionisations should be the same for the whole energy spectrum of incoming particles, in particular in the proton-edge region, which is often used for calibration. The number of ionisations produced by protons at the p-edge can be expressed as

$$N_{p-edge}^{gas}(D\rho^{gas}) = y_{p-edge}^{gas}(D\rho^{gas}) \times \bar{l}/W^{gas}$$

Here, y_{p-edge}^{gas} is the corresponding lineal energy calculated from the energy range tables published by International Commission on Radiation Units (ICRU)[4], W is the mean energy required to form an ion pair by protons[6] in a given gas and $\bar{l}$ is the mean chord length of the cavity. In Figure 3 (top), the number of ionisations for the two gases, N_{p-edge}^{C3H8} and $N_{p-edge}^{C3H8-TE}$, is plotted as a function of the site size $D\rho$. Due to the shorter ionisation mean free path length in propane, $N_{p-edge}^{C3H8} > N_{p-edge}^{C3H8-TE}$. In order to have $N_{p-edge}^{C3H8} = N_{p-edge}^{C3H8-TE}$, the density of propane must be reduced. If the N_{p-edge}^{C3H8} data are shifted to larger values of $D\rho$ by a factor of $(0.75)^{-1}$, they perfectly agree with the $N_{p-edge}^{C3H8-TE}$ data. For instance, in pure propane at $(D\rho)^{C_3H_8} = 0.075\ mg\ cm^{-2}$, $N_{p-edge}^{C3H8} = 3373$ is obtained, which is almost the same as $N_{p-edge}^{C3H8-TE} = 3375$ at $(D\rho)^{C_3H8-TE} = 0.1\ mg\ cm^{-2}$.

In order to have the same number of ionisations $N_{p\text{-}edge}$ in both gases, the gas density of pure C_3H_8 must be reduced by a factor of 0.75 when compared with the gas density of C_3H_8-TE. Consequently, to get the same equivalent site size d, the following equations are used:

$$d = \frac{(D\rho)^{C_3H_8-TE}}{(1\ g\ cm^{-3})};\ d = \frac{(D\rho)^{C_3H_8}}{0.75 \times (1\ g\ cm^{-3})} \qquad (3)$$

If plotted as a function of d, the number of ionisations in the two gases is almost the same (bottom Figure 3).

MATERIALS AND METHODS

The measurements were performed at INFN-LNL using a spherical TEPC with an internal diameter of 5 cm, an anode diameter of 100 μm and 3-mm-thick walls made of tissue-equivalent plastic (A-150). The TEPC was filled either with pure C_3H_8 gas or with the C_3H_8-TE gas mixture. The mass per area of the cavity diameter $D\rho$ was varied between 0.05 and 0.3 mg cm^{-2} by changing the gas density. To study the detector response for low linear energy transfer (LET) fields, the TEPC was exposed to a ^{137}Cs γ-source, whereas for high-LET fields, it was exposed to fast neutrons, 0.58 MeV in mean energy, produced from the $^{8}Be(p,n)$ reaction by bombarding a thick beryllium target with 3 MeV protons at the CN Van de Graaff accelerator of LNL. For both filling gases, the microdosimetric spectra for the ^{137}Cs and the neutron fields were collected at 10 different gas densities, giving a total of 40 spectra. At each density, the neutron and photon spectra were measured in the same working conditions of the TEPC.

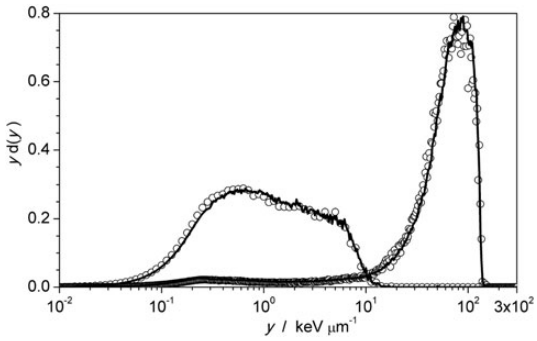

Figure 4. ^{137}Cs (left) and neutron microdosimetric spectra measured at $d = 2$ µm: C_3H_8-TE (line) and C_3H_8 (circles).

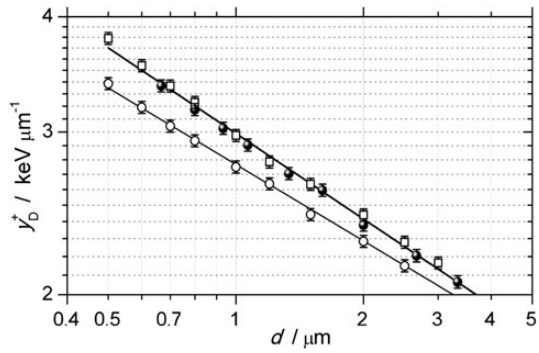

Figure 5. Dose-mean lineal energy y_D^+ for ^{137}Cs as a function of the equivalent site size d for C_3H_8 (circles) and C_3H_8-TE (squares). For details, see the text.

The neutron spectra for both gases were calibrated using the proton edge, by applying the technique discussed in detail by Conte *et al.*[7]. The most precise marker point (h_{TC}) is given by the intercept of the tangent through the inflection point of the fitted Fermi function with the abscissa. The lineal energy values at the p-edge, $y_{p-edge}(d)$, were calculated using the energy range tables published by ICRU[4] for the TE gas mixture. Afterwards, they were assigned to the marker h_{TC}. For each density, the same calibration factor was applied to the ^{137}Cs spectrum. To give an example, the neutron spectra measured in the two gases, at $(D\rho)^{C_3H_8} = 0.075\,mg\,cm^{-2}$ in propane and at $(D\rho)^{C_3H8-TE} = 0.1\,mg\,cm^{-2}$ in the TE gas, were calibrated, according to Equation (3), using $y_{p-edge}(1\,\mu m)$ for the TE gas mixture.

RESULTS AND DISCUSSION

To give an impression of the results, Figure 4 shows the microdosimetric spectra for ^{137}Cs on the left and for 0.58 MeV neutrons on the right at equivalent site size $d = 2$ µm. A good correspondence in the shape of the spectra is observed with only minor differences in the gamma spectra. A similar good agreement was observed also for the other gas densities.

To get an overview of the results in the whole range of site sizes, the dose-mean lineal energy for the gamma component was calculated. As the lower threshold of the spectral distributions was set at the noise level $y = 0.4$ keV µm^{-1}, this dose-mean lineal energy was indicated as y_D^+.

Figure 5 shows y_D^+ in pure propane (spheres) and in the TE gas mixture (squares) as a function of the equivalent site size d: the agreement between the y_D^+ for propane and for the TE gas as a function of the equivalent site size d is almost perfect within the estimated 'type A' standard uncertainties of 1.5 %. Within these uncertainties, the experimental data can be described by a power function of d (the straight lines in the figure). The y_D^+ in propane obtained without taking into account the 0.75 factor in Equation (3) is also shown (open circles). In this case, an underestimation between 5 and 10 % clearly demonstrates the improvement by using the 0.75 factor.

CONCLUSIONS

The aim of this work was to investigate how different is the response of a TEPC filled with pure C_3H_8 instead of the common C_3H_8-TE gas, in gamma and neutron fields. The comparison of the microdosimetric spectra for ^{137}Cs and neutron fields, in the equivalent diameter range investigated in this study, confirms that the C_3H_8-filled TEPC has almost the same response function as the C_3H_8-TE-filled detector if the mass per area in pure C_3H_8 is reduced by a factor of 0.75 when compared with C_3H_8-TE. By applying this factor, pure propane gas can be used as a substitute of the TE gas mixture. The microdosimetric spectra are almost the same, and the derived dose-mean lineal energies do not differ significantly, within the estimated uncertainty of 1.5 %. The applicability of Equation (3) in mixed fields and higher neutron energies is under investigation.

FUNDING

This work is supported by the Fifth Scientific Commission of the Istituto Nazionale di Fisica Nucleare (INFN) of Italy.

REFERENCES

1. International Commission on Radiation Units and Measurements. *Microdosimetry*. ICRU Report 36 (1983).
2. De Nardo, L., Colautti, P., Conte, V., Baek, W. Y., Grosswendt, B. and Tornielli, G. *Ionization-cluster distributions of α-particles in nanometric volumes of propane:*

measurement and calculation. Radiat. Environ. Biophys. **41**, 235–256 (2002).
3. Grosswendt, B. *Formation of ionization clusters in nanometric structures of propane-based tissue-equivalent gas or liquid water by electrons and α-particles*. Radiat. Environ. Biophys. **41**, 103–112 (2002).
4. International Commission on Radiation Units and Measurements. *Stopping Powers and Ranges for Protons and Alpha Particles*. ICRU Report 49 (1993).
5. Salvat, F., Fernández-Varea, J. and Sempau, J. *PENELOPE—2011: A Code System for Monte Carlo Simulation of Electron and Photon Transport*. OECD Nuclear Energy Agency (2011).
6. Baek, W., Grosswendt, B. and Willems, G. *W Values of protons in alkane-based TE gases and dimethylether in the energy range 1 to 100 keV: measurements and additivity models*. Radiat. Prot. Dosim. **110**, 61–68 (2004).
7. Conte, V., Moro, D., Grosswendt, B. and Colautti, P. *Lineal Energy Calibration of Mini Tissue Equivalent Gas-Proportional Counters (TEPC)*. AIP Conf. Proc. **1530**, 171–178 (2013).