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The heat defect in the water absorbed dose calorimeter [1]

A. Krauss * and M. Roos

Physikalisch-Technische Bundesanstalt, Labor 6.43, 38023 Braunschweig (Germany)

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Abstract

The water absorbed dose calorimeter can be established as a primary standard for the quantity, water absorbed dose, if the heat defect problem can be solved.

The method chosen is an experimental absolute determination using a total absorption calorimeter for high energy electrons, so that the heat defect need not necessarily be zero, which is a crucial condition, but only stable. First results, including investigations of the stability of the heat defect for various aqueous solutions, are presented.

INTRODUCTION

The quantity water absorbed dose has been fixed as the measurand in dosimetry for radiation therapy. The definition of the absorbed dose as the quotient $d\bar{\varepsilon}/dm$, where $\bar{\varepsilon}$ is the mean energy imparted by radiation to matter of mass m, allows water calorimetry to be used for the realization of this quantity according to its definition and to establish a water calorimeter as a primary standard for the water absorbed dose.

The experimental method is based upon a measurement of the radiation-induced temperature rise at a point in a water phantom. The calorimeter must therefore be operated in such a way that convection is suppressed and heat conduction is reduced to such an extent that a measurement uncertainty can be achieved which is appropriate for a primary standard measuring system [1, 2].

Furthermore, it must be known what portion of the mean energy imparted to water is converted into heat. The relative deviation of that portion from unity is the so-called "heat defect" h of the water and is defined by the following expression:

$$D_w = c_p \Delta\theta (1 - h)$$

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* Corresponding author.

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where D_w is the water absorbed dose, c_p the specific heat capacity of water, and $\Delta\theta$ the radiation-induced temperature rise at the point of measurement. The heat defect is due to the overall exothermicity and endothermicity of the chemical reactions initiated by the radiolysis of the water.

In the following a brief summary of basic information on the heat defect and the various methods used to take this effect into account is given. The radiation chemistry of water can be described by the Boyd–Carver–Dixon model [3] which predicts, for absorbed doses that are not too low, a zero heat defect for pure water in the case of high energy photon or electron radiation independent of radiation quality. Detailed calculations [4, 5], particularly for pure water containing dissolved gases in millimolar concentration, again predict a zero heat defect for water saturated with N_2 or H_2 gas, but a stable value of −2.4% for water saturated with an equal mixture of H_2 and O_2 gas.

Caloric measurements using a small, sealable glass vessel filled with purified water [5], confirmed the relative differences in the extent of the heat defect for water containing the above-mentioned gases, but gave no independent information on the absolute value.

Experiments involving large water calorimeters [6–8] with stagnant, distilled water open to the atmosphere showed that the water absorbed dose values obtained were some percent higher than the absorbed dose values derived from ionization chamber, Fricke dosimeter or graphite dose calorimeter measurements under similar conditions. Such dose deviations were attributed to the heat defect to get rough estimates, the results not being supported by theory. In any case it should be noted that the heat defect may strongly depend on organic impurities in the water. These impurities could be initially dissolved in the distilled water or be introduced through the water's contact with the various materials of the calorimeter.

However, although these experiments are not suited for an absolute determination of the heat defect for the water qualities (distilled water, air saturated) used in the calorimeters, they produce stable results and therefore also a stable heat defect during all experiments as a function of accumulated dose or as a function of the time the water remained in the calorimeter tank. This is confirmed by experiments with the water absorbed dose calorimeter with air saturated water at this laboratory.

This important finding is supported to some extent by the model calculations [5], which show that specific additives can ensure a stable non-zero heat defect, as for the H_2/O_2 system, or may stabilize the heat defect by sidestepping some impurities reactions, as is expected for the H_2 system.

To establish the water calorimeter as a primary standard for the water absorbed dose, the heat defect must be determined theoretically or experimentally for the water quality actually used in the calorimeter.

The latter approach is the one used in the heat defect problem at this laboratory, where work is in progress to find a system with a stable heat defect, the absolute value of which can be measured in a separate experiment.

In the present paper two experimental methods developed in this laboratory to investigate the heat defect in detail are described. Measurements with the absorbed dose calorimeter are presented which investigate to what extent the radiation-induced temperature rise is stable in distilled water saturated with different gases. Using a differential experimental method measurements have been performed to determine the absolute value of the heat defect as a function of the accumulated absorbed dose.

EXPERIMENTAL SETUP AND PROCEDURE

The water absorbed dose calorimeter

The water absorbed dose calorimeter developed in this laboratory is based on the original design by Domen [6]. Several modifications were made [1], for instance to accomplish the use of horizontal beams and to apply the 4°C method for the suppression of convection. To investigate the influence of various gases dissolved in water on the heat defect problem, further modifications had to be made to allow the water to be saturated with a gas or a gas mixture under stable and reproducible conditions. Figure 1 shows a schematic diagram of the peripheral features of the calorimeter.

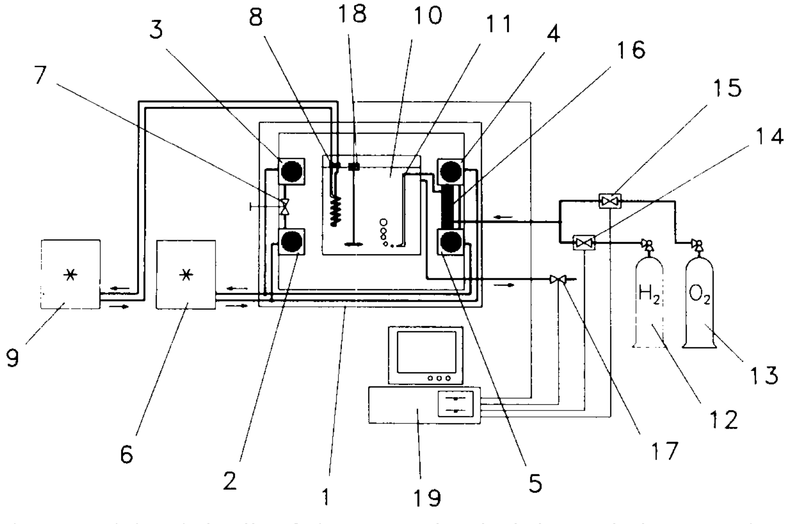

Fig. 1. Peripheral details of the water absorbed dose calorimeter. 1 is the temperature-stabilized outer enclosure with heat exchangers 2–5, cooler 6 and valve 7, to adjust the cooling liquid flow rate. A second heat exchanger 8 supplied by a separate cooler 9 allows acceleration of the cooling-down and heating-up procedures, for the water inside the perspex tank 10. Inside the tank are glass tubes 11 connected to the gas supply system 12,13 with gas flow meters 14,15, gas heat exchanger 16 and outlet valve 17. 18 indicates the mechanical stirrer and 19 the computer.

To saturate the water in the calorimeter with gas, several glass tubes reaching down to the bottom of the water-filled poly(methyl methacrylate) (PMMA) tank are connected to a gas supply system. Two computer-controlled gas flow meters (MKS 1159B) allow the gas flow to be adjusted within a range of 0–2000 $\mathrm{cm^3\,min^{-1}}$ with an uncertainty of 0.1%. Because the calorimeter is operated at a water temperature of 4°C, it is important to reduce the gas temperature to this level before bubbling the water. A special heat exchanger has therefore been designed, made from an aluminium block with dimensions of 21 cm × 12 cm × 6 cm through which three vertical channels with a diameter of 2.4 cm have been drilled. The channels are connected in series by small horizontal channels and are filled with special filter elements (sintered bronze) with holes of about 25 μm diameter. By adjusting the number of these filter elements and the gas flow, it is possible to bring the temperature of the gas flowing through the heat exchanger near to 4°C. The heat exchanger is fixed between two of the four actively cooled heat exchangers, which are placed on the inner walls of the temperature-stabilized enclosure of the calorimeter.

When the gas flow is switched on, a computer-controlled valve on the sealed lid of the calorimeter opens to the gas outlet tube.

The temperature-stabilized enclosure of the calorimeter consists of a wooden box with an 8 cm-thick polystyrene thermal shield against the surrounding room temperature. On two opposite inner walls of this box there are two pairs of actively cooled heat exchangers which are connected in parallel to a cooler. With the help of a manually adjustable valve it is possible to obtain equal flow rates of the cooling liquid (ethanol), resulting in all heat exchangers having a temperature level of about 2°C. On the front and the back of each exchanger there is a grid-shaped electrical heating element and a ventilator. These ventilators circulate the air around the body of the calorimeter and by switching the heating grids on and off by means of a computer-controlled electronic circuit the temperature of the circulating air is stabilized at 4°C.

During operation the temperature of the water inside the water phantom is maintained at 4°C by the temperature-stabilized enclosure and a deliberate reduction of the thermal shielding of the phantom. However, to accelerate the cooling-down and heating-up procedures, the water temperature can be changed by a heat exchanger inside the tank. This heat exchanger is connected to an additional cooler and can also be used to heat up the water.

Possible temperature gradients in the water must be eliminated before the series of measurement can be started. This can be done with the help of a computer-controlled mechanical stirrer. The computer also gets information on the temperature of the heat exchangers, the outer walls of the calorimeter and the circulating air from a total of 20 Pt-100 sensors.

To investigate the heat defect problem the water in the perspex phantom was saturated with a certian gas or gas mixture for more than 2 h with a gas flow of 500 $cm^3 min^{-1}$ before starting the series of caloric measurements. For measurements with the same gas the water was newly saturated in the morning of a new day. During the long-term ^{60}Co irradiation for the H_2 measurements, the water was bubbled with H_2 for 2 h (500 $cm^3 min^{-1}$) at time intervals of 12 h.

The irradiation time for a caloric measurement was about 120 s, producing a temperature increase at the point of measurement of about 0.5 mK, which corresponds to a water absorbed dose of about 2 Gy. After a series of normally eight irradiation runs the temperature gradients which had built up in the water were eliminated by stirring the water mechanically for about 15 min.

The irradiation geometry for the present experiment is identical to previous ones [2], but a ^{60}Co source with higher activity was used.

In this investigation caloric measurements were made with once-distilled water saturated with air, N_2, O_2, H_2 and a H_2/O_2 mixture (50:50) without changing the water with which the calorimete was filled. The same water was used in a second experiment for caloric measurements with the H_2 system as a function of the accumulated absorbed dose.

The total absorption calorimeter

The constructional and experimental details of the total absorption calorimeter have been described previously [9, 10]. This experiment allows the heat defect to be measured in absolute terms within an uncertainty of 0.5% for the correction factor. The method is based on the total absorption of 5 MeV electrons in a small water-filled vessel and an accurate measurement of the totally absorbed radiant energy of the electrons. In the first experimental step a relative measurement of the radiation-induced temperature increase was carried out and in the second step the same temperature increase was produced by electrical heating which does not result in a heat defect. The heat defect is the relative difference between the radiation energy W_r and the electrical heating energy W_e.

The calorimeter's stainless steel absorber vessel is filled with distilled water at a temperature of 30°C. This temperature is stabilized by a similar outer enclosure as described for the water absorbed dose calorimeter. The typical range of the accumulated absorbed dose extends from the order of 10 Gy up to about 200 Gy for a single measurement. For the results shown in Fig. 2 the irradiation time was about 150 s, which produced an increase in the temperature of the water of about 35 mK. This corresponds to a mean absorbed dose of about 140 Gy.

RESULTS AND DISCUSSION

Caloric measurements with the water absorbed dose calorimeter were carried out with various gases dissolved in water, i.e. air, N_2, H_2, O_2, and an equal mixture of H_2 and O_2. The total accumulated absorbed dose for these

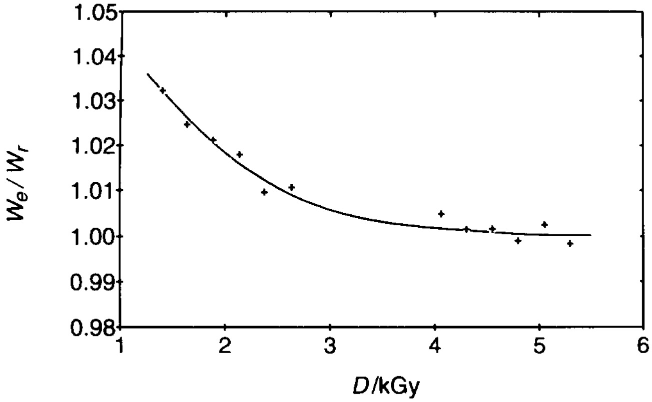

Fig. 2. Measurements with the total absorption calorimeter. Ratio of electrical heating energy and imparted radiation energy as a function of accumulated absorbed dose.

measurements ranged up to about 500 Gy and the relative standard devation of the mean of the measured temperature rise is about 0.1% for the measurements with a certain gas over a one-day period.

The relative differences in the calorimeter response for the different aqueous solutions produced in the same water filling are within a range of 1.2% with the mean value of the temperature increase for the H_2 system being nearly 1% higher than for the H_2/O_2 system.

When a series of caloric measurements is done on different days, as for the H_2 system, there is no significant change in the temperature rise measured, irrespective of whether the water has been saturated with another gas in between. These results indicate that the model calculations for pure water saturated with the gases mentioned are not directly applicable to the calorimeter system used in this experiment. The model [5] predicts relative differences with a bandwidth of about 4.5% with the H_2 system being 2.4% lower than the H_2/O_2 system.

To find a system with a stable heat defect it is important to investigate different aqueous solutions as a function of the accumulated absorbed dose. The first measurements were therefore made for the H_2 system in the dose range 0.5–13 kGy. The results do not allow definitive conclusions to be drawn, since other quantities of influence remain to be investigated, but they do show that the heat defect is at least stable within ±0.2%. Further detailed experimental investigations are needed for this and especially for the H_2/O_2 system, to prove the independence of the accumulated absorbed dose over a sufficiently wide range. Possible recovering effects between successive series of irradiation measurements must also be taken into account and investigated.

In a separate experiment the first measurements with the total absorption calorimeter were made to determine the absolute value of the heat defect. The stainless steel absorber vessel was filled with multiply-distilled water without saturating it with any gas.

The chief objective of this experiment was to prove the experimental method itself; therefore no great care was taken about impurities in the absorber vessel or on the detector materials. Caloric measurements were performed as a function of accumulated absorbed dose. Due to the performance of the beam transport system of the electron accelerator, the water inside the vessel was pre-irradiated up to an accumulated absorbed dose of about 1.3 kGy before irradiation measurements could be started. Figure 2 shows the results of these heat defect measurements. The ratio W_e/W_r of the electrical heating energy and the radiation energy imparted by the electrons to create the same temperature rise in the water, is shown. The heat defect h is therefore given by $1-(W_e/W_r)$. as can be seen from Fig. 2, the heat defect decreases with increasing accumulated dose and asymptotically reaches a value $h=0$ for an accumulated dose of about 5 kGy. This indicates that impurities, initially dissolved in the water, have been eliminated by the radiation.

For high purity pre-irradiated water a zero heat defect was measured with a total uncertainty of 0.44% for the respective correction factor [10].

CONCLUSIONS

The way to overcome the heat defect problem of the water absorbed dose calorimeter, chosen in this laboratory, is an experimental absolute determination for the water actually used in the absorbed dose calorimeter by means of a total absorption calorimeter, so the heat defect need not necessarily be zero but only stable. The water absorbed dose calorimeter gives additional information on the relative heat defect of various water qualities and its dependence on the accumulated dose.

Further detailed experiments are in progress to find a water system which offers a stable heat defect independent of accumulated dose or, for example, the time the water is kept in the calorimeter. These investigations are being performed using both types of calorimeter to bring the results into a broad overlapping region regarding the accumulated absorbed dose.

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