International dose-assurance in radiation technology

by K.H. Chadwick*

Radiation can solve some technological problems, and its application in modern society is increasing at a rate of about 15% each year [1]. Such applications range from food preservation to radiation cross-linked plastics; they are important to both the industrialized and the developing countries of the world.

Radiation can:

Prevent onions and potatoes from sprouting;
Disinfest insects in cereals, peas, beans, lentils, and dried food;
Destroy spoilage and pathogenic micro-organisms in food, e.g. salmonella in chicken;
Extend the shelf-life of fresh fish, fruit, and vegetables;
Sterilize a wide range of medical products;
Pasteurize or sterilize food-packaging materials;
Cure plastic coatings and paints;
Cross-link plastic insulation materials;
Make plastics heat-shrinkable;
Enhance rubber green strength for tyre construction;
Treat waste-water and sewage;
and the list is far from complete.

The extremely penetrating gamma- and X-radiations are used to treat bulk products, and modestly-penetrating, intense electron beams are used for surface or thin-product treatments. Doses extend from 10 Gy (1 krad) up to 100 kGy (10 Mrad). The correct use of radiation in preserving food and sterilizing medical products directly affects human health; in the treatment of plastics etc., the reliability of the finished product is at stake. Prior research and development relates the efficiency of each radiation process to the dose, so that in large-scale application the processor can use radiation dosimetry measurements during commissioning and operation as a form of quality control to guarantee the safety or reliability of the irradiated product [2, 3].

In scaling up from research to large-scale application, the processor moves from a situation where small samples are treated under carefully controlled conditions so that the radiation-dose is uniform throughout the sample, to the situation where bulk products have to be treated on an industrial scale and where it is impossible to ensure that all parts of the product receive the same dose. In this situation one has to accept a distribution of dose through the product and be able to measure that dose-distribution to ensure that it does not exceed the prescribed range.

Large industrial irradiation facilities are designed to deliver as uniform a dose-distribution in the product as possible. In gamma-ray facilities, boxes of product are moved past a large plaque-type radiation source. The surface of the box is thus uniformly irradiated so that the dose varies only within the product. Usually the boxes are irradiated from both sides to improve the uniformity in depth-dose but as it is impossible to make the dose exactly the same throughout the product there are always areas of maximum dose – on the outside of the box – and areas of minimum dose – in the mid-plane of the box. In the gamma-ray facility for food irradiation at Wageningen (Netherlands) for example, containers enter the irradiation chamber via a labyrinth and make two passes on either side of the source plaque and are thus irradiated from two sides. The speed of the container past the source plaque determines the dose acquired by the product: the higher the speed, the lower the dose. Figure 1 illustrates the distribution of dose for a one-sided and two-sided irradiation and shows the positions of maximum and minimum dose [3].

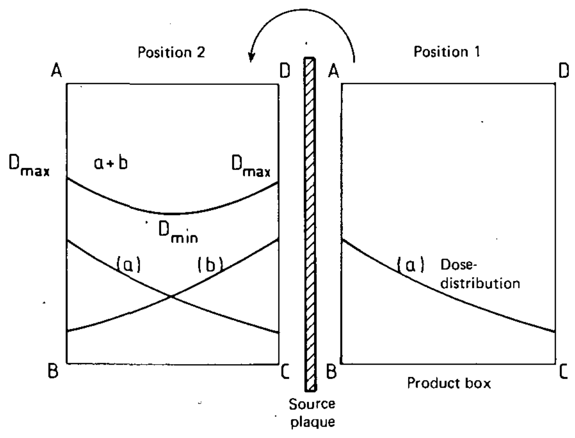

Figure 1. A representation of the depth-dose distributions in a product following one- and two-sided irradiation indicating the positions of maximum and minimum dose.

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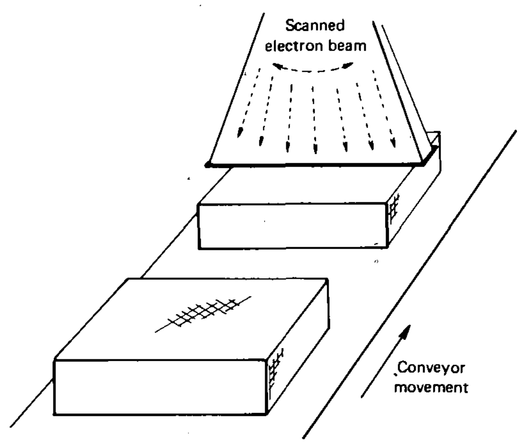

Figure 2. A schematic diagram showing a product box passing under a scanned electron beam in a one-sided irradiation.

In an electron-beam facility the product passes in one direction through the beam which is rapidly scanned perpendicularly to ensure a uniform surface-dose to the product. The dose-distribution varies as a function of depth in the product but is more complicated than for gamma rays. Figure 2 illustrates the passage of product under a scanned electron beam and Figure 3 presents the distribution with depth of dose.

Dosimetry problems in radiation processing

Problems may be encountered in the routine control of an industrial radiation process because conditions in the large irradiation facility differ considerably from those in which the dose-meter system was calibrated.

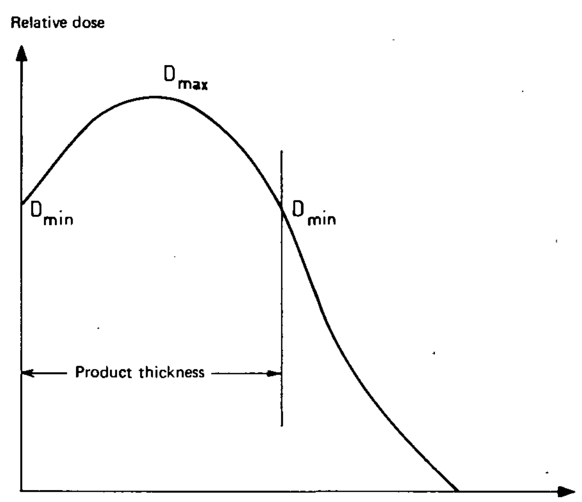

Figure 3. A representation of the depth-dose distribution in a product following one-sided electron irradiation. Note that the electrons have a finite range and that the product thickness is adjusted to ensure that it is less than the electron range.

These differences may lead to unexpected systematic errors in estimating the dose.

In a gamma-ray facility dose-meter systems are usually calibrated at a fixed, standardized dose-rate, at a fairly constant temperature, in less than 4 hours. In a large gamma-ray facility, the dose is accumulated at widely varying rates, over a period of many hours if not days, and in conditions where temperature increases of 20°C are not uncommon. In an electron-beam facility the dose is accumulated at a very high rate in a few seconds.

Many materials undergo measurable changes when exposed to high doses of radiation of the order of several kilograys, and hence could be used for dosimetry. However, many of the radiation-induced changes are inconvenient or difficult to measure, or are not sufficiently stable to permit reproducible estimates of dose to be made. Although the more convenient and more stable changes can be measured, the final products in all high-level dosimetry systems are the results of the interaction of initially induced, chemically reactive, unstable species. The rates at which the initial species are formed and the rates at which the unstable species interact with each other depend strongly on the environmental conditions of the dose meters – temperature, water content, presence of oxygen, etc. Thus, differences between conditions during calibration and routine use can cause systematic deviations in dose-meter response and lead to insidious, unexpected errors in dose-estimation.

One factor which influences the response of most high-level dose-meter systems is the temperature during irradiation: Figure 4 shows how the response of several different dose-meter systems depends on temperature. Another factor is the water-content at the time of irradiation: Figure 5 shows the change in response of a clear perspex dose meter after several months pre-irradiation storage at either 0% or at 80 to 100% relative humidity. Also shown in the figure is the effect of a 48-hour treatment in 80°C water before irradiation. Figure 6 presents the dependence of the response of a blue cellophane dose meter on relative humidity at 50 kGy.

These examples show that mistakes can be made in routine dosimetry during industrial irradiation due to differences between conditions during calibration and operation. Such unsuspected mistakes can be crucial to the operator because they may reflect on the reliability of the irradiated product. More importantly, however, accurate dosimetry can provide a unique quality control of the radiation process and is the basis for the regulatory acceptance of products which might affect public health. Medical products are considered to be sterile if only one unit in a million carries a viable bacterial spore. In practice, it is impossible to test sterilized products microbiologically to see if they meet this requirement. In food preservation it is equally impossible to test the effect of irradiation: by the time the test is made, the food is no longer preserved. Consequently quality control of these radiation processes must be based on the assurance that the product has been given the correct dose and this requires that the in-product dose be accurately measured.

An international intercomparison

With the increasing international trade in irradiated products, the authorities who assess their safety will require some concrete indication that the product has

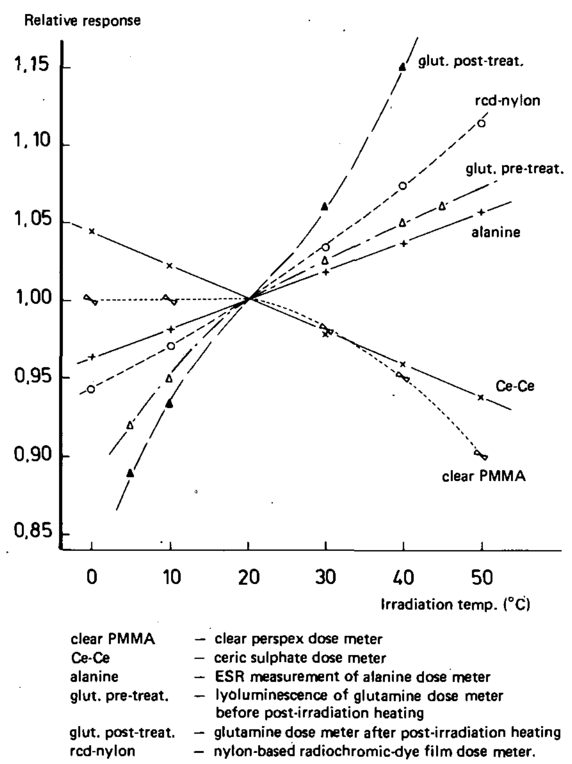

Figure 4. The dependence of the response of several dose-meter systems on the irradiation temperature. (Data derived from references [4, 10, 12 and 13].)

received the proper dose and that the measurements are reliable. With this in mind the IAEA established in 1977 a programme to develop an independent international high-dose intercomparison for industrial radiation processing. The Agency's ultimate goal is to provide an international dose-assurance service.

The Agency's intercomparison programme has developed a series of dose-meter systems which can be used 'in the product' during normal operation of an irradiation facility, in parallel with the operator's own routine dose-meter system. The aim is to confirm that no unexpected errors are arising in the facility's routine dose-meter system.

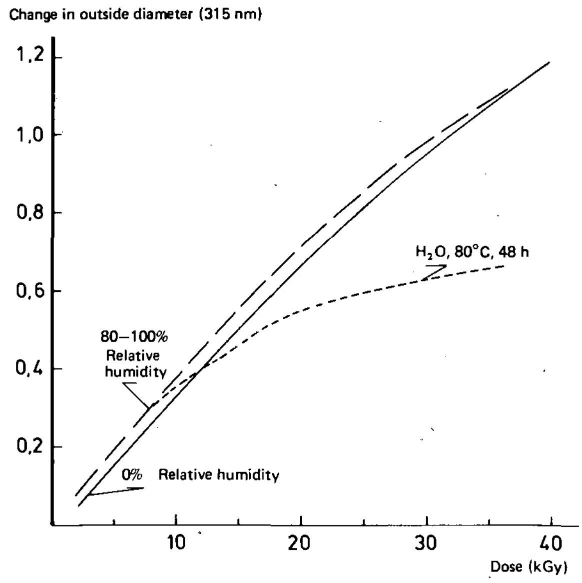

Figure 5. The effect of humidity treatment on the response of a clear polymethylmethacrylate (perspex) dose meter. Samples were stored for several months at the different humidities to equilibrate.

It should be stressed that, although a primary standard for dosimetry at kGy doses does not exist, this programme is not intended to provide such a standard, rather it is hoped that one will eventually become available under the auspices of the Bureau International des Poids et Mesures. The Agency's intercomparison dose-meter systems would then be

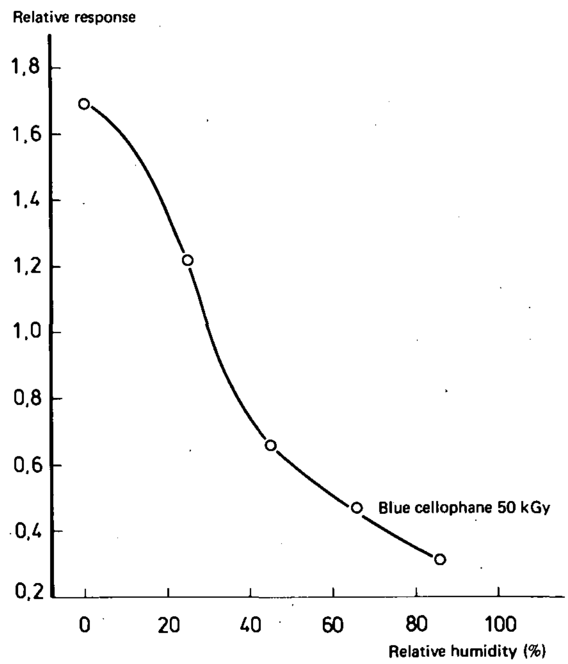

Figure 6. The relative response of the blue cellophane dose meter as a function of relative humidity. The dose level is 50 kGy, normalization is at 32% relative humidity (r.h.). (Data derived from reference [14].)

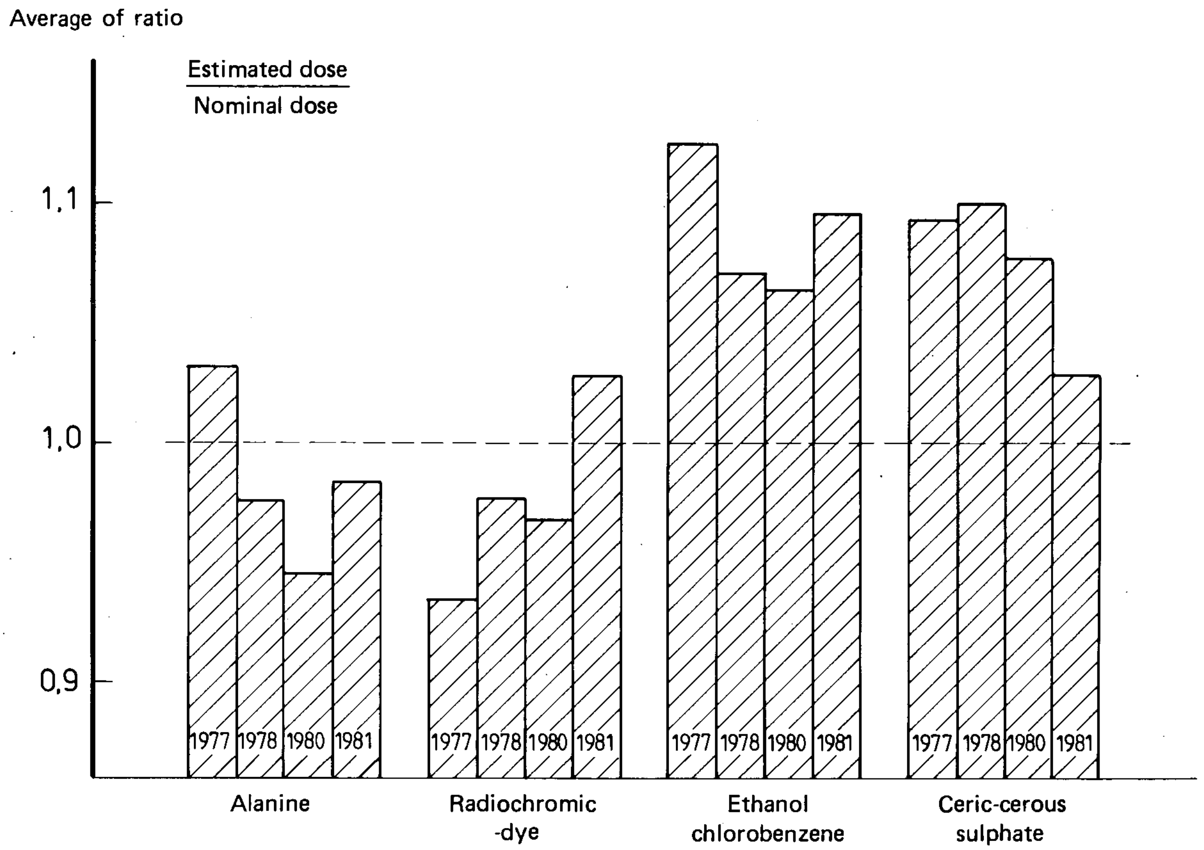

Figure 7. The average of the ratio of estimated dose to nominal dose determined in the four medium- and high-dose intercomparison studies by four dose-meter systems used. (1977 data exclude cross-measurements between dose-meter operating laboratories.)

calibrated against this standard. The programme will not provide a dose-meter calibration service to facility operators nor does it intend to develop one or two systems for universal routine use by radiation facility operators.

Selecting a suitable dose meter

At the start of the intercomparison it was decided that efforts would be concentrated on the development of dose-meter systems for use in gamma- and X-ray facilities; work on electron-beam systems has only started this year.

Because the dose meters are sent and returned by post from all over the world, the systems have to have a good pre-irradiation stability and a very stable dose-response: this implies that several very satisfactory routine dose-meter systems are unsuitable for the intercomparison. So far four suitable systems have been tested – ESR measurements in alanine [4]; radiochromic-dye films [5]; ceric-cerous sulphate solutions [6]; and ethanol chlorobenzene [7]. Initially a preliminary intercomparison (1977) and a comprehensive intercomparison (1978) [8] were carried out at laboratory level; and in 1980 a pilot study was undertaken at the in-plant/in-product level. All these studies were especially designed to take a wide variety of climates into account. A fifth system, lyoluminscence of glutamine [9, 10], was included in a later research study because it has recently been developed for practical use and because it performed very well in the range 0.01 to 3 kGy (1 to 300 krad).

The research study was set up in 1981 to solve a persistent discrepancy between the results from the alanine and radiochromic-dye film dose meters and the ceric-cerous sulphate and ethanol chlorobenzene dose meters. In the intercomparisons and the pilot study the four dose-meter systems proved to be reliable and, with some reservations, all four remained contenders for use in the intercomparison. However, when comparisons were made with the nominal dose quoted by the irradiating laboratories or facilities, the alanine and radiochromic-dye film dose-estimates were consistently 3 to 5% below the nominal, and the ceric-cerous sulphate and ethanol chlorobenzene dose-estimates were 6 to 10% above the nominal (Fig.7).

In the 1981 research study four factors considered most likely to influence dose-meter response were varied. The factors were dose, dose-rate, irradiation temperature, and storage temperature. These were not disclosed to the dose-meter laboratories who were asked to make an initial dose-estimate assuming a set of base conditions. Details about the factors were then provided one-by-one to the laboratories, who then provided sequential corrections to their first dose-estimates. In this way it was hoped that the cause of the discrepancy in dose-estimates would be revealed as a consequence of one or more of the correction factors. Unfortunately this did not happen. The research study did, however, reveal a long-term temperature-humidity instability in the radiochromic-dye films, previously suspected on the basis of one or two results in the intercomparisons. It also revealed that more research was necessary into the high-dose stability of the glutamine-lyoluminescence system and it showed that the alanine and ceric-cerous sulphate systems behaved consistently and in agreement with each other, whilst the ethanol chlorobenzene system consistently over-estimated doses by 10%.

In the preliminary intercomparison, three dose-meter systems performed reliably in the range 0.01 to 3 kGy (1 to 300 krad), but three other systems had serious drawbacks. As a consequence of these results no comprehensive intercomparison was made for the three reliable systems (ESR measurement of alanine, thick radiochromic-dye films, and lyoluminescence of glutamine) and these systems have been used in an in-plant/in-product pilot study during 1981. In this pilot study the alanine system again behaved consistently and the glutamine system behaved well – although its performance at one plant was not entirely consistent. Long-term temperature-humidity instability was again indicated for thick radiochromic-dye films.

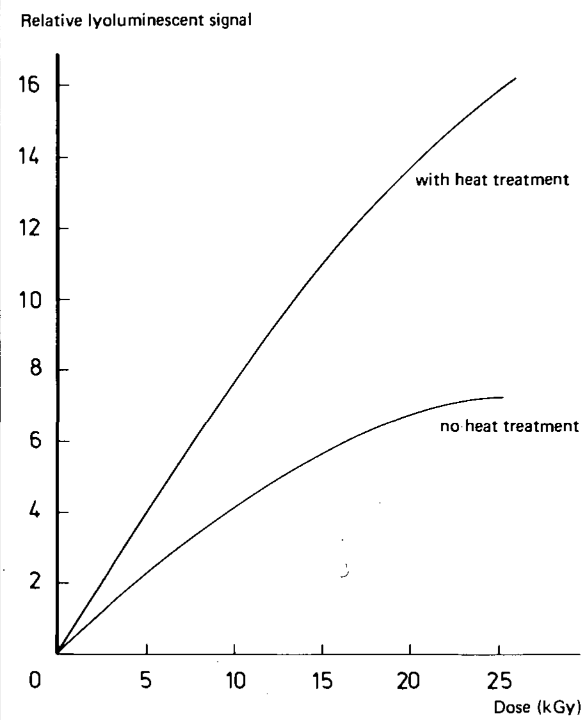

Figure 8. The effect of a post-irradiation heat treatment of 115°C for 5 hours on the dose-response relationship for the lyoluminescence of glutamine. (Figure from reference [10].)

Co-ordinated research programme

In parallel with the intercomparison programme, the Agency established a co-ordinated research programme which concentrated on specific problems:

- The effect of environmental variables, temperature, humidity etc. on the stability and accuracy of high-level dose-meter systems;
- The development of newer dose-meter systems for use in intercomparison services;
- The development of dose-meter systems for the measurement of intense electron beams.

The research programme's main aim is to consolidate the information-base for intercomparison dose-meter systems. Several unexpected results have been detected and the development of the lyoluminescence dose-meter system has been positively stimulated by the programme. The results will not only influence the intercomparison dose-meter systems but will lead to improvements in the use of routine dose meters.

One notable result of the co-ordinated research programme has been the development of a post-irradiation heat-treatment of glutamine dose meters before read-out. This reduces the effect of post-irradiation storage temperature on the response of the dose meter [10]. It had been noted that storage of the glutamine for extended periods of time after irradiation did not cause the lyoluminescent signal to fade, but the signal did depend on temperature and humidity during storage. Treatment at 115°C for five hours increases the response of the dose meter substantially and at the same time almost eliminates any effect of varying post-irradiation storage conditions. This is shown in Figure 8 which presents the relative increase in lyoluminescence signal as a result of the heat treatment. Unfortunately the heat treatment, whilst increasing the dose-meter response also increases the sensitivity of the response to irradiation temperature as can be seen in Figure 4.

The programme also showed that, although the radiochromic-dye films have an exceptionally stable response at the very high dose-rates found in electron-beam facilities, in the lower dose-rates encountered in gamma-irradiation facilities a dose-rate effect can be detected. However, this occurs only when the film has

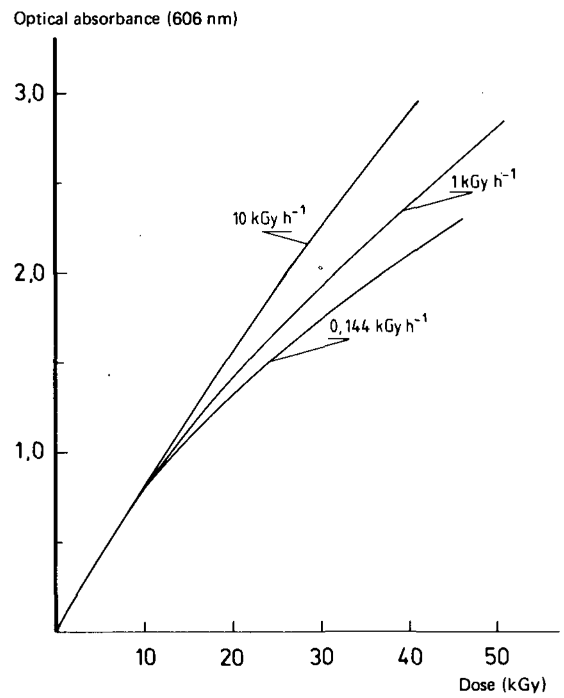

Figure 9. The dose-response of the radiochromic-dye film based on nylon as a function of dose-rate when the film is equilibrated at 32% relative humidity (r.h.). (Figure from reference [11].)

been equilibrated at certain values of relative humidity (Figure 9). Research also indicated that the development of the final colour in this dose-meter film was quite complex. In very dry film a different colour-forming species was found: this was stable until the film was exposed to humid air, when it was converted into the normal species. These investigations into what goes on in a dose meter between irradiation and read-out invariably lead to considerable improvements in eventual performance [11].

Another research study revealed that although the response of the alanine dose meter was not affected by normal exposure to daylight during handling for read-out, a four-week exposure induced a 20% loss of signal; and after being exposed for four weeks to strong UV light, three-quarters of the signal was lost. The interesting feature is that the 75% loss of signal – determined by measuring peak-peak distance on the electron-spin-resonance spectrum of the free radicals – is accompanied by a very noticeable change in spectrum shape. Integration of the ESR spectrum to determine the total number of free radicals revealed that the UV had faded only 30%, so the UV must convert many of the original radicals into a second species. Hence, even in this dose-meter system, which has behaved so consistently in the intercomparison exercises, the effect induced by radiation is not completely stable, and the system needs to be handled with care to obtain the optimum results.

An international dose-assurance service

In this series of intercomparisons it has become clear that while several dose-meter systems are performing well under the extremely demanding conditions, the alanine system behaved consistently and could cover the complete dose-range from 10 Gy to 100 kGy. The Agency has therefore been advised to set up its dose-assurance service for gamma-irradiation facilities in the dose-range 10 Gy to 100 kGy using the measurement of electron-spin-resonance of free radicals induced in alanine as the dose-transfer system. Initially a pilot service will be operated to assess the organizational problems involved in running such a service and to assess the behaviour of the dose-measurement system in practice.

So that the service may achieve a reputation as a completely independent, unbiased international service, the Agency will co-ordinate the distribution and retrieval of the dose meters and collate all the data. Confidentiality will be preserved for the facility operator by using coded dose meters and the operator will be able to compare his estimate of the dose with the Agency's. It has been proposed that an 'investigation limit' should be introduced when a difference of more than 10% is found between the dose estimated by the operator and that by the Agency. A difference of 10% would imply a systematic discrepancy between the two measurement systems, and would require further investigation. In such a case the Agency could repeat the measurements at the facility using a back-up system in tandem with the normal one. If the results confirmed that the facility operator's routine system was not operating properly, the Agency could offer expert help to discover the causes of the dosimetric errors.

Once the service has been set up, the next step must be to develop, via a series of intercomparison exercises, a dose-meter system suitable for electron-beam measurement. This seems likely to be technically more complicated than for the gamma- and X-ray facilities, but the lessons already learned in the programme should help the development of an electron-beam dosimetry intercomparison service.

Eventually, the Agency's international independent Dose-Assurance Service could create the situation when public health authorities or regulating authorities may decide to approve irradiated products, especially those concerning public health, only when they are accompanied by supporting documents indicating that the quality-control dosimetry is assured by the Agency's service. Some people may think that this is unnecessary and implies a mistrust of their own dosimetric capabilities but I do not believe that this is true. I know that many irradiation facility operators are able to do accurate dosimetry, but I also know that it is easy to make substantial systematic errors quite unknowingly. I am convinced that an independent dose-assurance service can only improve confidence and trade in irradiated goods, and thus benefit both the consumer and the radiation-processing industry.

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