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THE SUITABILITY OF INTRINSIC AND ADDED MATERIALS AS DOSE METERS FOR RADIATION PROCESSING OF PARTICULATE FOODS

D. A. E. EHLERMANN
Institute of Process Engineering, Federal Research Centre for Nutrition, Karlsruhe, F.R.G.

Abstract—Radiation dosimetry applied to food irradiation still has some unresolved issues. One—most substances used in radiation dosimetry are foreign to food and toxic in many cases—might be settled by screening food compounds for their dosimetric suitability. Chemiluminescence of some crystalline food compounds is well known; electron spin resonance spectroscopy of alanine—a food compound—is about to become an internationally accepted standard in radiation dosimetry; little is known about thermoluminescence as many food compounds melt or sublimate at the necessary reading temperatures. Preliminary results of a screening study of food compounds for their dosimetric capabilities are reported. Also the application of such dose meter systems to measurements in free-flowing particulate foods is described.

INTRODUCTION

Correct application of dosimetry to radiation processing of food is still an issue under discussion (Bögl *et al.*, 1988). Some studies are concerned with finding a single dose meter system covering the full range of food irradiation from 10 Gy to 10 kGy. Other studies are devoted to the problems of external influences such as low and high temperatures and to the problem of increasing the accuracy with which rather narrow dose ranges can be reliably measured. One specific problem is associated with the use of suspended dose meters in a stream of free-flowing particulate food and bulk particulate solids. In such cases the problem of lost dose meters may be encountered. The inability to retrieve all implemented dose meters after radiation processing implies the release of the dosimetric materials to the food during subsequent processing. If the compounds used for dosimetry are foreign to food and toxic, as indeed some of the usual dose meter substances are, such a loss might taint the whole consignment of food.

Principles of dosimetry for food irradiation and guidelines for dose meter selection are described elsewhere (Chadwick *et al.*, 1977) and concepts for particular measurements can be derived (Chadwick and Oosterheert, 1986). An especially important issue is the traceability to national and to internationally accepted standards. The available international service (Nam, 1985; Nam and Regulla, 1989) may contribute to the solution of the problem. From the application of such more general rules and from practical experience in the utilization of dosimetry for the increasing commercial-scale exploitation of radiation processing the development of a new and better suited dose meter system can be undertaken.

SELECTION OF FOOD COMPOUNDS

Methods of radiation dosimetry are well established (Mahesh and Vij, 1985) and several compounds with dosimetric capabilities are also food components. Several publications describe the application of lyoluminescence (Ettinger and Puite, 1982; Ettinger *et al.*, 1978; Puite and Ettinger, 1982); less specific information is available on thermoluminescence where usually specially designed or doped materials are employed in order to achieve high sensitivity and reproducibility. Dust and sand are concomitant substances in many types of food as a result of field and harvesting procedures. One compound, quartz, is suitable for dosimetric purposes. Furthermore, fine quartz sand of known origin and well determined dosimetric properties may be employed for dosimetry. One food compound, the amino acid alanine, contained in proteins, is now emerging as an internationally accepted standard for dosimetry using electron spin resonance (ESR) spectroscopy (Regulla, 1990); this dose meter system is also the basis for the International Dose Assurance Service (Nam and Regulla, 1989) and under study by several national metrological laboratories. An ideal case would be the use of homogeneous foods or foods containing homogeneous parts, where that component exhibits dosimetric capabilities. Whole pepper, white or black, where the grains serve as dose meters, might be suitable (Ehlermann, 1988).

CALIBRATION OF SOME FOOD DOSE METERS

The following fine powders were used as dose meter materials: saccharose and table salt—household quality; the samples were homogenized to increase reproducibility. Fine-grain quartz (Merck) was

obtained washed and glowed. L-Glutamine (Merck) and L-alanine (Serva) was of biochemical grade. Commercially available lithium fluoride and borate powders (Harshaw TL-100 and TL-800) were included in the study. These materials are foreign to and undesirable in food. However, their suitability for dosimetry and for such studies on free-flowing particulate foods is well established. During an initial run, also dyed nylon films (Far West) were employed. These usually serve as routine dose/monitors at our facilities. Finally, in a case study on grain irradiation only glutamine and quartz were used and LiF served for comparison purposes.

As ESR measurements were not feasible, chemi- and thermoluminescence were used as a read-out method. Chemiluminescence was determined using an LKB 1251 luminometer and integration period of 10 s; thermoluminescence was measured using an ELSEC 7185 Thermoluminescent Dating System and the peak height at 195 and 200°C for LiF and quartz, respectively. For chemiluminescence sufficient solubility of the crystals of food compounds is necessary in order to avoid excessively long integration periods for the light output. With thermoluminescence the melting and sublimation points should be well above the temperature at which light emission of the crystals is stimulated. Both methods are well established at our laboratories and used also for studies on the identification of irradiated foods (Delincée, 1989). For alanine instead of ESR spectroscopy an alternative read-out procedure is available using indirect oxidation of ferrous ions (van Laere *et al.*, 1989).

All samples were irradiated with 10 MeV electrons at an average dose rate of 10^6 Gy/s and the radiation field was calibrated by Fricke dosimetry. In order to compare readings from different instruments and to allow for the wide range of sensitivity, the results are presented in arbitrary units. The thermo- and chemiluminescence responses of several materials up to 1.5 kGy are shown in Fig. 1. Saccharose shows early saturation, whereas glutamine, quartz and lithium borate are useful also in the range up to 10 kGy. As radiation processing of grain in bulk aims at the elimination of insect infestation our interest centred around the applicable dose of about 200 Gy. These calibrations are shown in Fig. 2. Ordinary table salt reveals a non-linear response curve. Quartz and glutamine have a linear response in the dose region of interest and alanine and lithium fluoride are perfectly linear. The response curve of quartz for the full dose ranging up to 10 kGy is non-monotonous and approaches saturation; however, it can be used for dosimetry. Quartz powder may be a suitable dose meter material to be used with particulate food radiation processing. Its dosimetric properties, such as dose rate and temperature dependence, as well as fading, shall be established in further studies. Every point in Figs 1 and 2 respresents 5–10 individual measurements and the respective standard deviations approximate to the size of the symbols used.

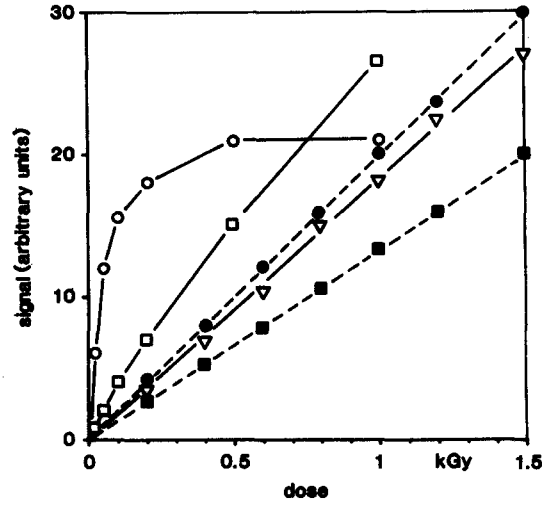

Fig. 1. Calibration curves for several dose-meter materials (10 MeV electrons; dose rate 10^6 Gy/s). Chemiluminescence: (○) saccharose; (□) table salt; (▽) glutamine; thermoluminescence: (●) quartz; (■) $Li_2B_4O_7$.

APPLICATION TO BULK PROCESSING

A pilot plant at the site of the electron linear accelerator was available (Ehlermann *et al.*, 1988; Schubert and Ehlermann, 1988). It consists of feeding and receiving silos, a pneumatic transport system and a vibrating trough conveyor in the irradiation zone. The flow pattern of particulate foods such as whole wheat was carefully established. Likewise the depth–dose distribution in the filled trough was determined during stand-still. It had to be expected that a mathematical folding of velocity distributions and depth–dose curves would render the overall dose distribution as measured by dose meters suspended in the stream of the wheat. For this purpose dose meter powder was filled into small gelatine capsules (5 mm dia., 13 mm long) and the average density was adjusted to the density of the grain. The iron weights used for this purpose also enable magnetic sorting and retrieving of the capsules. The

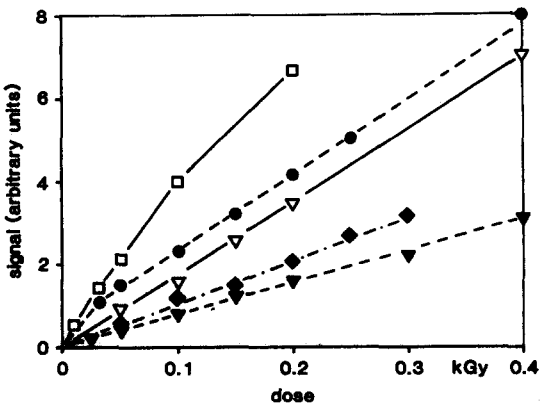

Fig. 2. Calibration curves for several dose-meter materials (10 MeV electrons; dose rate 10^6 Gy/s). Chemiluminescence: (□) table salt; (▽) glutamine; thermoluminescence: (●) quartz; (▼) LiF; optical absorbance: (◆) alanine.

path of individual capsules through the irradiation zone was carefully established. When the dose meters were mixed strategically and at random into the grain in the feeding silo the capsules appeared evenly distributed over the cross-section of the irradiation zone. It was also established that the capsules did not drift to the surface or the bottom of the conveyor volume. The feeding inlet of the vibrating trough was adjusted in such a way that a filling height of 45 mm was always maintained. For the bulk density of wheat of 0.8 g/cm^3 this corresponds to an equivalent water layer of 37 mm or to a depth at which the dose shall be about 50% of the surface dose. The maximum dose was determined to be 120% of the surface dose. Consequently, the ratio of the maximum to the minimum dose (uniformity ratio) should be 2.4.

In a preliminary experiment, using dyed nylon films, the target dose was 1 kGy according to the sensitivity of the films. Figure 3 shows the resulting frequency distribution. The uniformity ratio is slightly above 2.0 and in good agreement with the expected value. Consequently, the experiment was repeated using LiF as a reference material for dosimetry powders, glutamine as a food compound, and quartz as an inert material concomitant to grain. All dose meter capsules were added simultaneously during one single run of pilot processing of grain. For the purpose of easier comparison Gaussian fits to the measurements instead of the group values are presented (Fig. 4). With lithium fluoride the measurements were extraordinarily reproducible and the frequency distribution corresponded to the one given in Fig. 3. The radiation treatment had been adjusted for a minimum dose of 200 Gy which was easily achieved. When glutamine and quartz were used as dose meter materials the resulting frequency distributions widened. This can be partially attributed to the larger variabilities of these dose meter systems. A slight shift in the mean value for glutamine was observed. However, the difference was not statistically significant. The experiments will be continued in order to clarify the observed discrepancies as well as to study which intrinsic material is best suited for dosimetry, in stream, of bulk particulate foods. Furthermore, measures to reduce the uniformity ratio by changing and adapting the velocity distributions of the grains in the radiation processing area are under study.

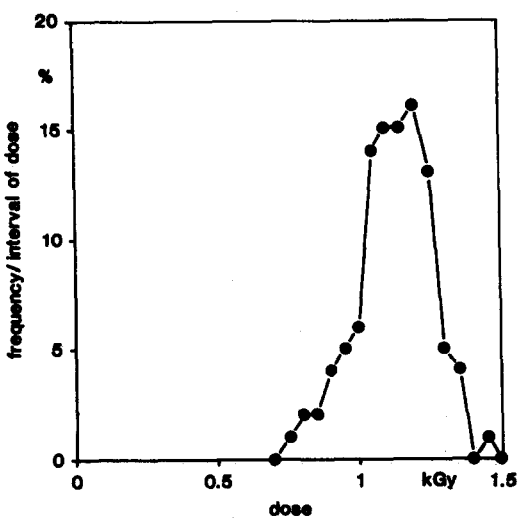

Fig. 3. Frequency distribution of dose (class width 50 Gy) for grains irradiated by 10 MeV electrons in a vibrating trough conveyor (layer height 45 mm; total 100 dyed nylon films).

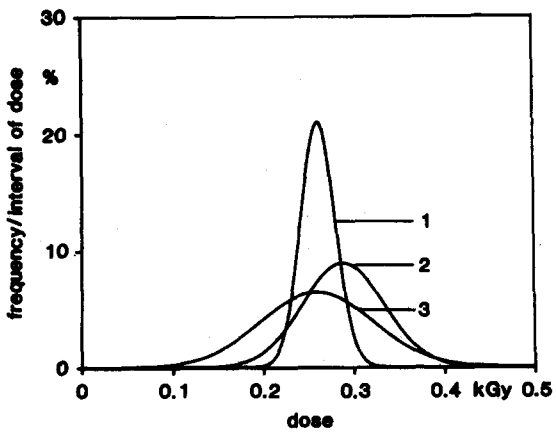

Fig. 4. Comparison of frequency distributions of dose for grains (conditions as in Fig. 3; curves are Gaussian fits) measured by several dose meter types (in parentheses number of capsules suspended): 1 = LiF (40); 2 = glutamine (60); 3 = quartz (60).

CONCLUSIONS

Food compounds or concomitant substances inherent in grain may be used as dosimetric materials which are neither foreign to food nor toxic. Some of the materials have good dosimetric properties, e.g. glutamine and quartz which were used in the reported study. If filled in capsules and compensated for bulk density of particulate foods, such capsules may be added strategically and at random to the bulk stream through the irradiation zone. The resulting frequency distributions for the dose are in good agreement with the expected values calculated from measurements in the motionless state.

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