

## 5 Irradiation

*Alistair S. Grandison*

### 5.1 Introduction

Irradiation has probably been the subject of more controversy and adverse publicity prior to its implementation than any other method of food preservation. In many countries, including the UK, irradiation has been viewed with suspicion by the public, largely as a result of adverse and frequently misinformed reporting by the media. In the UK this has resulted in the paradoxical situation where irradiation of many foods is permitted, but not actually carried out. On the other hand, many nonfood items such as pharmaceuticals, cosmetics, medical products and plastics are routinely irradiated. Yet, the process provides a means of improving the quality and safety of certain foods, while causing minimal chemical damage.

Irradiation of food is not a new idea. Since the discovery of X-rays in the late 19th century, the possibility of controlling bacterial populations by radiation has been understood. Intensive research on food irradiation has been carried out for over 50 years, and the safety and 'wholesomeness' of irradiated food have been established to the satisfaction of most scientists.

### 5.2 Principles of Irradiation

Irradiation literally means exposure to radiation. In practice three types of radiation may be used for food preservation: Gamma ( $\gamma$ ) rays, X-rays or high-energy electron beams ( $\beta$  particles). These are termed ionising radiations. Although the equipment and properties differ, the three radiation types are all capable of producing ionisation and excitation of the atoms in the target material, but their energy is limited so that they do not interact with the nuclei to induce radioactivity. Gamma rays and X-rays are part of the electromagnetic spectrum, and are identical in their physical properties, although they differ in origin.

The energy of constituent particles or photons of ionising radiations is expressed in electron volts (eV), or more conveniently in MeV ( $1 \text{ MeV} = 1.602 \times 10^{-13} \text{ J}$ ). One eV is equal to the kinetic energy gained by an electron on being accelerated through a potential difference of 1 V. An important and sometimes confusing distinction exists between radiation energy and dose. When ionising radiations penetrate a food, energy is absorbed. This is the 'absorbed dose' and is expressed in Grays (Gy), where 1 Gy is equal to an absorbed energy of  $1 \text{ J kg}^{-1}$ . Thus, while radiation energy is a fixed property for a particular radiation type, the absorbed dose varies in relation to the intensity of radiations, exposure time and composition of the food.

Gamma rays are produced from radioisotopes and hence they have fixed energies. In practice Co 60 is the major isotope source. This isotope is specifically manufactured for irradiation and is not a nuclear waste product. An alternative radioisotope is Cs 137, which is a byproduct of nuclear fuel reprocessing, and is used very much less widely than Co 60.

Electron beams and X-rays are machine sources, which are powered by electricity. Hence they exhibit a continuous spectrum of energies depending on the type and conditions of the machinery. They hold a major advantage over isotopes in that they can be switched on and off and can in no way be linked to the nuclear industry.

The mode of action of ionising radiation can be considered in three phases:

- the primary physical action of radiation on atoms;
- the chemical consequences of these physical actions;
- the biological consequences to living cells in food or contaminating organisms.

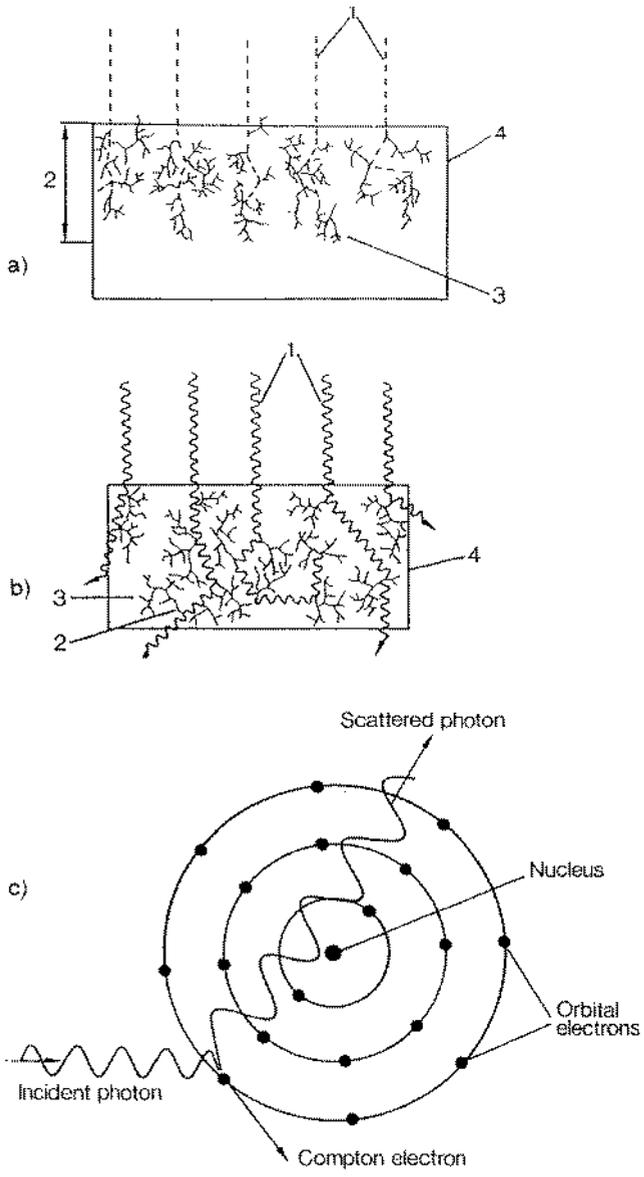
### 5.2.1

#### Physical Effects

Although the energies of the three radiation types are comparable, and the results of ionisation are the same, there are differences in their mode of action as shown in Fig. 5.1.

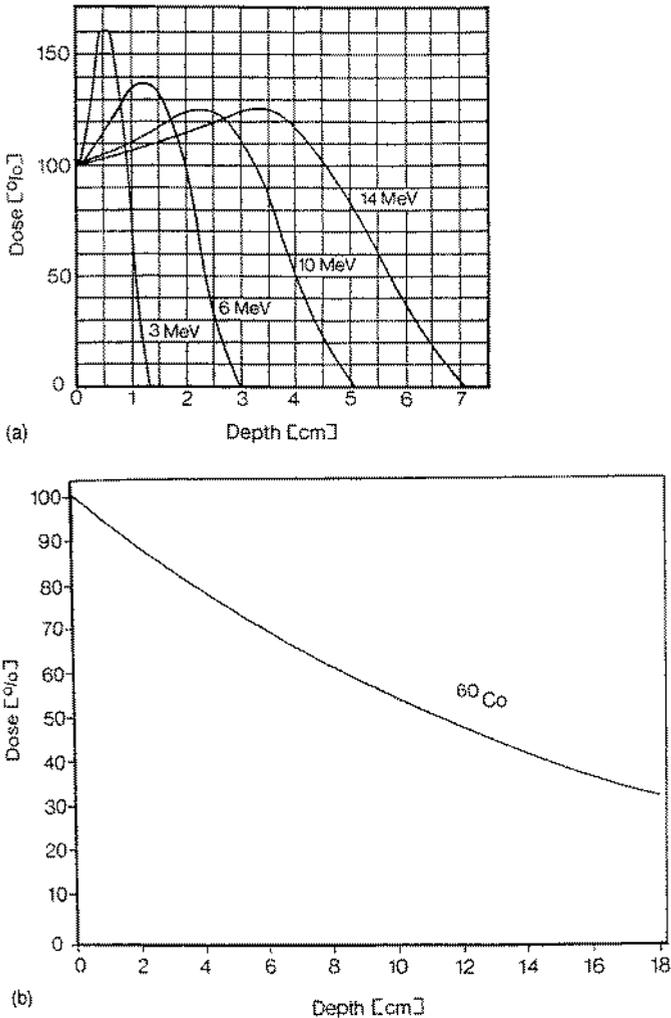
High-energy electrons interact with the orbital electrons of the medium, giving up their energy. The orbital electrons are either ejected from the atom entirely, resulting in ionisation, or moved to an orbital of higher energy, resulting in excitation. Ejected (secondary) electrons of sufficient energy can go on to produce further ionisations and excitations in surrounding atoms (see Fig. 5.1 a).

X-rays and  $\gamma$  rays can be considered to be photons; and the most important interaction is by the Compton effect (see Fig. 5.1 b, c). The incident photons eject electrons from atoms in the target material, giving up some of their energy and changing direction. A single photon can give rise to many Compton effects and can penetrate deeply into the target material. Ejected electrons which have sufficient energy (approx. 100 eV) go on in turn to cause many further excitations and ionisations. Such electrons are known as delta rays. It has been calculated that a single Compton effect can result in 30–40 000 ionisations and 45–80 000 excitations [1].



**Fig. 5.1** Interaction of radiation with matter, adapted from Diehl [3]. (a) Electron radiation: 1, primary electron beam; 2, depth of penetration; 3, secondary electrons; 4, irradiated medium. (b) Gamma or X-radiation: 1,  $\gamma$  or X-ray photons; 2, Compton electrons; 3, secondary electrons; 4, irradiated medium. (c) Compton effect.

The radiation-induced chemical changes produced by  $\gamma$  or X-ray photons or by electron beams are exactly the same, because ionisations and excitations are ultimately produced by high-energy electrons in both cases. However, an important difference between photons and high-energy electrons is the depth of penetration into the food. Electrons give up their energy within a few centimetres of the food surface, depending on their energy, whereas  $\gamma$  or X-rays penetrate much more deeply, as illustrated in Fig. 5.1 a, b. Depth-dose distribution curves for electrons and  $\gamma$  rays are shown in Fig. 5.2 for irradiation of water, which acts as a reasonable model for foods.



**Fig. 5.2** Depth-dose distribution in water from one side: (a) electrons of different energy, (b)  $\gamma$  radiation; adapted from Diehl [3].

The difference in the shape of the curve between the two radiation types reflects the difference in their mechanism of action. With electrons, it can be seen that the effective dose is greatest at some distance into the food, as more secondary electrons are produced at that distance. The primary electrons lose their energy by interacting with water, so that the practical limit for electron irradiation is about 4 cm using the maximum permitted energy (10 MeV). Gamma rays, however, become depleted continuously as they penetrate the target and the effective depth is much greater. X-rays follow a similar pattern to Co 60 radiations. Two-sided irradiation permits the treatment of thicker packages of food (see Fig. 5.3) but it is still quite limited for electrons (approx. 8 cm).

The net result of these primary physical effects is a deposition of energy within the material, giving rise to excited molecules and ions. These effects are non-specific, with no preference for any particular atoms or molecules. Hall et al. [2] estimated that the timescale of these primary effects is  $10^{-14}$  s. It is an essential feature that radiations do not have sufficient energy to interact with the nuclei, dislodging protons or neutrons, otherwise radioactivity could be induced in the

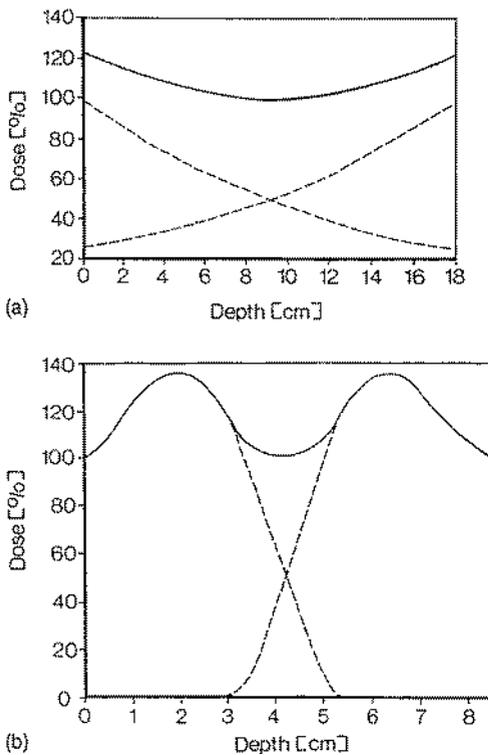


Fig. 5.3 Depth-dose distribution in water from two sides: (a)  $\gamma$  radiation, (b) 10 MeV electrons. Dashed lines indicate dose distribution for one-sided irradiation; adapted from Diehl [3].

target atoms. For this reason, energies are limited to 5 MeV and 10 MeV for X-rays and electrons respectively. Cobalt 60 produces  $\gamma$  rays of fixed energy well below that required to interfere with the nucleus.

### 5.2.2

#### Chemical Effects

The secondary chemical effects of irradiation result from breakdown of the excited molecules and ions and their reaction with neighbouring molecules, giving a cascade of reactions. The overall process by which these reactions produce stable end-products is known as 'radiolysis'. These reactions are considered in detail elsewhere [3] and only a brief summary is given here. The ions and excited molecules contain abnormal amounts of energy, which is lost through a combination of physical processes (fluorescence, conversion to heat, or transfer to neighbouring molecules) and chemical reactions. This occurs irrespective of whether they are free or molecular components. The primary reactions include isomerisation and dissociation within molecules and reactions with neighbouring species. The new products formed include free radicals, i.e. atoms or molecules with one or more unpaired electrons, which are available to form a chemical bond and are thus highly reactive. Because most foods contain substantial quantities of water and contaminating organisms contain water in their cell structure, the radiolytic products of water are particularly important. These include hydrogen, hydroxy radicals ( $\text{H}^\bullet$ ,  $\bullet\text{OH}$ ),  $e_{\text{aq}}^-$ ,  $\text{H}_2$ ,  $\text{H}_2\text{O}_2$  and  $\text{H}_3\text{O}^+$ . These species are highly chemically reactive and react with many substances, although not water molecules. Hall et al. [2] estimated the timescale of the secondary effects to be within  $10^{-2}$  s. In most foods, free radicals have a short lifetime ( $<10^{-3}$  s). However, in dried or frozen foods, or foods containing hard components such as bones or shells, free radicals may persist for longer.

The major components of foods and contaminating organisms, such as proteins, carbohydrates, fats and nucleic acids, as well as minor components such as vitamins, are all chemically altered to some extent following irradiation. This can be through direct effects of the incident electrons or Compton electrons. In aqueous solutions, reactions occur through secondary effects by interaction with the radiolytic products of water. The relative importance of primary and secondary effects depends on the concentration of the component in question. The chemical changes are important in terms of their effects on living food contaminants whose elimination is a major objective of food irradiation. However, it is also essential to consider effects on the components of foods inasmuch as this may affect their quality, e.g. nutritional status, texture, off flavours. A further consideration is their effects on living foods such as fruits and vegetables, where the goal is to delay ripening or senescence.

## 5.2.3

**Biological Effects**

The major purpose of irradiating food is to cause changes in living cells. These can either be contaminating organisms such as bacteria or insects, or cells of living foods such as raw fruits and vegetables. Ionising radiation is lethal to all forms of life, the lethal dose being inversely related to the size and complexity of the organism (see Table 5.1). The exact mechanism of action on cells is not fully understood, but the chemical changes described above are known to alter cell membrane structure, reduce enzyme activity, reduce nucleic acid synthesis, affect energy metabolism through phosphorylation and produce compositional changes in cellular DNA. The latter is believed to be far and away the most important component of activity but membrane effects may play an additional role.

The DNA damage may be caused by *direct effects* whereby ionisations and excitations occur in the nucleic acid molecules themselves. Alternatively, the radiations may produce free radicals from other molecules, especially water, which diffuse towards and cause damage to the DNA through *indirect effects*. Direct effects predominate under dry conditions, such as when dry spores are irradiated, while indirect effects are more important under wetter conditions, such as within the cell structures of fruits or vegetative bacterial cells.

It should be noted that remarkably little chemical damage to the system is required to cause cell lethality, because DNA molecules are enormous compared to other molecules in the cell and thus present a large target. Yet, only a very small amount of damage to the molecule is required to render the cell irreparably damaged. This is clearly illustrated in two separate studies reported by Diehl [3], which calculated that during irradiation of cells:

- A radiation dose of 0.1 kGy would damage 2.8% of DNA molecules, which would be lethal to most cells, while enzyme activity would only fall by 0.14%, which is barely detectable, and only 0.005% of amino acid molecules would be affected, which is completely undetectable [5].
- A dose of 10.0 kGy would affect 0.0072% of water molecules and 0.072% of glucose molecules, but a single DNA molecule ( $10^9$  Da) would be damaged at about 4000 locations, including 70 doublestrand breaks [6].

**Table 5.1** Approximate lethal doses of radiation for different organisms; data from [4].

Organism	Lethal dose (kGy)
Mammals	0.005–0.01
Insects	0.01–1.0
Vegetative bacteria	0.5–10.0
Sporulating bacteria	10–50
Viruses	10–200

This is fortuitous for food irradiation, as the low doses required for efficacy of the process result in only minimal chemical damage to the food.

The phenomenon also explains the variation in lethal doses between organisms (see Table 5.1). The target presented by the DNA in mammalian cells is considerably larger than that of insect cells, which in turn are much larger than the genome of bacterial cells; and, consequently, radiation sensitivity follows the same pattern. In contrast, viruses have a much smaller nucleic acid content; and the high doses required for their elimination make irradiation an unlikely treatment procedure. Fortunately, it is unusual for foodborne viruses to cause health problems in humans.

### 5.3

#### Equipment

##### 5.3.1

#### Isotope Sources

Co 60 is the major isotope source for commercial irradiation. It is manufactured in specific reactors and over 80% of the world supply is produced in Canada. Co 60 is produced from nonradioactive Co 59, which is compressed into small pellets and fitted into stainless steel tubes or rods a little larger than pencils. These are bombarded with neutrons in a nuclear reactor over a period of about 1 year to produce highly purified Co 60, which decays in a controlled manner to stable Ni 60, with the emission of  $\gamma$  rays with energies of 1.17 MeV and 1.33 MeV and a half-life of about 5.2 years. Co 60 is water-insoluble and thus presents minimal risk for environmental contamination.

Cs 137 is an alternative possibility, but is much less widely used than Co 60. It decays to stable Ba 137, emitting a  $\gamma$  photon (0.662 MeV) with a half-life of 30 years. It is unlikely to gain more importance and future discussion of radioisotopes will be confined to Co 60.

In practice, a radioactive 'source' is comprised of a number of Co 60 tubes arranged into the appropriate geometric pattern (e.g. plaques or cylinders). The short half-life of Co 60 means that the source will be depleted by approximately 1% per month. This depletion must be considered in calculations of dose and requires that tubes be replaced periodically. In practice, individual rods are replaced in rotation at intervals maintaining the total source energy at a fairly constant level. An individual rod may be utilised for three or four half-lives when activity has decayed to about 10% of its original level. When not in use, the source is normally held under a sufficient depth of water to completely absorb radiation to the surface so that personnel may safely enter the radiation cell area to load and unload radionuclides.

Examples of irradiation plants are shown in Figs. 5.4 and 5.5. Designs can be batch or continuous, the latter being more appropriate for large-scale processing.

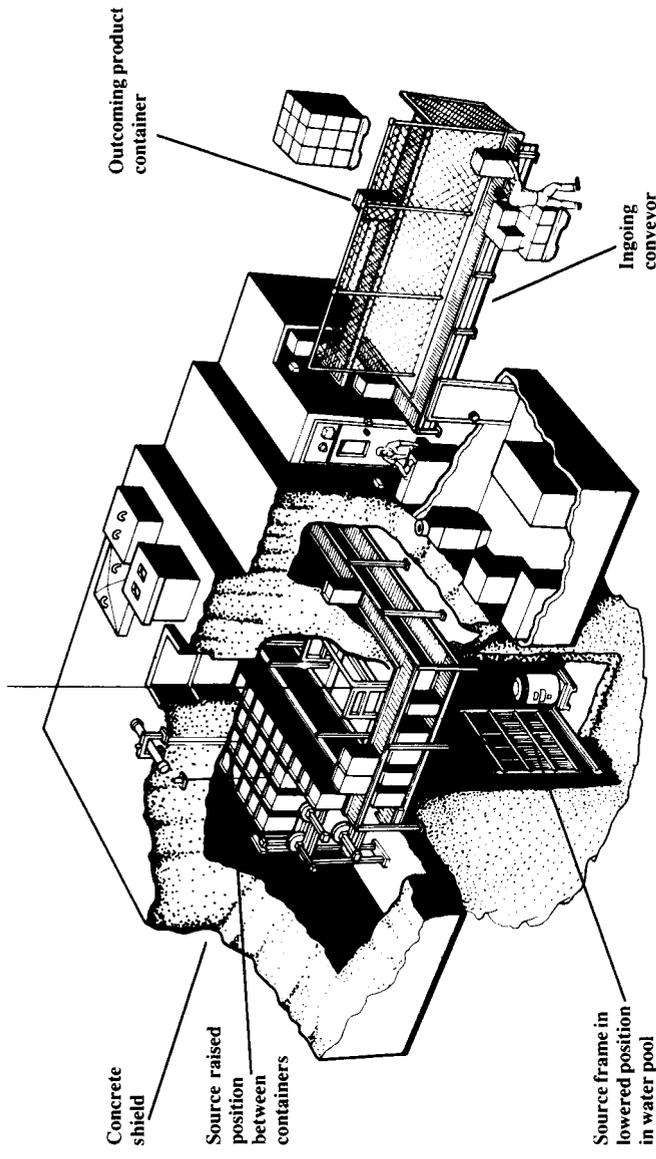


Fig. 5.4 Commercial automatic tote box  $\gamma$  ( $^{60}\text{Co}$ ) irradiator; by courtesy of Atomic Energy of Canada Ltd.

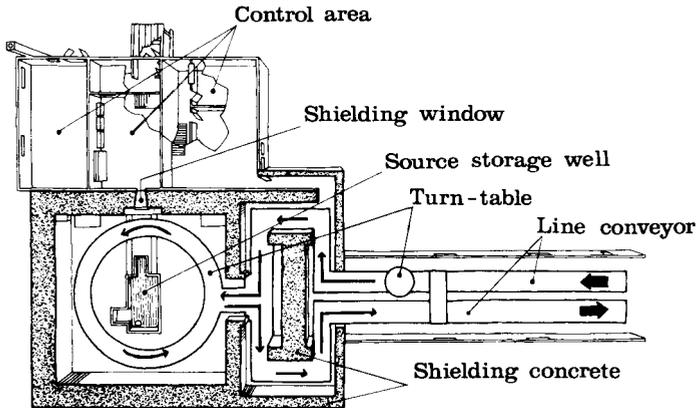


Fig. 5.5 Plan of commercial potato irradiator, by courtesy of Kawasaki Heavy Industries.

As the source emits radiation in all directions and the rate of emission cannot be controlled, it is essential to control product movement past the source in the most efficient way possible, to make best use of the radiations. Another aim is to achieve the lowest possible dose uniformity ratio (see Section 5.3.3). For this reason, containerised products may follow a complex path around the source, often two or more product units in depth, the units being turned to effect two-sided irradiation. An example of product progress around a source is shown in Fig. 5.6.

The dose rates provided by isotope sources are generally low, so that irradiation may take around 1 h to complete. Therefore, product movement is often sequential with a finite time allowed in each position without movement, to allow absorption of sufficient radiation.

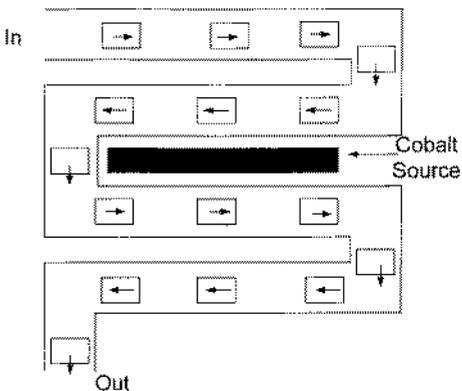


Fig. 5.6 Example of progress of containerised product around a  $\gamma$  irradiation source.

## 5.3.2

**Machine Sources**

Both electron and X-ray machines use electrons, which are accelerated to speeds approaching the speed of light by the application of energy from electric fields in an evacuated tube. The resulting electron beams possess a considerable amount of kinetic energy.

The main designs of electron irradiator available are the Dynamitron, which will produce electron energies up to 4.5 MeV, or linear accelerators for higher energies. In either case, the resulting beam diameter is only a few millimetres or centimetres. To allow an even dose distribution in the product, it is necessary to scan the beam using a scanning magnet, which creates an alternating mag-

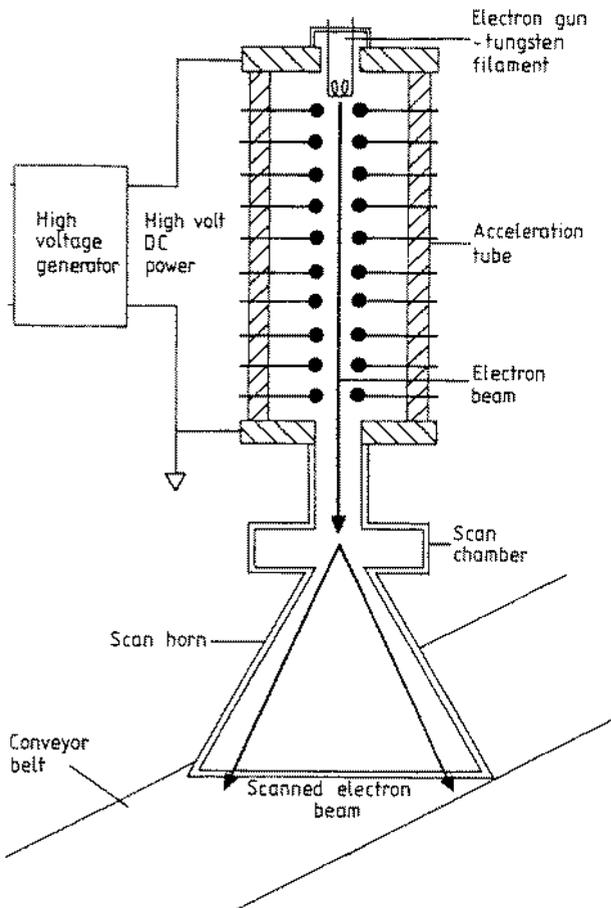


Fig. 5.7 Simplified construction of electron beam machine; by courtesy of Leatherhead Food RA, UK.

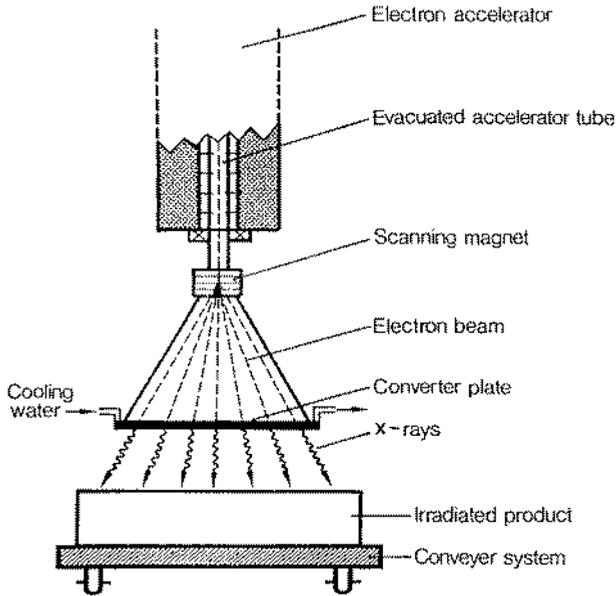


Fig. 5.8 Electron accelerator with X-ray converter; from Diehl [3], with permission.

netic field (analogous to the horizontal scan of a television tube) which moves the beam back and forwards at 100–200 Hz. An even, fan-shaped field of emitted electrons is created, through which the food is conveyed. A simplified diagram of an electron beam machine is shown in Fig. 5.7.

As described in Section 5.2.1, the low penetration of electrons requires that this technology is limited either to surface treatment of food or to foods of limited thickness (8 cm max., with two-sided irradiation).

When electrons strike a target, they produce X-rays which can be utilised to give greater penetration depth, but which suffer from the disadvantage of a low conversion efficiency. The efficiency of conversion depends on the energy of the electrons and the atomic number of the target material. In practice, therefore, X-rays are produced by firing high-energy electrons at a heavy metal target plate, as shown schematically in Fig. 5.8. Even with 10 MeV electrons and a tungsten (atomic number 74) plate, the efficiency of conversion is only 32%, hence cooling water must be applied to the converter plate.

Both electrons and X-rays deliver much higher dose rates than isotope sources, so that processing is complete in a matter of seconds. Also the beams may be directed, so that the complex transport of packages around the source is not required.

## 5.3.3

**Control and Dosimetry**

As with any processing technology, radiation treatment must be controlled and validated; and hence the level of treatment must be monitored in a quantifiable form. The subject is considered in detail by Ehlermann [7]. The basic parameter of importance is the absorbed dose, although information on dose rate may be important in some circumstances. A knowledge of absorbed dose is necessary, both to conform to legal limits and to ensure that the advisory technological considerations have been met. In other words, to know whether the food has received sufficient treatment without exceeding either the legal limit or the threshold dose for sensory impairment. The dose absorbed by a food depends on the magnitude of the radiation field, the absorption characteristics of the food and the exposure time.

A further consideration is the dose uniformity ratio, i.e. the ratio between the maximum and minimum dose absorbed by different parts of a food piece or within a food container. Radiation processing results in a range of absorbed doses in the product, in the same way as heating results in a range of temperatures. Dose is normally expressed in terms of average dose absorbed by the food during the treatment. Dose distribution depends partly on the type of radiation used, but is affected by the geometry of individual food units and the way in which the food is packaged and loaded into containers for processing. The ideal uniformity ratio of 1 is not achievable in any type of plant, a value of about 1.5–2.0 being a rough guideline for practical applications.

Validation of dose and quality assurance (as well as optimising plant performance) can be carried out using dosimeters. Routine dosimetry is carried out by attaching dosimeters to packages and then reading on completion of treatment. The dosimeters are usually in the form of plastic strips whose absorption characteristics change in a linear fashion in relation to a given irradiation dose. Radiation-induced changes are measured with a spectrophotometer at the appropriate wavelength. It is essential that routine dosimeters be ultimately related to a primary standard at a specialised national standards laboratory, e.g. the National Physical Laboratory in the UK. The primary standard is the actual energy absorbed by water as determined by calorimetry, i.e. measurement of the temperature rise and hence heat absorbed during irradiation.

Chemical dosimetry systems such as the Fricke system can be used as reference systems to ensure the reliability of routine systems. This system is based on the conversion of ferrous to ferric ions in acidic solutions, measured with a spectrophotometer, which is highly accurate but too complex for routine use.

#### 5.4 Safety Aspects

The safety of consuming irradiated foods is no longer in serious question; and this is discussed in other parts of this chapter. However, the safe operation of irradiation facilities warrants further consideration. In particular, precautions must be taken to protect workers, the public and the environment from accidental exposure. These considerations must encompass both the irradiation facilities and the transport and disposal of the radioactive materials associated with  $\gamma$  plants. There are more than 170  $\gamma$  plants and 600 electron beam facilities in operation worldwide [3], but only a small fraction of these are used for treatment of food. As with any industrial process, there is the potential for accidents, but radiation processing is one of the most strictly regulated and controlled industries, with an excellent safety record [8].

Any design of plant must incorporate a 'cell' to contain the radiation source. Commercial plants usually have walls constructed of concrete, while lead may be employed in smaller plants. Gamma sources are held under water when not in use. Irradiators are designed with overlapping protection to prevent leaking of radiation to workers or the public; and examples are shown in Figs. 5.4 and 5.5. It is of paramount importance that personnel cannot enter the cell when irradiation is taking place, as exposure for even a few seconds could deliver a lethal dose. Precautions against uncontrolled entry, such as interlocks, are therefore an essential feature. Transportation of radioactive source materials is carried out in special casks, which are designed to survive the most severe accidents and disasters. Waste radioactive materials are returned to the manufacturer and either reused or stored until harmless. The quantity of waste is, in fact, very small. The North American Food Irradiation Processing Alliance [9] report that, in 40 years of transporting radioactive materials, there has never been a problem with the materials and, during 35 years of operating irradiation facilities, there has never been a fatality. This is an excellent record compared to other industries, such as those involving shipping toxic materials or crude oil and petroleum products.

It should be remembered that electron and X-ray equipment carries no problem of transporting, storing or disposal of radioactive materials.

#### 5.5 Effects on the Properties of Food

The radiation doses suitable for food systems cause very little physicochemical damage to the actual food, but potential effects on the major components will be considered.

The mineral content of foods is unaffected by irradiation, although the status of minerals could be changed, e.g. extent of binding to other chemicals. This could feasibly affect their bioavailability.

The major effect of radiation on carbohydrates concerns the breakdown of the glycosidic bond in starch, pectin or cellulose to give smaller carbohydrates. This can lead to a reduction in the viscosity of starches, or a loss of texture in some foods, for example leading to the softening of some fruits.

Irradiation of proteins at doses up to 35 kGy causes no discernible reduction in amino acid content [4] and hence no reduction in their protein nutritional quality. Changes in secondary and tertiary structures are also minimal, although there may be isolated examples where functionality is impaired. For example, the whipping quality of egg whites is impaired following irradiation. As stated previously, effects on enzyme action are very limited and are probably undetectable at the doses likely to be used for foods.

Insignificant changes in the physical (viscosity, melting point) and many chemical properties (iodine value, peroxide number etc.) of lipids are produced by irradiation up to 50 kGy [4]. The major concern with fat-containing foods is the acceleration of autoxidation when irradiated in the presence of oxygen. Unsaturated fatty acids are converted to hydroperoxyl radicals, which form unstable hydroperoxides, which break down to a range of mainly carbonyl compounds. Many of the latter have low odour thresholds and lead to rancid off flavours. It should be noted that the same endproducts are found following longterm storage of unirradiated lipids. This is obviously a concern when irradiating foods which contain significant quantities of unsaturated fats; and the irradiation and subsequent storage of these foods in the absence of oxygen is advisable.

Loss of vitamins during processing is an obvious concern and has been studied in great detail in a variety of foods [10, 11]. Inactivation results mainly from their reaction with radiolytic products of water and is dependent on the chemical structure of the vitamin. The degree of inactivation is also determined by the composition of the food, as other food components can act as 'quencher', in competition for the reactive products. This has led to an overestimation of vitamin losses in the literature where irradiation of pure solutions of vitamins has been studied. Another consideration is the role of postirradiation storage where, in common with other processing methods, further breakdown of some vitamins may occur. Hence, losses of vitamins during irradiation vary from vitamin to vitamin and from food to food and are obviously dose-dependent. In addition, subsequent storage or processing of food must be considered.

According to Stewart [1], the sensitivity of water-soluble vitamins to irradiation follows the order: vitamin B<sub>1</sub>>vitamin C>vitamin B<sub>6</sub>>vitamin B<sub>2</sub>>folate=niacin>vitamin B<sub>12</sub>, while the sensitivity of fat-soluble vitamins follows the order: vitamin E>carotene>vitamin A>vitamin D>vitamin K.

As a general rule, vitamin losses during food irradiation are quite modest compared to other forms of processing. In fact, many studies have demonstrated 100% retention of individual vitamins following low or medium dose treatments.

## 5.6

### Detection Methods for Irradiated Foods

There is a clear need to be able to distinguish between irradiated and nonirradiated foods. This would permit proof of authenticity of products labelled as irradiated, or conversely, of unlabelled products which had been irradiated. In addition, it would be useful to be able to estimate accurately the dose to which a food had been treated. Solving these issues would improve consumer confidence in the technology and would benefit international trade in irradiated foods, especially where legislation differs between countries. However, as discussed in Section 5.5, the physicochemical changes occurring during food irradiation are minimal and thus detection is difficult. The changes measured may not be specific to radiation processing and may alter during subsequent storage. Also the compositional and structural differences between foods and the different doses required for different purposes mean that it is unlikely that a universal detection method is possible. Various approaches have been studied in many different foods with some success; and the subject has been reviewed in full by Stewart [12] and Diehl [3]. Methods currently available depend on physical, chemical, biological and microbiological changes occurring during irradiation and are summarised only briefly below.

Electron Spin Resonance (ESR) can detect free radicals produced by radiation, but these are very short-lived in high moisture foods (see Section 5.2.2). However, in foods containing components with high dry matter, such as bones, shells, seeds or crystalline sugars, the free radicals remain stable and may be detected. ESR has been successfully demonstrated in some meats and poultry, fish and shellfish, berries, nuts, stone fruit spices and dried products.

Luminescence techniques are also very promising and are based on the fact that excited electrons become trapped in some materials during irradiation. The trapped energy can be released and measured as emitted light, either by heat in thermoluminescence (TL), or by light in photostimulated luminescence (PSL). In fact the luminescence is produced in trapped mineral grains rather than in the actual food, but successful tests have been demonstrated in a wide range of foods where mineral grains can be physically separated.

Other physical principles, including viscosity changes to starch, changes in electrical impedance of living tissues and near infrared reflectance, hold some promise but suffer from errors due to variation between unprocessed samples, dose thresholds and elapsed time from processing.

The most promising chemical methods are the detection of long-chain hydrocarbons and 2-alkylcyclobutanones which are formed by radiolysis of lipids and are hence limited to foods with quite high fat contents. Numerous other potentially useful chemical changes have been studied with limited success.

DNA is the main target for ionising radiation; and hence it is logical that DNA damage should be an index for detection. The 'comet assay' is used to detect the presence of tails of fragmented DNA produced by irradiation, on electrophoresis gels, as opposed to more distinct nuclei from nonirradiated samples.

It is limited by the fact that cooking and other processing damages DNA; but it is nonetheless promising, as it is applicable to most foods.

Microbiological methods involve looking at changes in populations of microorganisms which may have resulted from irradiation. They are nonspecific, but may act as useful screening procedures for large numbers of samples.

A further development is the use of antibody assays in which antibodies are raised to products of radiolysis and incorporated into enzyme-linked immunosorbent assays. These tests are rapid and specific, but not yet in routine use.

## 5.7 Applications and Potential Applications

In practice, the application of irradiation is limited by legal requirements. Approximately 40 countries have cleared food irradiation within specified dose limits for specific foods. This does not mean, however, that the process is carried out in all these countries, or indeed that irradiated foods are freely available within them, the UK being a prime example of a country where irradiation of many foods is permitted, but none is actually carried out. The picture is further complicated by trade agreements, labelling requirements, etc.; and legal questions will not be pursued here. Legislation generally requires that irradiated foods, or foods containing irradiated ingredients, be labelled appropriately. The 'radura' symbol (see Fig. 5.9) is accepted by many countries.

One general principle has been to limit the overall average dose to 10 kGy, which essentially means that radappertisation of foods is not an option. This figure was adopted by the WHO/FAO Codex Alimentarius Commission in 1983 [13, 14] as a result of masses of research studying the nutritional, toxicological and microbiological properties of foods irradiated up to this level. There was never any implication, however, that larger doses were unsafe; and there have been exceptions to this generalisation. South Africa has permitted average doses up to 45 kGy to be used in the production of shelf-stable meat products and several countries have permitted doses greater than 10 kGy for the treatment of spices. A recent joint FAO/IAEA/WHO study group [15] concluded that doses greater than 10 kGy can be considered safe and nutritionally adequate and it would not be surprising to see more widespread national clearances of higher doses in the future.



Fig. 5.9 Radura symbol indicates that a food has been irradiated.

Two basic purposes can be achieved by food irradiation:

- extension of storage life
- prevention of foodborne illness.

It is difficult to classify the applications of food irradiation, as the process may be acting through different mechanisms in different foods or at different doses. Some foods are much more suitable for irradiation than others and the factors determining shelf life vary between foods. This section will therefore consider the general effects and mechanisms of irradiation, followed by a brief overview of applications in the major food classes.

### 5.7.1

#### General Effects and Mechanisms of Irradiation

##### 5.7.1.1 Inactivation of Microorganisms

When a population of microorganisms is irradiated, a proportion of the cells will be damaged or killed, depending on the dose. In a similar way to heat treatment, the number of surviving organisms decreases exponentially as dose is increased. A common measure of the radiation sensitivity in bacteria is the  $D_{10}$  value, which is the dose required to kill 90% of the population. Fig. 5.10 shows

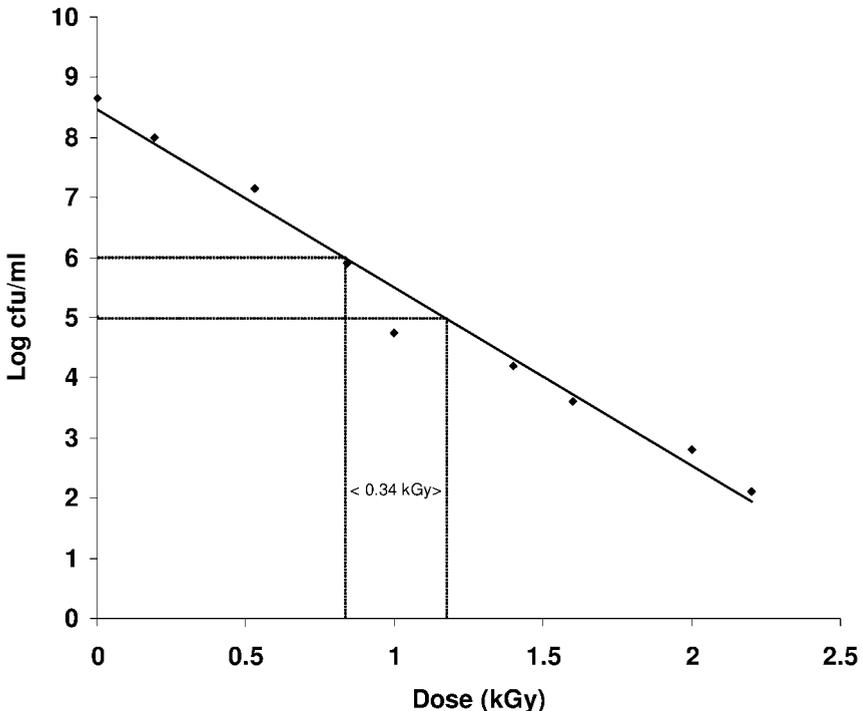


Fig. 5.10 Survival curve for electron-irradiated *Escherichia coli*.

a typical survival curve for *Escherichia coli* (data from Fielding et al. [16]) showing a  $D_{10}$  value of 0.34 kGy.

Radiation resistance varies widely among different species of bacteria, yeasts and moulds. Bacterial spores are generally more resistant than vegetative cells (see Table 5.1), which is at least partly due to their lower moisture content. Vegetative cells may contain 70% water, whereas spores contain less than 10% water. Hence indirect damage to DNA through the radiolytic products of water is much less likely in spores. An extremely important concept determining the radiation sensitivity of different species and genera of bacteria is their widely varying ability to repair DNA.  $D_{10}$  values in the range 0.03–10.0 kGy have been reported. Some organisms have developed highly efficient repair mechanisms, but fortunately these species are not pathogenic and have no role in food spoilage.

The sensitivity of microorganisms to radiation is also related to environmental conditions. Temperature of irradiation,  $a_w$ , pH and the presence of salts, nutrients or toxins such as organic acids all exert a great effect on microbial populations after irradiation. The mechanisms of effect may be through modification of the lethality of the applied dose, for example in dry or frozen environments the effectiveness may be reduced because of suppression of indirect effects caused by radiolytic products of water. Alternatively, the environmental conditions may undoubtedly affect the ability of the organisms to repair themselves and their ability to reproduce subsequent to treatment. Generally, reducing  $a_w$  by freezing, water removal or the addition of osmotically active substances increases the resistance of microorganisms, as the secondary effects due to radiolytic products of water are reduced. The presence of food components generally reduces sensitivity, as they can be considered to compete with microorganisms for interaction with the radiolytic products of water. pH also affects lethality in two ways: (a) radiolysis of water is pH-sensitive, and (b) pH changes affect the general functioning of cells, including the efficiency of DNA repair mechanisms. In addition, the recovery of surviving cells following irradiation is pH-dependent [16].

Radiation treatments aimed at the inactivation of microorganisms are conveniently classified as:

- Radappertisation: a treatment which aims to reduce the number and/or activity of microorganisms to such a level that they are undetectable. Properly packaged radappertised foods should keep indefinitely, without refrigeration. Doses in the range 25–50 kGy are normally required.
- Radicidation: this aims to reduce the number of viable spore-forming pathogenic bacteria to an undetectable level. Doses of 2–8 kGy are normally required.
- Radurisation: a treatment sufficient to enhance the keeping quality of foods through a substantial reduction in the numbers of viable specific spoilage organisms. Doses vary with the type of food and level of contamination, but are often in the range 1–5 kGy.

#### 5.7.1.2 Inhibition of Sprouting

The shelf life of tuber and bulb crops, such as potatoes, yams, garlic and onions, may be extended by irradiation at low dose levels. Sprouting is the major sign of deterioration during storage of these products and occurs after a time lag (dormant period) after harvest. The duration of the dormant period differs between different crops, different agricultural practices and different storage conditions, but is usually a number of weeks.

It is believed that the inhibitory effect of irradiation on sprouting results from a combination of two metabolic effects [17]. Firstly, irradiation impairs the synthesis of endogenous growth hormones such as gibberellin and indolyl-3-acetic acid, which are known to control dormancy and sprouting. Secondly, nucleic acid synthesis in the bud tissues, which form the sprouts, is thought to be suppressed. Treatments in the range 0.03–0.25 kGy are effective, depending on the commodity, while higher doses may cause deterioration of the tissue.

#### 5.7.1.3 Delay of Ripening and Senescence

Living fruits and vegetables may be irradiated to extend shelf life by delaying the physiological and biochemical processes leading to ripening. The mechanisms involved are complex and not well understood; and it is probable that different mechanisms predominate in different cultivars.

In some fruits, ripening is associated with a rapid increase in the rate of respiration and associated quality changes (flavour, colour, texture, etc.) known as the 'climacteric', which more or less coincides with eating ripeness. This represents the completion of maturation and is followed by senescence. Ripening is triggered by ethylene, which is then produced autocatalytically by the fruit. Climacteric fruit such as tomatoes, bananas or mangoes can either be harvested at full ripeness for immediate consumption, or harvested before the climacteric for storage and transport before ripening (either through endogenous or exogenous ethylene). Irradiation can be used either to delay senescence in fully ripe fruit, or to extend the preclimacteric life of unripe fruit. Applied doses of radiation are usually limited to 2 kGy and often much less, due to radiation injury to the fruit, leading to discolouration or textural damage.

Vegetables and nonclimacteric fruit, e.g. citrus, strawberries, cherries, are usually fully mature at harvest and respiration rate declines steadily thereafter. Irradiation can be used to delay the rate of senescence in these products in some instances, although other purposes such as control of fungi or sprout inhibition may be more important.

#### 5.7.1.4 Insect Disinfestation

Insects can cause damage to food as well as leading to consumer objections. Fortunately insects are sensitive to irradiation (see Table 5.1) and can be controlled by doses of 0.1–1.0 kGy. These doses may not necessarily cause immediate lethality, but will effectively stop reproduction and egg development.

#### 5.7.1.5 Elimination of Parasites

Relatively few foodborne parasites afflict humans. The two major groups are singlecelled protozoa and intestinal worms (helminths) which can occur in meats, fish, fruit and vegetables. Irradiation treatment is feasible although not widespread.

#### 5.7.1.6 Miscellaneous Effects on Food Properties and Processing

Although chemical changes to food resulting from irradiation are very limited, it is possible that irradiation could produce beneficial changes to the eating or processing quality of certain foods. There have been reports of improvements in the flavour of some foods following processing, but these are not particularly well substantiated. Chemical changes could result in textural changes in the food. The most likely examples are depolymerisation of macromolecules such as starch, which could lead to altered baking performance or changes in drying characteristics. Irradiation may cause cellular injury in some fruits, giving rise to easier release of cell contents and hence increased juice recovery from berry fruits.

#### 5.7.1.7 Combination Treatments

Combining food processes is a strategy that permits effective processing while minimising the severity of treatment. The benefits of combining low dose irradiation with heat, low temperatures, high pressures, modified atmosphere packaging or chemical preservatives have been described [18, 19]. In some cases, an additive effect of combining processes may be obtained, but synergistic combinations are sometimes observed.

The major drawback of these strategies is the economics of such complex processing regimes.

### 5.7.2

#### Applications to Particular Food Classes

A brief outline of the potential application of radiation processing of the major food classes is given. It should be reiterated that the actual practices adopted by different countries usually result from local legislation and public attitudes rather than the technical feasibility and scientific evaluation of product quality and safety.

#### 5.7.2.1 Meat and Meat Products

The application of irradiation for control of bacteria and parasites in meat and poultry is discussed in detail by Molins [20]. A major problem with these products is the development of undesirable flavours and odours, which have variously been described as 'wet dog' or 'goaty'. The precise chemical nature of the

irradiated flavour is unclear despite much research, but lipid oxidation is a contributing factor. The phenomenon is dose-dependent, species-dependent and can be minimised by irradiating at low (preferably sub-zero) temperatures or under vacuum or in an oxygen-free atmosphere. It is notable that the flavours and odours are transient and have been shown in chicken and different meats to be reduced or disappear following storage for a period of days. The sensory problems may also be masked by subsequent cooking. The phenomenon clearly limits applications and applied doses even though there is no suggestion of toxicological problems. Appropriate dose-temperature combinations that yield acceptable flavour, while achieving the intended purpose, must be evaluated for any application.

Radiation sterilisation of meat and meat products was the objective of much research in the 1950s and 1960s, largely aimed at military applications. The high doses required, e.g. 25–75 kGy, are usually prohibitive on legislative grounds, but the process is technically feasible if combined with vacuum and very low temperatures (say  $-40^{\circ}\text{C}$ ) during processing [21]. Radiation-sterilised, shelf-stable precooked meals incorporating meat and poultry have been produced for astronauts and military purposes and are produced commercially in South Africa for those taking part in outdoor pursuits.

Effective shelf life extensions of many fresh, cured or processed meats and poultry have been demonstrated with doses in the range 0.5–2.5 kGy. Increases in shelf life of 2–3 times or more, without detriment to the sensory quality, are reported [20]. Irradiation cannot, however, make up for poor manufacturing practice. For example, Roberts and Weese [22] demonstrated that quality and shelf life extension of excellent initial quality ground beef patties [ $<10^2$  aerobic colony-forming units (CFU)  $\text{g}^{-1}$ ] was much better than lower initial quality ( $10^4$  CFU  $\text{g}^{-1}$ ) materials when irradiated at the same dose. The former were microbiologically acceptable for up to 42 days at  $4^{\circ}\text{C}$ . Also, subsequent handling of irradiated meats or poultry, which may give rise to recontamination following treatment, should be avoided. It is preferable to irradiate products already packaged for retail. Packaging considerations are important for both preventing recontamination and inhibiting growth of contaminating microorganisms. Vacuum or modified atmosphere packaging may be useful options. It is important to note that such products continue to require refrigeration following treatment.

Elimination of pathogens in vegetative cell form is a further aim of irradiating meat and poultry products and may go hand in hand with shelf life extension. Many studies have reported effective destruction of different pathogens in meat – reviewed by many authors including Farkas [23] and Lee et al. [24]. Much work has focused on *Campylobacter* spp in pork and *Salmonella* spp in chicken, but recent outbreaks of food poisoning attributed to *Escherichia coli* 0157:H7 in hamburgers and other meats have gained attention and accelerated FDA approval of treatment of red meat in the USA.

### 5.7.2.2 Fish and Shellfish

Both finfish and shellfish are highly perishable unless frozen on board fishing vessels or very shortly after harvesting. If unfrozen, their quality depends on rapid ice cooling. Irradiation can play a role in preservation and distribution of unfrozen fish and shellfish, with minimal sensory problems. The subject is reviewed extensively by Kilgen [25] and Nickerson et al. [26]. The mechanism of action involves reducing the microbial load (hence extending refrigerated shelf life), inactivating parasites and reducing pathogenic organisms. Low and medium dose irradiation (up to 5 kGy) has been studied as a means of preserving many varieties of finfish and commercially important shellfish, and shelf life extensions of up to 1 month under refrigerated storage have been reported in many species, without product deterioration. On board irradiation of eviscerated, fresh, iced fish has been suggested.

Problems have been noted with fatty fish, such as flounder and sole, which must be irradiated and stored in the absence of oxygen to avoid rancidity. Salmon and trout exhibit bleaching of the carotenoid pigments following irradiation, which may limit the application [4]. Combination treatments involving low dose irradiation (1 kGy) with either chemical dips, e.g. potassium sorbate or sodium tripolyphosphate, or elevated CO<sub>2</sub> atmospheres may be effective for the treatment of fresh fish [27].

Strategies for irradiation treatment of frozen fish products, dried fish, fish paste and other fish products have been investigated [27].

### 5.7.2.3 Fruits and Vegetables

There are a number of purposes for irradiating fresh fruits and vegetables, including the extension of shelf life by the delay of ripening and senescence, the control of fungal pathogens which lead to rotting and insect disinfestation [28]. Radurisation and radication of processed fruits and vegetables have also been studied. The unusual property of fruits and vegetables is that they consist of living tissue, hence irradiation may effect life changes on the product and such changes may not be immediately apparent, but result in delayed effects.

The main problems associated with these products are textural problems which result from radiation-induced depolymerisation of cellulose, hemicellulose, starch and pectin, leading to softening of the tissue. The effect is dose-related and hence severely limits the doses applied. Nutritional losses at such doses are minimal. Other disorders include discolouration of skin, internal browning and increased susceptibility to chilling injury. Tolerance to radiation varies markedly, for example some citrus fruits can withstand doses of 7.5 kGy, whereas avocados may be sensitive to 0.1 kGy.

The conditions used for irradiating any fruit or vegetable cultivars are very specific, depending on the intended purpose and the susceptibility of the tissue to irradiation damage; and specific examples are discussed in detail by Urbain [4] and Thomas [17]. Combination treatments involving low dose irradiation and hot water or chemical dips, or modified atmospheres, may be effective.

#### 5.7.2.4 Bulbs and Tubers

Potatoes, sweet potatoes, yams, ginger, onions, shallots and garlic may be treated by irradiation to produce effective storage life extensions due to inhibition of sprouting as described in Section 5.6.1.2. A general guideline for dose requirements is 0.02–0.09 kGy for bulb crops and 0.05–0.15 kGy for tubers [29]. These low doses have no measurable detrimental effect on nutritional quality and are too low to produce significant reductions in microbiological contaminants.

#### 5.7.2.5 Spices and Herbs

Spices and herbs are dry materials which may contain large numbers of bacterial and fungal species, including organisms of public health significance. Small quantities of contaminated herbs and spices could inoculate large numbers of food portions and hence decontamination is essential. Chemical fumigation with ethylene oxide is now banned in many countries, on account of its toxic and potentially carcinogenic properties, and radiation treatment offers a viable alternative [27]. Fortunately, being dry products, herbs and spices are resistant to ionising radiation and can usually tolerate doses up to 10 kGy. In general, doses in the range 3–10 kGy are employed, which gives a reduction in the aerobic viable count to below  $10^3$  CFU g<sup>-1</sup> or  $10^4$  CFU g<sup>-1</sup>, which is considered equivalent to chemical fumigation. Individual treatments are discussed in detail by Farkas [30].

#### 5.7.2.6 Cereals and Cereal Products

Lorenz [31] reviewed the application of radiation to cereals and cereal products. Insects are the major problem during the storage of grains and seeds. Disinfestation is therefore the main purpose of the irradiation of cereal grain. e.g. wheat, maize, rice, barley. This can be achieved with doses of 0.2–0.5 kGy, with minimal change to the properties of flour or other cereal products [27].

Chemical fumigants, such as methyl bromide gas, are considered a health risk and could be phased out in favour of the irradiation disinfestation of grain.

Radurisation of flour for bread making, at a dose of 0.75 kGy, to control the 'rope' defect caused by *Bacillus subtilis*, gives rise to a 50% increase in the shelf life of the resulting bread. However, higher doses lead to reduced bread quality. Alternatively, finished loaves and other baked goods may be irradiated to increase storage life by suppression of mould growth, with a dose of 5 kGy.

#### 5.7.2.7 Other Miscellaneous Foods

Radiation processing of most foods has been investigated at some point. Some foods which do not appear in the above categories are described below.

Milk and dairy products are very susceptible to radiation-induced flavour changes, even at very low doses. Significant application to dairy products is

therefore unlikely, with the possible exception of Camembert cheese produced from raw milk.

Control of *Salmonella* in eggs would be a very beneficial application. However, irradiation of whole eggs causes weakening of yolk membrane and loss of yolk appearance. There may be potential for the irradiation of liquid egg white, yolk or other egg products.

Nuts contain high levels of oil and again are susceptible to lipid oxidation. Low doses to control insects, or sprouting are feasible, but higher doses required to control microbial growth are less likely.

Ready meals, such as those served on airlines or in hospitals and other institutions, offer a potential application for radiation treatments. The major problem is the variety of ingredients, each with different characteristics and radiation sensitivities. However, careful selection of ingredients, the use of low doses and combination treatments may lead to some practical applications.

## References

- 1 Stewart, E. M. **2001**, Food Irradiation Chemistry, in *Food Irradiation: Principles and Applications*, ed. R. Molins, Wiley Interscience, Chichester, pp. 37–76.
- 2 Hall, K. L., Bolt, R. O., Carroll, J. G. **1963** Radiation Chemistry of Pure Compounds, in *Radiation Effects on Organic Materials*, ed. R. O. Bolt, J. G. Carroll, Academic Press, New York, pp. 63–125.
- 3 Diehl, J. F. **1995**, *Safety of Irradiated Foods*, Marcel Dekker, New York.
- 4 Urbain, W. M. **1986**, *Food Irradiation*, Academic Press, London.
- 5 Pollard, E. C. **1966**, Phenomenology of Radiation Effects on Microorganisms, in *Encyclopedia of Medical Radiology*, vol. 2, 2nd edn, ed. A. Zuppinger, Springer, New York, pp. 1–34.
- 6 Brynjolfsson, A. **1981**, Chemiclearance of Food Irradiation: its Scientific Basis, in *Combination Processes in Food Irradiation*, Proceedings Series, International Atomic Energy Agency, Vienna, pp. 367–373.
- 7 Ehlermann, D. **2001**, Process Control and Dosimetry in Food Irradiation, in *Food Irradiation: Principles and Applications*, ed. R. Molins, Wiley Interscience, Chichester, pp. 387–413.
- 8 IAEA **1992**, *Radiation Safety of Gamma and Electron Irradiation Facilities*, Safety Series 107, International Atomic Energy Agency, Vienna.
- 9 Anon. **2001**, *Food Irradiation: Questions and Answers*, Technical Document, Food Irradiation Processing Alliance.
- 10 WHO **1994**, *Safety and Nutritional Adequacy of Irradiated Food*, World Health Organization, Geneva.
- 11 Thayer, D. W., Fox, J. B., Lakrotz, L. **1991**, Effects of Ionising Radiation on Vitamins, in *Food Irradiation*, ed. S. Thorne, Elsevier Applied Science, London, pp. 285–325.
- 12 Stewart, E. M. **2001**, Detection Methods for Irradiated Foods, in *Food Irradiation: Principles and Applications*, ed. R. Molins, Wiley Interscience, Chichester, pp. 347–386.
- 13 CAC **1984**, *Codex General Standard for Irradiated Foods*, Vol. XV, E-1, CODEX STAN 106-1983, Joint FAO/WHO Food Standards Programme, Rome.
- 14 CAC **1984**, *Recommended International Code of Practice for the Operation of Radiation Facilities Used for the Treatment of Foods*, Vol. XV, E-1, CAC/RCP 19-1979 (rev. 1), Joint FAO/WHO Food Standards Programme, Rome.
- 15 WHO **1999**, *High Dose Irradiation: Wholesomeness of Food Irradiated with Doses Above 10 kGy*, Technical Report Series No. 890, World Health Organisation, Geneva.

- 16 Fielding, L. M., Cook, P. E., Grandison, A. S. 1994, The Effect of Electron Beam Irradiation and Modified pH on the Survival and Recovery of *Escherichia coli*, *J. Appl. Bacteriol.* 76, 412–416.
- 17 Thomas, P. 1990, Irradiation of Fruits and Vegetables, in *Food Irradiation: Principles and Applications*, ed. R. Molins, Wiley Interscience, Chichester, pp. 213–240.
- 18 Campbell-Platt, G., Grandison, A. S. 1990, Food Irradiation and Combination Processes, *Radiat. Phys. Chem.* 35, 253–257.
- 19 Patterson, M. 2001, Combination Treatments Involving Food Irradiation, in *Food Irradiation: Principles and Applications*, ed. R. Molins, Wiley Interscience, Chichester, pp. 313–327.
- 20 Molins, R. A. 2001, Irradiation of Meats and Poultry, in *Food Irradiation: Principles and Applications*, ed. R. Molins, Wiley Interscience, Chichester, pp. 131–191.
- 21 Thayer, D. W. 2001, Development of Irradiated Shelf-Stable Meat and Poultry Products, in *Food Irradiation: Principles and Applications*, ed. R. Molins, Wiley Interscience, Chichester, pp. 329–345.
- 22 Roberts, W. T., Weese, J. O. 1998, Shelf-Life of Ground Beef Patties Treated by Gamma Radiation, *J. Food Prot.* 61, 1387–1389.
- 23 Farkas, J. 1987, Decontamination, Including Parasite Control, of Dried, Chilled and Frozen Foods by Irradiation, *Acta Aliment.* 16, 351–384.
- 24 Lee, M., Sebranek, J. G., Olson, D. G., Dickson, J. S. 1996, Irradiation and Packaging of Fresh Meat and Poultry, *J. Food Prot.* 59, 62–72.
- 25 Kilgen, M. B. 2001, Irradiation Processing of Fish and Shellfish Products, in *Food Irradiation: Principles and Applications*, ed. R. Molins, Wiley Interscience, Chichester, pp. 193–211.
- 26 Nickerson, J. F. R., Licciardello, J. J., Ron-sivalli, L. J. 1983, Radurization and Rad-icidation: Fish and Shellfish, in *Preserva-tion of Food by Ionizing Radiation*, ed. E. S. Josephson, M. S. Peterson, CRC Press, Boca Raton, pp. 12–82.
- 27 Wilkinson, V. M., Gould, G. W. 1996, *Food Irradiation: A Reference Guide*, Butterworth-Heinemann, Oxford.
- 28 Kader, A. A. 1986, Potential Applications of Ionising Radiation in Postharvest Handling of Fresh Fruits and Vegetables, *Food Technol.* 40, 117–121.
- 29 Thomas, P. 2001, Radiation Treatment for Control of Sprouting, in *Food Irradia-tion: Principles and Applications*, ed. R. Molins, Wiley Interscience, Chichester, pp. 241–271.
- 30 Farkas, J. 2001, Radiation Decontamina-tion of Spices, Herbs, Condiments and Other Dried Food Ingredients, in *Food Irradiation: Principles and Applications*, ed. R. Molins, Wiley Interscience, Chi-chester, pp. 291–312.
- 31 Lorenz, K. 1975, Irradiation of Cereal Grains and Cereal Grain Products, *CRC Crit. Rev. Food Sci. Nutr.* 6, 317–382.