Irradiation of Spices – a Review

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Abstract

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Food irradiation is a process of exposing food to ionising radiation such as gamma rays emitted from the radioisotopes ^{60}Co and ^{137}Cs, or high energy electrons and X-rays produced by machine sources. The use of ionising radiation to destroy harmful biological organisms in food is considered a safe, well proven process that has found many applications. Depending on the absorbed dose of radiation, various effects can be achieved resulting in reduced storage losses, extended shelf life and/or improved microbiological and parasitological safety of foods. The most common irradiated commercial products are spices and vegetable seasonings. Spice irradiation is increasingly recognised as a method that reduces post-harvest losses, ensures hygienic quality, and facilitates trade with food products. This article reviews recent activities concerning food irradiation, focusing on the irradiation of spices and dried vegetable seasonings from the food safety aspect.

Keywords: food irradiation; spices; essential oils; GC; GC/MS; sensory quality; olfactometry; antioxidant activity; EPR spectroscopy; viscometry

Contamination of food with microorganisms, particularly pathogenic non-sporeforming bacteria, is one of the most significant public health problems and an important cause of human suffering all over the world. According to the World Health Organisation (WHO), in 1992 the infectious and parasitic diseases represented the most frequent cause of death (35%) worldwide, the majority of which occurred in developing countries (LOAHARANU 1994). While the thermal pasteurisation of liquid foods is a well-established and satisfactory method of terminal decontamination/disinfection of such commodities, it has been shown inappropriate for solid foods and dry ingredients, or for fresh foods whose raw characteristics must be maintained to fulfil specific market requirements. Due to these reasons and keeping in mind the universality of the food spoilage problem, food irradiation has become one of the most promising programmes that attracted many countries during the movement to use "Atoms for Peace" (BOISSEAU 1994). However, practical limitations precluded early industrial and commercial development and the application of these concepts. Thus, significant industrial utilisation of food irradiation has become widespread only recently. To speed up the process of irradiation implementation, the WHO Food Safety Unit has described food irradiation as possibly the most significant contribution to public health to be made by food science and technology since the pasteurisation of milk at the end of the 19th century.

Radiation pasteurisation with low doses of gamma rays, X-rays, and electrons effectively controls foodborne pathogens. Irradiation leads to the destruction of pathogenic non-spore forming foodborne bacteria and parasitic organisms, such as trichina. As a consequence, it protects the consumers from microorganisms-related diseases such as salmonellosis, hemorrhagic diarrhoea caused by *Escherichia coli*,

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or gastroenteritis from *Vibrio vulnificus* (Thayer *et al*. 1996). The application of ionising radiation in food processing is based mainly on the principle that ionising radiation causes very effective disruption of DNA molecules in the nuclei of cells (Diehl 1995) rendering them inactivated. Therefore microorganisms, insect gametes, and plant meristems are prevented from their reproduction, which consequently results in various preservative effects as a function of the absorbed radiation dose (Table 1), while chemical or other radiation-induced changes in food are minimal (Thayer 1990).

An important reason for the relatively high sensitivity of DNA to the effects of ionising radiation is the fact that DNA molecules are much larger than other molecular structures inside the cell. The damage is either direct, caused by reactive oxygen-centred (•OH) radicals originating from the radiolysis of water, or indirect. In the case of an indirect hit, the damage to the nucleic acids occurs when radiation ionises an adjacent molecule, which in turn reacts with the genetic material. In view of the fact that water is a major component of most foods and microbes, it is often the adjacent molecule that ends up producing a lethal product (Grecz *et al*. 1983). According to the paper published by Arena (1971), ionising radiation causes water molecule to loose an electron producing H_2O^+. This product immediately reacts with other water molecules to produce a number of compounds, including hydrogen and hydroxyl radicals ($OH^•$), molecular hydrogen, oxygen, and hydrogen peroxide (H_2O_2). Hydroxyl radicals are very reactive and are known to interfere with the bonds between nucleic acids within a single strand or between opposite strands. Although biological systems have a capacity to repair both single-stranded and double-stranded breaks of the DNA backbone, the damage occurring from ionising radiation is random and extensive (Razskazovskiy *et al*. 2003). Therefore, recovery processes in bacteria after their radiation damage are unlikely to occur.

The differences in sensitivity to radiation among microorganisms are related to the differences in their chemical and physical structure, and in their ability to recover from the radiation injury. The amount of radiation energy required to control microorganisms in food, therefore, varies depending on the resistance of the particular species and the number of organisms present. Besides inherent abilities of microorganisms, several environmental factors such as the composition of the medium, moisture content, temperature during irradiation, presence or absence of oxygen, and others, significantly influence their radiation resistance, particularly in vegetative cells. The actual dose employed is a balance between that what is needed and that what can be tolerated by the product without objectionable changes (e.g. off-flavours, texture changes, flavour alterations).

According to the Codex General Standard for Irradiated Foods (CAC 2003), ionising radiation foreseen for food processing is limited to high energy photons (gamma rays of radionuclides ^{60}Co and, to a much smaller extent, ^{137}Cs, or X-rays from machine sources with energies up to 5 MeV, or accelerated electrons with energies up to 10 MeV produced by electron accelerating machines. These types of radiation are chosen because:
- they produce the desired food preservative effects;
- they do not induce radioactivity in foods or packaging materials;
- they are available in quantities and at costs that allow commercial use of the irradiation process (Farkas 2004).

The radiation treatment causes only minimal temperature rise in the product and can be applied through packaging materials including those that

Table 1. Directives for dose requirements of various applications of food irradiation (Farkas 2006)

Preservative effects and types of application	Dose requirements (kGy)
Killing and sterilising insects (disinfestations of food)	0.2–0.8
Prevention of reproduction of food-borne parasites	0.1–3.0
Decrease of after-ripening and delaying senescence of some fruit and vegetables; extension of shelf-life of food by reduction of microbial populations	0.5–5.0
Elimination of viable non-sporeforming pathogenic microorganisms (other than viruses) in fresh and frozen food	1.0–7.0
Reduction or elimination of microbial of microbial population in dry food ingredients	3.0–10

cannot withstand heat. This means also that the radiation treatment can be performed also after packaging, thus re-contamination or re-infestation of the product is avoided.

Long-term animal feeding studies have demonstrated that radiation-pasteurised or -sterilised foods are safe and nutritious also for humans (Thayer *et al.* 1996). Toxicological and nutritional tests have confirmed the safety of foods irradiated at doses below 10 kGy (Thayer 1994; Smith & Pillai 2004). The Directive 1999/3/EC established a Community list of foods and food ingredients that may be treated with ionising radiation (EC 1999). According to this directive, maximum allowed overall average absorbed dose is 10 kGy for dried aromatic herbs, spices, and vegetable seasonings. The U.S. Food and Drug Administration (FDA) set a limit for irradiation treatment of culinary herbs, seeds, spices, vegetable seasonings, and blends of these aromatic vegetable substances that must not exceed 30 kGy (Bendini *et al.* 1998; Olson 1998).

The radiation pasteurisation process for many foods has been approved or endorsed by many world agencies and associations such as the FDA, WHO, the Codex Alimentarius Commission, the American Medical Association, the Institute of Food Technologies, and the health authorities in approximately 40 countries (Thayer *et al.* 1996).

Safety and effectiveness of irradiation

In 1983, Becker stated that food cannot become radioactive through exposure to gamma rays from ^{60}Co, ^{137}Cs, X-rays of 5 MeV or less, or accelerated electrons with energy levels below 10 MeV (Becker 1983). This is a very mild treatment as a radiation dose of 1 kGy represents the absorption of only enough energy to increase the temperature of the product by 0.36°C. In fact, heating, drying, and cooking may cause higher nutritional losses. Moreover, certain carcinogenic aromatic and heterocyclic ring compounds produced during thermal processing of food at high temperatures were not identified in food after irradiation (Becker 1983). In general, food macronutrients (carbohydrates, proteins, and lipids) and most micronutrients (mainly water-soluble and fat-soluble vitamins) are not appreciably affected by 10 kGy-range ionising dose with regard to their nutrient contents. However, with higher radiation doses (above 10 kGy – exceeding permitted limit in the EU), the structural properties of the fibrous carbohydrates can be degraded, and lipids can become somewhat rancid, leading to a loss of food quality (Miller 2005). Moreover, the irradiation of lipids at high doses, and especially in the presence of oxygen, can lead to the formation of liquid hydroperoxides. The oxidation products formed often have undesirable odours and flavours (rancidity). It is known that the unsaturated fatty acids are more prone to develop rancidity. Lipid oxidation can be significantly reduced by freezing, and/or by oxygen removal prior to irradiation.

Of the micronutrients, thiamine is of concern because of its relatively high sensitivity to the effects of radiation. Foods that contain thiamine (e.g. pork) are suitable as indicators of food safety regarding the irradiation treatment (Miller 2005). Minerals have been shown to remain stable (Diehl 1995).

Besides the nutritional and sensory values, the wholesomeness (lack of mutagenicity, teratogenicity, and toxicity) of irradiated foods has been studied extensively (Thayer 1990). Neither short nor multigenerational feeding studies have produced any evidence of toxicological effects in mammals due to ingestion of irradiated foods. In fact, multigenerational studies with animals have demonstrated that the ingestion of irradiated foods is completely safe and that the nutritive value remains essentially unchanged (Thayer *et al.* 1996). The data support the conclusion that properly processed irradiated foods are wholesome. Radiolytic changes in foods are minimal and are predictable from the radiation chemistry of principal food components. Furthermore, possible radiolytic products derived, e.g. from lipids (most of which are saturated and unsaturated hydrocarbons, aldehydes, and 2-alkylcyclobutanones) are neither unique nor toxicologically significant in the quantities found in irradiated foods (Chinn 1979; Urbain 1986; Swalow 1991; Diehl 1994, 1995). Besides the improvements in food safety through the destruction of microflora, irradiation provides also other benefits. These include an increased shelf life of fruits, vegetables, spices (Thayer & Rajkowski 1999), and meat (Thayer 1993; Murano *et al.* 1998), and provide a suitable alternative to chemical treatments, especially for the decontamination of fruits, vegetables, and spices (Boisseau 1994).

Irradiation of spices and dried vegetable seasonings

Spices, even when used in small amounts, present a potential source of microbial pollution for foodstuffs to which they were added. Spices often originate in developing countries where harvest and storage conditions are inadequately controlled with respect to food hygiene. Thus, they may have been exposed to a high level of natural contamination by mesophylic, sporogenic, and asporogenic bacteria, hyphomycetes, and faecal coliforms (Bendini *et al.* 1998). Most spices are dried in the open air and can become seriously contaminated by air- and soil-borne bacteria, fungi, and insects. Microorganisms of public health significance such as *Salmonella*, *Escherichia coli*, *Clostridium perfringens*, *Bacillus cereus*, and toxigenic moulds can also be present. Bacterial plate counts of one to 100 million per gram of spice are usual (Bendini *et al.* 1998). Good manufacturing practices during harvest and processing could improve their hygienic quality, but frequently not to an extent sufficient to obtain an acceptable microbiological purity level (WHO 1999). Contaminated dry plant ingredients cause serious troubles in the food processing industry. Therefore, many commercial food processors fumigate spices with methyl bromide to eliminate insects or with ethylene oxide to eliminate bacteria and moulds. However, it has been found that both methyl bromide and ethylene oxide are extremely toxic compounds. Moreover, methyl bromide is potentially capable of depleting the atmospheric ozone layer. Ethylene oxide has been banned in Europe because of safety and environmental concerns, and its use for the treatment of ground spice has been banned in the United States (Loaharanu 1994). The U.S. Clean Air Act and the Montreal Protocol of the Vienna Convention require that any substance listed as ozone depleting be withdrawn from production and use by the year 2001.

Spices, herbs, and dried vegetable seasonings are currently treated with ionising radiation to eliminate microbial contamination. It was unambiguously confirmed that the treatment with ionising energy is more effective against bacteria than the thermal treatment, and does not leave chemical residues in the food product (Tjaberg *et al.* 1972; Loaharanu 1994; Thayer *et al.* 1996; Olson 1998). Thus, ethylene oxide and methyl bromide treatments can be effectively replaced by food irradiation, which in fact is less harmful to the spices than heat sterilisation, which implicates the loss of thermolabile aromatic volatiles and/or causes additional thermally induced changes (e.g. thermal decomposition or production of thermally induced radicals).

Effect of irradiation dose on volatiles and impact on organoleptic changes

Many approaches can be applied for studying the effects of irradiation on the volatile compounds and the impact on organoleptic changes of volatile oils in spices. These include methods of gas chromatography with flame-ionisation detection (GC/FID), gas chromatography-mass spectrometry (GC/MS), sensory evaluation, and GC-olfactometry. To compare two methods of decontamination, powdered black pepper was irradiated with different recommended doses of gamma rays (5 and 10 kGy, respectively) and treated with microwaves for different periods (20, 40 and 75 s) (Emam *et al.* 1995). The results obtained indicate that irradiation treatment with controlled doses of gamma irradiation is a safe and suitable technique for decontamination of black pepper. In comparison with microwave treatment, irradiation does not result in extensive loss of flavour compounds.

In contrast to the radiation treatment, the thermal treatment of black pepper (using 130°C hot dry steam for 4 min; internal temperature of the treated berries was 95°C) caused a significant increase in the content of monoterpenes (Sádecká *et al.* 2005a). These changes might have resulted from the formation of thermal isomerisation products of some terpenes as shown by Richard and Jennings (1971) and Farag Zaied *et al.* (1996). However, the qualitative composition of volatile oils obtained from control, thermally treated sample, and from irradiated samples of black pepper at various doses (up to 30 kGy) was identical. Statistical analysis of the effects of irradiation on the total content of volatile oils in spices showed no significant differences between irradiated and non-irradiated samples. Sádecká *et al.* (2005b) showed that the most important changes were observed in the black pepper sample irradiated up to the dose of 30 kGy, which resulted in a threefold increase of caryophyllene oxide concentration and a parallel decrease of sesquiterpene caryophyllene, in comparison with an untreated sample. Nevertheless, such a dose of ionising energy exceeds three times the level permitted by the EU legislation (EC 1999). The observed changes induced in terpenes by irradiation could be explained by oxidation or hydroxylation of aromatic rings in terpene molecules. Urbain (1986) showed that the products with a low moisture content (10%) are prone to the formation of free radicals by irradiation which leads to an increased production of oxides and alcohols. In addition, the configuration of the side groups on the terpene skeleton, especially the position of double bonds and functional groups, can result in a variety of compounds produced.

The research has demonstrated that gamma irradiation at the dose of 10 kGy (toxicologically and nutritionally confirmed maximum safe dose) can eliminate microbial load of spices without causing any significant organoleptic or chemical alterations (Farkas 1973, 1985, 1987; Farkas *et al.* 1973; Kiss *et al.* 1978; Ito *et al.* 1985; Mossei 1985; Narvaiz *et al.* 1989; Sádecká *et al.* 2004, 2005a, b). Lescano *et al.* (1991) showed that even the treatment at the dose of 30 kGy of ginger, turmeric, cayenne pepper, onion, and garlic powders did not alter their seasoning capacity (odour, flavour and pungency). Analogous results were reported in our paper published recently (Sádecká *et al.* 2005b). GC-olfactometry analysis using the method of Aroma Extract Dilution Analysis (AEDA) (Grosch 1993) of volatile oils in black pepper, oregano, and pimento revealed that even a high dose of radiation (30 kGy) had no significant effect and the overall aroma of the treated spices remained unaltered. The exposure of carbohydrates, proteins and lipids to high doses of ionising radiation is known to result in the formation of additional, radiolytically induced compounds. Their origin, structural properties, and quantity are notably influenced by the presence of water. Bendini *et al.* (1998) showed that dried foods, such as spices, are less sensitive to the ionisation energy than the hydrated ones. In 1992, USA authorised the ionisation treatment at doses below 30 kGy for microbial decontamination of dried or dehydrated herbs, spices, and vegetable seasonings that are used in small amounts as food ingredients. On the contrary, Antonelli *et al.* (1998) observed that the composition of dried basil leaf essential oils treated at doses of 5 and 10 kGy was different in comparison to the blank sample. They concluded that radiation caused more evident changes in the composition profiles than the microwave treatment. Subsequently, a sensory test confirmed significant differences between the extracts. The panellists preferred the gamma treated sample, while the microwaved sample was the least appreciated. This discordance in the literature data indicates that it is necessary to standardise analytical procedures in order to obtain results that are intercomparable.

Ionising radiation level versus antioxidant activity

Only a few studies address the influence of irradiation procedures on the antioxidant activity of herbs and spices. Murcia *et al.* (2004) evaluated the effect of this processing technique on the antioxidant properties of seven dessert spices (anise, cinnamon, ginger, liquorice, mint, nutmeg, and vanilla). In comparison with the non-irradiated samples, the water extracts of the irradiated spices at 1, 3, 5, and 10 kGy did not show any significant difference in the antioxidant activity in the radical-scavenging assays used. Farag and Khawas (1998) evaluated the antioxidant properties of anise, caraway, cumin, and fennel essential oils extracted from untreated, gamma-irradiated and microwaved seeds. Gamma-irradiation at 10 kGy and microwave treatments did not affect the antioxidant property of the essential oils under study. In addition, essential oils extracted from gamma-irradiated fruits were more effective antioxidants in sunflower oil than those produced from microwaved fruits. Topuz and Ozdemir (2004) analysed the capsaicinoid content insundried and dehydrated paprika samples that were irradiated at doses from 2.5 kGy to 10 kGy. The content of capsaicin, dihydrocapsaicin, and homodihydrocapsaicin was increased by about 10% in the samples irradiated at the dose of 10 kGy.

The effects of irradiation by electron beam on the colour and the contents of volatile oils in five-spice powder (prickly ash, star aniseed, cinnamon, clove, and fennel) and chilli were assessed by Lianzhong *et al.* (1998). Irradiation enhanced the UV absorption of aqueous extracts of spices, but the darkening phenomenon of spices due to irradiation was temporary. Calucci *et al.* (2003) studied the effects of gamma-irradiation at 10 kGy on the free radical formation and the antioxidant contents of nine aromatic herbs and spices (basil, bird pepper, black pepper, cinnamon, nutmeg, oregano, parsley, rosemary, and sage). Irradiation resulted in a general increase of quinone radical content (measured using electron paramagnetic resonance (EPR) spectroscopy) in all samples investigated, and in a significant decrease of the total ascorbate and carotenoids content of some spices. Calenberg *et al.* (1998) found no significant differences between EPR spectra of the samples of white pepper, sweet paprika, and nutmeg irradiated with electron beams or X-rays at 0–10 kGy. Several studies applied EPR spectroscopy to investigate free radicals formed by the gamma-radiation treatment of ground black pepper, oregano, allspice, ginger, and clove, and to evaluate the influence of the absorbed dose of gamma radiation on the radical-scavenging potential of alcoholic extracts (Franco *et al.* 2004; Suhaj *et al.* 2006; Polovka *et al.* 2006, 2007). The observed radical-scavenging (antioxidant) activity of spice extracts was only slightly influenced by the gamma radiation treatment (Polovka *et al.* 2006, 2007; Suhaj *et al.* 2006).

Detection and potential detection methods for spices irradiation

For the international food trade, simple and reliable methods are needed to identify irradiated foodstuffs. Numerous studies have dealt with the detection methods applicable to irradiated herbs and spices and have also concluded that food irradiation can be considered a radiologically, microbiologically, and toxicologically safe technology. Nevertheless, questions focusing on nutrient loss, free radical and radiolytic by-products formation, and changes of antioxidant properties during irradiation are still being discussed in the scientific community. In 1993, the European Commission gave a mandate to the European Committee for Standardisation (CEN) to standardise the methods for the detection of irradiated foods. These European Standards have been adopted by the Codex Alimentarius Commission as general methods and are referred to in the Codex General Standard for Irradiated Foods in section 6.4 on 'Post-irradiation verification'(Code of Federal Regulation 2004). In the case of spices, the most important methods are viscosity measurement, electron spin resonance (ESR), and thermoluminiscence (TL).

In relation to the formation of paramagnetic species upon gamma-irradiation food processing, EPR spectroscopy represents a unique detection technique for their characterisation and investigation. In 2000, CEN issued the EN 1787 EPR method for the detection of irradiated foods containing cellulose. The gamma radiation treatment of plant products containing cellulose leads to the generation of a typical three-line EPR signal (Yordanov *et al.* 1998, 2000; Raffi *et al.* 2000), attributable to "cellulosic" radicals. Later, more standardised methods were published for the detection of irradiated foods (i.e. EN 13708, EN 13751, and EN 13784). EN 1784 and EN 1785 describe methods for the detection of irradiated food containing fat, and EN 1786 a method for the detection of irradiated food containing bone. Unfortunately, their application is limited by the lifetime of the radiolytically produced free radicals (Yordanov *et al.* 1998; Formanek *et al.* 1999; Raffi *et al.* 2000; Yordanov *et al.* 2000; Delincée & Soika 2002; Bayram & Delincée 2004; Suhaj *et al.* 2006; Polovka *et al.* 2006, 2007). Raffi and Stocker (1996) observed that, even though electron spin resonance is known to be a very sensitive method, in the case of spices it did not lead to favourable results because the main radio-induced signal decreased too fast with the storage time and disappeared before the maximal usual commercial storage time. On the other hand, other authors (Polonia *et al.* 1995) showed a prolonged appearance of cellulose peaks in dried paprika. This enabled to include the EPR method in the European protocol (Anonymous 1995). For the purpose of post-radiation identification of cellulose containing foods, the presence of relatively weak satellite lines of this "cellulosic" radical species was accepted as an unambiguous evidence for gamma-radiation treatment (Yordanov *et al.* 1998, 2000). However, it was observed that the EPR intensity of the "cellulosic" triplet signal gradually decreased with the storage time, and that the rate of disappearance was dependent on temperature, humidity, the presence of oxygen, and other factors (Yordanov *et al.* 1998, 2000, 2004; Bayram & Delincée 2004; Polovka *et al.* 2006, 2007; Suhaj *et al.* 2006). In general, the cellulosic EPR signal disappeared within 70 days to 90 days after the irradiation process (Raffi *et al.* 2000). Consequently, the method was deemed to fail in the verification of the gamma-radiation treatment and, consequently, it was recommended that thermoluminescence techniques should be used (Yordanov *et al.* 1998; Raffi *et al.* 2000; Delincée & Soika 2002; Bayram & Delincée 2004). Recently, Yordanov *et al.* (2000) pointed to the different thermal behaviour of EPR signals of non-irradiated and gamma-radiation treated foods containing cellulose, even after a long storage period after radiation, when the specific "cellulosic" EPR signal is extremely low, and recommended this technique as a method to identify gamma-radiation processed foods.

Formanek *et al.* (1999) utilised EPR spectroscopy and viscometry (with two different sample preparation methods) to detect irradiated black pepper samples. They concluded that the identification of irradiation at doses above 8 kGy is possible using these methods, at least within one month of post-irradiation storage at ambient temperature. The viscosity measurement was reported to be a promising method for the identification of irradiated spices by Mohr and Wichmann (1985) and Heide *et al.* (1987, 1988). They described a method using heat gelatinisation of starch in different spices. In agreement with their findings, Farkas *et al.* (1990a, b) reported a dramatic decrease in the dispersion viscosity of heat gelatinised suspensions of several irradiated spices with the starch content compared to that of unirradiated samples. This approach may provide a relatively simple diagnostic technique for the detection of irradiation treatment of starch-containing spices. Because the effect mentioned seems to be related to the radio-depolymerisation of starch in the irradiated spices, additional analytical techniques have been tested to investigate the starch damage in black and white peppers (Farkas *et al.* 1990a, b). The colorimetrically determined reducing sugar content as well as the alcohol-induced turbidity of hot-water extracts indicated an increased starch damage in the pepper samples as a function of the irradiation dose. However, the effect of irradiation had a less dramatic response in these tests than in the viscometric test. The moisture content influences partial radio-depolymerisation of starch. According to the experimental results, the technique of differential scanning calorimetry (DSC), measuring the energy and temperature characteristics of heat gelatinisation of starches, can not rival the sensitivity of viscometric measurements in the detection of radiation-induced changes (Farkas *et al.* 1990a). Formanek *et al.* (1994) and Barabassy *et al.* (1995, 1996) suggested an alternative method to that of heat gelatinisation of starch, which is less time-consuming because it does not require heat gelatinisation. In the case of cinnamon and allspice (15% suspensions, particle size less than 0.16 mm), the apparent viscosities seemed to be as sensitive to the irradiation dose as those of heat gelatinised spices. A comparison of three physical methods (viscometry, DSC, NIR-near infrared spectrophotometry) used in the identification of irradiated spices (cinnamon and allspice) revealed that the apparent viscosity test demonstrated the best response sensitivity when applied to samples irradiated with different doses. The storage time did not influence the apparent viscosity values. The identification limit of the viscometric method was determined at 2–3 kGy, whereas the limit of NIR spectrophotometric method was determined at 4–5 kGy, respectively. These two methods enabled to distinguish and correctly order the irradiated samples. Ukai and Shimoyama (2003) focused on the thermal behaviour of free organic radicals induced in irradiated black pepper. They found that the radical evolution in the irradiated pepper obeys a single exponential function and yields a unique time constant. Chabane *et al.* (2001) used thermoluminescence, EPR spectroscopy, and viscosimetric measurements to determine whether or not a spice had been irradiated. They confirmed that thermoluminescence, using the EN 1788 (2001) official protocol, with an alternative method for the extraction of mineral impurities, led to the proof of irradiation. EPR could be used as a proof of irradiation just up to several weeks after irradiation, and only for some spices.

Polovka *et al.* (2006, 2007) and Suhaj *et al.* (2006) recently confirmed using EPR spectroscopy that the gamma-radiation treatment of cellulose-containing spice samples such as ground black pepper (*Piper nigrum* L.), allspice berries (*Pimenta officinalis* L.), ginger root (*Zingiber officinale* Rosc.), dried clove buds (*Caryophyllus aromaticus* L.), and dried oregano leaves (*Origanum vulgare* L.) resulted in the dose-dependent generation of paramagnetic species of different structures and properties. Their behaviour is significantly affected by temperature, relative humidity, and storage conditions. All these factors have to be taken into account in order to survey the changes induced by the absorption of gamma-radiation. EPR spectra of all reference (non-irradiated) samples represent a broad singlet line with unresolved hyperfine splitting, attributable to Mn(II) ions, upon which an additional narrow EPR signal is superimposed, assigned to stable semiquinone radicals produced by the oxidation of polyphenolics in plants. The analysis of the individual EPR spectra of radiation treated spices showed the formation of new paramagnetic structures of different origin (mostly cellulose and carbohydrate), which exhibited diverse thermal stability and lifetime. The differences between spices indicate that although the spice matrices are very similar, they represent a complex system, and the impact of gamma-irradiation is strongly influenced by the presence of its characteristic specific constituents.

In addition to EPR spectroscopy, microgel electrophoresis (DNA comet assay) may be used for the identification of irradiated spices (Khan *et al.* 2002). The detection was successful in the case of poppy seeds, cardamom seeds, caraway seeds, and nigella seeds, but not in pomegranate seeds, ginger root, juniper berries, black peppercorns, nutmeg seed, and rosemary leaves. Nevertheless, for some irradiated foods, DNA comet assay is a rapid and inexpensive screening test. The direct epifluorescent filter technique/aerobic plate count (DEFT/APC) is the European Standard EN 13783 (2001) screening method for the detection of the irradiation treatment of herbs and spices.

Consumer acceptability of food irradiation

Despite the obvious benefits of the application of gamma-radiation on food, this technology remains vastly underestimated in the food industry. It has not been widely accepted and adopted yet, from our point of view due to two primary reasons, both associated with the three important sources of ionising radiation themselves, radioisotopes ^{60}Co and/or ^{137}Cs, and electron accelerators. The hindering factors in the way of commercial implementation of the food irradiation process are politics and consumer advocacy. Similar situation occurred with the heat pasteurisation of milk in the past (Farkas 2006).

Consumer attitudes to food irradiation are perceived as a crucial issue. The use of the treatment as a commercial food process depends on its acceptance by consumers. The analyses of attitudes, which vary according to country, national traditions, and political climate, have been extensively reviewed (Board 1991; Loaharanu 1993; Bruhn 1995). In the 1980s, the major concerns of consumer organisations included safety, nutrition, detection, and labelling of irradiated food. There were fears that the process would be used to upgrade low-quality products. In 1987, the International Organisation of Consumers Unions (IOCU), representing consumer organisations in member states across Europe, Asia, and Latin America, adopted a resolution on food irradiation calling for a worldwide moratorium on the subject (Feenstra & Scholten 1991). At the same time, a number of consumer organisations, including the London Food Commission and the National Coalition to Stop Food Irradiation, questioned the integrity and competence of food irradiation promoters. Health and environmental pressure groups opposed the introduction of the technology. In addition, the media emphasised concerns about food irradiation. Anti-food irradiation groups were successful in influencing legislation, with major food companies taking anti-irradiation stances (Pszczola 1990; Satin 1993). The opposition to food irradiation still exists. Recent actions by opponents of food irradiation include picketing, making inflammatory demands, and pressurising legislation. However, the IOCU has taken a more independent and unbiased approach to food irradiation. In a joint IOCU/International Consultative Group on Food Irradiation seminar on food irradiation and consumers (IAEA 1993), a number of recommendations were agreed on areas including applications, trade and environmental implications, regulation and enforcement, consumer acceptance and labelling. It is recognised that the attitudes of consumer organisations can strongly influence consumer opinions (Taylor 1989). Consumer resistance to food irradiation appears to be linked to the growth in popularity of additive-free, minimally processed foods, and environmentally acceptable food processing techniques. However, recent consumer surveys in the USA indicated that the concern about irradiation is smaller than about other food-related issues, such as food additives, pesticides, and animal drug residues (Resurreccion *et al.* 1995). The concern about the use of irradiation for foods treating appears to centre on the safety of the process. This is often linked to the fear and confusion about radiation itself and the lack of understanding of the process. Providing science-based information on food irradiation leads to positive consumer attitudes.

Consumer surveys have revealed the acceptability rates ranging from 45% to more than 90%, depending on the food commodity and the way of presentation (Fox 2002). Opinion polls reflect the level of awareness and quality of the information provided. The information about the process tends to promote acceptance. Nowadays, other authors (Nayga *et al.* 2004) reported that consumers would purchase irradiated foods, depending on their level of concern and awareness and the provision of sufficient background information. These findings emphasise the importance of educating the public on the controversy, technology, and the benefits of irradiation, especially since the public has been shown to be more receptive to the negative argument.

The key issue with the consumers is the labelling of irradiated foods. There appears to be a marked influence of informative labelling on consumers' willingness to buy irradiated food. The labelling to provide identification is not sufficient. The information that describes the purpose of the treatment promotes consumers' acceptance, e.g. for irradiated chicken, the words 'treated by irradiation to control *Salmonella* and other foodborne bacteria' (Pszczola 1993). Additional consumer education and information needs to be available in the place where the product is marketed.

There is evidence enabling to suggest that, if irradiated products offer clear advantages, and if the science-based information on the process is readily available, many consumers would be ready to buy irradiated foods.

CONCLUSION

Food irradiation has been shown to be a safe and effective process in controlling microbial contamination without adverse side effects and chemical residues that can be used to improve the safety of our food supply. The future research should focus on the organoleptic quality of irradiated foods, mainly spices, in the dependence on the dose used for ionising radiation. In this context, it is necessary to standardise analytical procedures for an unambiguous identification of gamma-radiation treatment and its influence on food quality, in order to obtain results that are intercomparable. Finally, with the aim of a better protection and objective consumer information, respecting their freedom of choice, it is necessary to make an international agreement on a careful and consistent labelling of irradiated food products.

Recently, scientists, educators, government officials, and suppliers of food irradiation equipment and services formed the International Council on Food Irradiation (ICFI 2004) to gather and disseminate information – based on sound science – on the safety and benefits of food irradiation. Scientists have the responsibility to help the consumer understand the irradiation/radiation process and its potential to improve our lives and protect our health, as does pasteurisation of milk, canning, and freezing.

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