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**Review Article** 

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# **Radiation Technology for Food Irradiation**

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#### **Abstract**

Radiation technology is considered to be a promising alternative for its high efficiency in pathogen inactivation, organic pollutants oxidation, malodor nuisance elimination, and some other characteristics enhancement, which will facilitate the downstream process of sludge treatment and disposal. Food irradiation is a process of exposing food to ionizing radiation such as gamma rays emitted from the radioisotopes Cobalt-60 and Caesium-137, or high energy electrons and X-rays produced by accelerators. The use of ionizing radiation to destroy harmful biological organisms in food is considered safe, well-proven, wholesome, and toxicologically safe over many years that can be used to increase the microbiological safety and shelf life of a wide range of foods. It is a cold process and can be used to pasteurize and sterilize foods without causing changes in food's freshness and texture, unlike heat. Unlike chemical fumigants, irradiation does not leave any harmful toxic residues in food and is more effective. Candidates for radiation decontamination are mainly poultry, red meat, egg products, grains, dried spices, vegetables, fruits, and fishery products. Nevertheless, today, radiation technology for irradiated food has become a standard technology worldwide and it holds a promise for enhancing the safety of many minimally processed foods.

Keywords: Radiation technology; Food irradiation; Cobalt-60; Biological organisms; Ionizing radiation

#### Introduction

Radiation is the emission or transmission of energy in the form of waves or particles through space or through a material medium which is often categorized as either ionizing or non-ionizing depending on the energy of the radiated particles. Ionizing radiation has long been indispensable in various sectors. Ionizing radiation in its various forms was discovered in 1895-1896 by Rontgen and Becquerel [1]. At the same time, therapeutic use was proposed for the first time and soon also the bactericidal effect of this new radiation was described. The story reported here will not illustrate all those early observations, ideas, fantasies, approaches, promises, and attempts to exploit ionizing radiation for some purpose. There have been a few great ideas which, however, for other reasons did not come to practical implications. A rather scurrile attempt was

the proposal to mix food with radioactive substances thus using 'internal' irradiation to achieve the desired effect. Food irradiation was quite an academic issue during the first half of the 20th century. It was only Eisenhower's program "Atoms for Peace" which gave the incentive to US institutions to expand research outside military applications, including food irradiation. Radiation principles explain how gamma rays, e-beams, and X-rays interact with matter. These interactions result in the formation of energetic electrons at random throughout the matter, which causes the formation of energetic molecular ions. These ions may be subject to electron capture and dissociation, as well as rapid rearrangement through ion-molecule reactions, or they may dissociate with time depending on the complexity of the molecular ion. Natural radiation and radioactiv-



ity in the environment make up the very largest part of the accumulated annual dose to human beings who are not occupationally exposed to ionizing radiation from other sources during their daily work activity [1,2].

Food irradiation has about 100 years of history and it was developed as a scientifically established technology and safe food process during the second half of the 20th century [3]. Food irradiation is a process that exposes agricultural and food commodities to ionizing radiation to enhance their shelf life and microbial safety [4]. Contamination of food with microorganisms, particularly pathogenic non-spore-forming bacteria, is one of the most significant public health problems and an important cause of human suffering all over the world [5]. Ionizing radiation is very effective in decontaminating foods, in particular in reducing and inhibiting pathogen and spoilage bacteria in raw materials and highly perishable foods. They're effective against insects and mites which can affect agricultural commodities and can be used as a quarantine treatment to eliminate pests in exported commodities to prevent their introduction into new areas [6]. Irradiation is very effective in reducing the germination of tubers and bulbs after harvesting and efficient in delaying the ripening of fruit and vegetables [7]. It is the ultimate minimal processing technology that has been profoundly studied. Although food is commonly irradiated for example with microwaves, the term food irradiation is used to describe a process where food is exposed to ionizing energy, utilizing gamma photons emitted by Cobalt-60 or Cesium-137 radioisotopes, or electron beams (high energy of up to 10 MeV), or machine-generated X-rays (high energy of up to 5 MeV) [8]. The electromagnetic radiation from the first two types of sources have good penetration ability, while accelerated electrons have low penetrability. None of these energy sources induce radioactivity in the food or its packaging, and the treatment has many technologically and technically feasible applications including significantly improving the microbiological safety and storage stability of foods. Radiation technology can complement and supplement existing technologies to ensure food security and safety. It provides an effective alternative to fumigants that are being phased out due to their adverse effects on the environment and human health [9].

#### **Radiation Sources**

Food irradiation is the best-studied food technology ever. More than 60 years of research is well documented, and during this period the sensitivity of analytical methods has increased tremendously. To avoid any measurable induced radioactivity, the number of permitted radiation sources has been limited to gamma rays from cobalt-60 or cesium-137 to electron beam up to 10 MeV particle energy, and X-rays generated from converting electrons with up to 5 MeV from machine sources. In other words, any radiation known to induce radioactivity – for example, neutrons – is not permissible [10].

# Gamma irradiator

Gamma irradiator is regarded as the simplest form of irradi-

ation, and photons are spontaneously emitted by radioactive isotopes of cobalt (Co-60) or cesium (Cs-137). The photons are relatively higher in frequency and hence energy in comparison to X-ray photons. The penetration depth can be several feet and can target microorganisms anywhere within that range. Even though gamma ( $\gamma$ ) irradiation can be simple in concept, in practice, it can be more challenging. The radioactive isotopes are produced by exposing them to a nuclear reactor core, and even after the source is selected, logistically, the exercise is complicated as the source cannot be switched off. Moreover, they do not come with directional or intensity controls [11], so the intensity must be attenuated by absorbers.

#### **Electron beam**

High-energy electron beams are produced in an electron gun, and it is easier to direct the electrons using a magnetic field. The high-energy electrons are focused into a narrow beam spot and this spot of incident electrons is scanned across food as it travels perpendicular to the beam direction, through the irradiator. The word 'irradiation' in this case could be misleading as food is not exposed to electromagnetic radiation or beta rays, but the process has a similar impact to gamma (γ) rays irradiation. Shielding during the process is still necessary but not to the extent of gamma ( $\gamma$ ) rays where concrete bunkers are used. For commercial use, the most important characteristics of an accelerator are its electron energy and average beam power. Therefore, industrial electron accelerators are usually classified according to their energy range, which is divided into low- (80-300 keV), medium- (300 keV-5 MeV), and high-energy ranges (above 5 MeV). The main drawback of the e-beam is its low penetration depth [12].

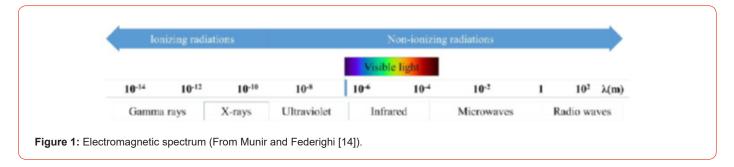
#### X-ray

X-rays are also generated by machines and can be switched off if necessary, which is a big advantage. Here electrons are accelerated at a metallic target (e.g., tantalum, tungsten, or gold) and this generates a stream of X-rays. The process is not efficient; much of the E-beam energy is lost as heat, but the X-ray conversion efficiency increases with the increasing atomic number of the metallic target material and with increasing incident E-beam energy. Nevertheless, X-ray irradiation is finding more favor in association with E-beams, and other radiation processing applications as they are more penetrating than E-beams, making it possible to process large bulk packages without the need for radioactive material. It is likely that X-ray irradiation will become more widespread in the future as technology advances [13].

#### The Radiolysis of Water

Because foodstuffs generally contain a significant amount of water, a short discussion of the radiolysis of water by irradiation with ionizing radiation (primarily  $\gamma$ -photons but also X-rays, UV photons, electrons, and neutrons) is necessary.

Non-particle ionizing radiation ( $\gamma$ -photons and X-rays) is electromagnetic radiation of very short wavelength (high frequency) as depicted in Figure 1.



A huge literature exists on this subject so only a few general sources are cited. The subject has been extensively explored because the radiolysis of water generates highly reactive species, including  $e_{aq}^-$ , H, OH, H $_2$ O $_2$ , HO $_2$ ,  $HO_2^-$ , O $_2$ , O, H $_2$ , H $_2$ , H $_3$ , H $_4$ , and possibly others [15]. These species are either oxidizing agents (e.g.,

 ${
m O_2}$ ,  ${
m O_2}$ ,  ${
m H_2O_2}$ , OH, O, O<sup>-</sup>) or reducing agents  $\left(H_2,H,e_{aq}^-,O_2^{2-},O_2^-\right)$  some of them being thermodynamically quite powerful, as measured by the standard reduction potentials summarized in Table 1.

Table 1: Selected standard redox potentials for selected radicals [15].

| Redox Couple                                     | $E^0$ / $V_{\rm she}$ | Redox Couple                     | $E^0$ / $V_{ m she}$ | Redox Couple  | $E^0/V_{\rm she}$ |
|--|-----------------------|----------------------------------|----------------------|---|-------------------|
| $e_{aq}^-$                                       | -2.87                 | OH/H <sub>2</sub> O              | 2.72                 | H <sub>2</sub> O <sub>2</sub> /H <sub>2</sub> O               | 1.77              |
| H/H <sup>+</sup>                                 | -2.31                 | H₂/H⁺                            | 0                    | 0 <sup>-</sup> /H <sub>2</sub> 0                              | 1.77              |
| H/H <sup>-</sup>                                 | 0.05                  | O <sub>2</sub> /H <sub>2</sub> O | 1.23                 | 02/02   | -0.16             |
| (O <sub>2</sub> , H+)/HO <sub>2</sub>            | 0.12                  | 0 <sub>3</sub> /0 <sub>3</sub>   | 0.83                 | 0 <sub>2</sub> +/0 <sub>2</sub>                               | 3.2               |
| NH <sub>3</sub> +/NH <sub>3</sub>                | 2.13                  | NH <sub>2</sub> /NH <sub>2</sub> | 0.7                  | NH <sub>2</sub> OH <sup>+</sup> /NH <sub>2</sub> OH           | ≤1.26             |
| NO+/NO   | 1.21                  | NO <sub>2</sub> /NO <sub>2</sub> | 1.04                 | NO <sub>2</sub> +/NO <sub>2</sub>                             | 1.51              |
| NO <sub>3</sub> -/NO <sub>3</sub> <sup>2</sup> - | <-0.40                | NO <sub>3</sub> /NO <sub>3</sub> | 2.5                  | N <sub>2</sub> H <sub>4</sub> +/N <sub>2</sub> H <sub>4</sub> | 0.01              |
| N <sub>3</sub> /N <sub>3</sub>                   | 1.33                  |                                  |                      |   |                   |

The table also contains nitrogenous species that may be generated via the homogeneous decomposition of  $N_2$  into highly reactive nitrogen atoms that may then react with water and water radiolysis products to generate a range of other species, including  $NH_3$  and the oxyanions. In neutron-irradiated systems, nitrogen is also

produced by the nuclear reaction  $^{16}O_8(^1n_0,^0e_{-1})^{16}N_7$  with  $^{16}N_7$  being radioactive. Because of neutron activation phenomena and the general unavailability of neutron sources except for research purposes, neutron irradiation is of little interest in the food industry.

**Table 2:** Model for the radiolysis of water as modified from Burns and Moore [16] and Macdonald et.al [17]. to model the primary coolant chemistry of BWRs, with the rate constant of Reaction (30) being used as the "calibrating" parameter.

| No. | Rate Constant (L/Mol-sec) | Activation Energy (Kcal/Mol) | Chemical Reactions  |
|-----|---------------------------|------------------------------|---|
| 1   | 1.60E+01                  | 3                            | $\mathbf{e}_{aq}^{-}$ + $\mathrm{H}_2\mathrm{O}$ = $\mathrm{H}$ + $\mathrm{OH}^{-}$ |
| 2   | 2.40E+10                  | 3                            | $e_{aq}^-$ + H $^+$ = H   |
| 3   | 2.40E+10                  | 3                            | $e_{aq}^-$ + OH = OH  |
| 4   | 1.30E+10                  | 3                            | $e_{aq}^- + H_2O_2 = OH + OH$   |
| 5   | 1.00E+10                  | 3                            | $H + H = H_2$   |
| 6   | 2.00E+10                  | 3                            | TRUE  |
| 7   | 1.90E+10                  | 3                            | $e_{aq}^{-} + 0_2 = 0_2^{-}$  |
| 8   | 0                         | 0                            | $2 e_{aq}^{-} + 2H_2O = 20H^2 + H_2$  |
| 9   | 4.50E+09                  | 3                            | $OH + OH = H_2O_2$  |

| 10 | 1.20E+10 | 3    | $OH + HO_2 = H_2O + O_2$  |  |
|----|----------|------|---|--|
| 11 | 1.20E+10 | 3    | $OH + O_2^- = OH^- + O_2$   |  |
| 12 | 2.00E+07 | 3    | $OH^{\cdot} + H = \mathcal{C}_{aq}^{-} + H_{2}O$                  |  |
| 13 | 4.50E+08 | 3    | $e_{aq}^{-}$ + H + H <sub>2</sub> O = OH + H <sub>2</sub>         |  |
| 14 | 0        | 0    | $e_{aq}^{-}$ + H0 <sub>2</sub> + H <sub>2</sub> 0 = 0H + 20H      |  |
| 15 | 1.44E+11 | 3    | $H^+ + OH^- = H_2O$   |  |
| 16 | 2.60E-05 | 3    | H <sub>2</sub> O = H+ + OH  |  |
| 17 | 2.00E+10 | 3    | $H + OH = H_2O$   |  |
| 18 | 3.40E+07 | 4.6  | OH + H <sub>2</sub> = H + H <sub>2</sub> O                        |  |
| 19 | 2.70E+07 | 3.4  | $OH + H_2O_2 = H_2O + HO_2$                                       |  |
| 20 | 4.40E+07 | 4.5  | $H + H_2O_2 = OH + H_2O$  |  |
| 21 | 1.90E+10 | 3    | $H + O_2 = HO_2$  |  |
| 22 | 8.00E+05 | 3    | HO <sub>2</sub> = O <sub>2</sub> -+ H+                            |  |
| 23 | 5.00E+10 | 3    | $O_2^- + H^+ = HO_2$  |  |
| 24 | 2.70E+06 | 4.5  | 2HO <sub>2</sub> = H <sub>2</sub> O <sub>2</sub> + O <sub>2</sub> |  |
| 25 | 0        | 0    | $20_{2}^{-} + 2H_{2}O = H_{2}O_{2} + O_{2} + 2OH$                 |  |
| 26 | 2.00E+10 | 3    | $H + HO_2 = H_2O_2$   |  |
| 27 | 2.00E+10 | 3    | $H + O_2^- = HO_2$  |  |
| 28 | 0        | 0    | $e_{aq}^{-} + O_{2}^{-} + H_{2}O = HO_{2}^{-} + OH$               |  |
| 29 | 1.80E+08 | 4.5  | $OH^- + H_2O_2 = HO_2^- + H_2O$                                   |  |
| 30 | 2.00E-06 | 14.8 | $2H_2O_2 = 2H_2O + O_2$   |  |
| 31 | 1.04E-04 | 3    | $H + H_2O = H_2 + OH$   |  |
| 32 | 1.02E+04 | 3    | $H_2O + HO_2^- = H_2O_2 + OH$                                     |  |
| 33 | 1.50E+07 | 4.5  | $HO_2 + O_2 = O_2 + HO_2$   |  |
| 34 | 7.70E-04 | 7.3  | H <sub>2</sub> O <sub>2</sub> = 20H                               |  |

Table 3: G-values (no./100 eV) for low LET radiation (γ radiation) at different temperatures by using relations from [20].

| Species                       | 25 °C | 70 °C | 100 °C | 130 °C | 150 °C | 250 °C |
|-------------------------------|-------|-------|--------|--------|--------|--------|
| $e_{aq}^-$                    | 2.75  | 2.961 | 3.101  | 3.232  | 3.311  | 3.513  |
| Н                             | 0.604 | 0.67  | 0.71   | 0.756  | 0.796  | 1.182  |
| ОН                            | 2.807 | 3.275 | 3.573  | 3.868  | 4.065  | 5.122  |
| H+                            | 2.75  | 2.961 | 3.101  | 3.232  | 3.311  | 3.513  |
| OH.                           | 0     | 0     | 0      | 0      | 0      | 0      |
| 02                            | 0     | 0     | 0      | 0      | 0      | 0      |
| 02-                           | 0     | 0     | 0      | 0      | 0      | 0      |
| H <sub>2</sub>                | 0.438 | 0.461 | 0.472  | 0.481  | 0.489  | 0.561  |
| H <sub>2</sub> O <sub>2</sub> | 0.711 | 0.639 | 0.59   | 0.541  | 0.509  | 0.347  |
| HO <sub>2</sub> -             | 0     | 0     | 0      | 0      | 0      | 0      |
| HO <sub>2</sub>               | 0     | 0     | 0      | 0      | 0      | 0      |

Numerous models have been proposed for the radiolysis of water and one such model that has been employed to model the coolants in water-cooled nuclear power reactors is presented in Table 2. This particular model has been cleansed of unacceptable,

non-elementary reactions, such  $e_{aq}^- + e_{aq}^- + 2H_2O \rightarrow H_2 + 2OH^-$ , which may be decomposed into two elementary reactions  $\left[2\left(e_{aq}^- + H_2O \rightarrow H_2 + OH^-\right), H + H \rightarrow H_2\right]$  that were already in-

cluded in the original model. Furthermore, such a reaction postulates reaction between species of like charge, which are discounted based on coulombic repulsion. As shown, the simplest way of removing these reactions from established codes is to simply set their rate constants equal to zero.

All models comprise a source term of primary radiolysis products that are envisioned to form within the spurs (track of the ionizing particle) in <  $10^{-12}$  s [14], including  $e_{aq}^-$  -,  $H_2O^+$  (H+), H, OH, OH-, H<sub>2</sub>, and H<sub>2</sub>O<sub>2</sub>, and a set of reactions between these species at longer times as they diffuse from the spurs to produce the non-primary radiolysis products (Table 2). Accordingly to that reactions are not counted twice, the radiolytic yiels (G vales, # of a given species that are produced per 100 eV of energy absorbed by the water) must be primary yields. A set of G-values for low linear energy transfer radiation like y-photon radiation are summarized in Table 3. Note that the yields for secondary species have been set equal to zero in an attempt to correct for the fact that they are not primary products that are generated within the  $< 10^{-12}$  s timescale. However, there is no assurance that the G-values are in fact primary yields, and it is known that the yield of  $e_{aq}^-$  and OH, for example, decreases sharply with time over a few nanoseconds range [18,19]. Accordingly, only yields that are determined using sub-nanosecond pulse techniques are arguably "primary" yields and, perhaps, the true primary yields are unmeasurable using currently available techniques. Nevertheless, such models have been successfully used to calculate properties, such as the electrochemical corrosion potential (ECP) of alloys in nuclear reactor coolant circuits, for example.

The interaction of ionizing radiation with biological systems has been studied extensively and a comprehensive review has been published by Reisz, et.al. [21]. Briefly, the interactions can be classified into direct interactions and indirect interactions. In direct interactions, the IR interacts directly with the biologically active molecule (e.g., DNA, amino acids, etc) primarily via scissoring of C-S-H, C-O-H, and C-N-H bonds to produce free radical and other species, while the indirect interactions result from the radiolysis of intercellular water to produce highly reactive products that then react with the biologically active components of the system, as briefly described above. Specific issues in the IR preservation of food have been reviewed by Catanescu and Tofana [22] while the international standards are articulated in Ref 10. The dose rates employed in γ-photon (60Co) irradiation range up to 0.5 Gy/s, depending upon the food and the microorganisms present, although many dose rates are apparently much lower (e.g., 1.12x10<sup>-3</sup> Gy/s [23]). In any event, at a dose rate of 0.1 Gy/s it takes 500,000 s (139hrs) to accumulate a 50 kGy dose, which is the upper end of the range employed in the food industry.

A cartoon depicting the effect of IR on biological systems is presented in Figure 2 [21]. As shown, the indirect effect involves the attack by radiolysis species on lipids, DNA, RNA, and proteins, amongst other entities, primarily by the oxidizing species, such as OH and  $\rm H_2O_2$ , which are powerful oxidizing agents (Table 1) and  $\rm O_2^-$  that is a mild reducing agent but which produces a strong oxidizing agent ( $\rm O_2$ ). Of course, the actual processes are much more complex than those depicted in Figure 2, but the general idea is conveyed.

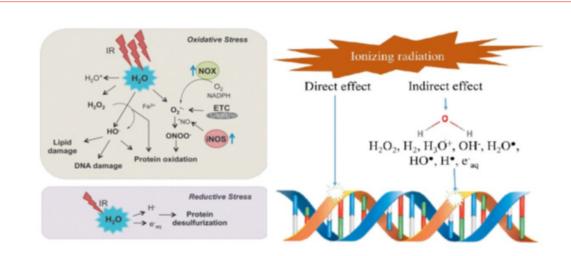


Figure 2: (a) Interaction of ionizing radiation (IR) with biological systems. (From Reisz, et.al. [21]). (b) Direct vs indirect effects of IR in biological systems (From Munir and Federighi [14]).

Many biochemical processes are redox sensitive [21,22,25]. From an electrochemical viewpoint, the driving force for redox processes is the redox potential ( $E_{\rm redox}$ ), which is a mixed potential that is established by a balance of the partial anodic and cathodic processes at an interface such that the net current is zero. Often, this potential is erroneously identified with an equilibrium potential as calculated using the Nernst equation [26-33]. Instead,  $E_{\rm redox}$  can

be calculated using a mixed potential model (MPM) [34] that was originally developed for calculating the electrochemical corrosion potential (ECP) of steels in water-cooled nuclear reactor coolant circuits. In that application,  $\mathbf{E}_{\text{redox}}$  provides a measure of the driving force for corrosion reactions, such as metal electrode solution and passivation to occur. In fact, to a good approximation,  $\mathbf{E}_{\text{redox}}$  may be thought of as being equivalent to the potential applied between

the tip of the Luggin reference electrode probe and the metal in a classical electrochemical potentiostat experiment. However, it is important to recognize the difference between  $E_{\mbox{\tiny redox}}$  and the ECP (corrosion potential) as the former refers to a system for which the anodic current due to the dissolution of the substrate is zero while the latter corresponds to a system in which the substrate is undergoing electro dissolution (corrosion). Since redox potentials are commonly measured with Pt or Au indicator electrodes, this non-reacting substrate constraint is practically satisfied especially because both substrates display catalytic activity toward charge transfer (REDOX) reactions, such as H<sub>2</sub>/H<sup>+</sup> (HER), O<sub>2</sub>/H<sub>2</sub>O (OER), and Fe2+/Fe3+, where HER and OER indicate the hydrogen and oxygen electrode reactions, respectively. Accordingly, the exchange current densities for the partial reactions are very much higher than for the same reactions on stainless steels, for example, and these affect the value of the measured potential. An example of the calculated Eredox vs [0,] at 25 °C in pure water containing 1 ppm

of hydrogen is shown in Figure 3. Also plotted are the equilibrium potentials for the oxygen electrode reaction (OER) and the hydrogen electrode reaction (HER) for comparison with Eredox. As seen, Eredox displays a sigmoidal variation with composition ([H $_2$ ]/[O $_2$ ]) and approaches  $E^e_{HER}$  and  $E^e_{HER}$  asymptotically at limitingly low and high [O $_2$ ], respectively. Note that  $E^e_{HER}$  is independent of the composition because, in this calculation, the hydrogen content is fixed (1 ppm) while  $E^e_{HER}$  becomes more positive as the oxygen content of the solution increases, as specified by the Nernst equation. Furthermore, the reader will note the large difference between  $E_{\rm redox}$  and  $E^e_{HER}$  and  $E^e_{HER}$  except at the composition extremes where the redox potential is determined by the equilibrium potentials of one of the redox reactions. This accounts for the frequently observed disparity between the measured  $E_{\rm redox}$  and that calculated by using the Nernst equation, which is a common but erroneous practice in biology.

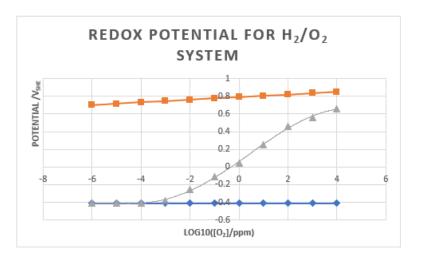


Figure 3: Plot of  $E_{redox}$  vs Log10[O2/ppm] (grey line) at 25 °C in pure water containing 1 ppm of hydrogen. Also plotted are the equilibrium potentials for the HER (blue line) and the OER (red line). [H2] = 1 ppm.

Another issue that complicates our ability to calculate  $E_{redox}$  is the fact that no exchange current density or transfer coefficient data are available for the energetic radiolysis species, such  $e_{aq}^-$  , H, OH,  $HO_2$ ,  $HO_2$ ,  $O_2$ ,  $O_2$ ,  $O_2$ ,  $O_3$ , and O. However, a general rule of thumb that has been gleaned by extensive modelling and experimental work in this area is that the contribution that any species makes to the redox potential (or to ECP) is roughly proportional to its concentration and, because the concentrations of these radiolytic species are orders of magnitude lower than those of O2, H2O2, and H<sub>2</sub>, their impact is minor (see below). Thus, the redox potential in irradiated systems is dominated by  $O_2$ ,  $H_2O_2$ , and  $H_2$  but that does not imply that the more energetic species are not important in redox reactions since, while their concentrations are low, they are generally of much higher reactivity than are the dominant species. Furthermore, because of their low concentration, the partial currents due to the redox reactions involving these highly active species are mass transfer limited and hence insensitive to the kinetic parameters. The most effective way of addressing this issue is to define new concentrations as  $[H_2^*] = [H_2] + 0.5[H] + 0.5[\textit{e}_{aq}^-] + [O_2^-]$ ,  $[H_2O_2^*] = [H_2O_2] + 0.5[OH] + [HO_2^-] + [O_2^{-2}^-] + [HO_2]$  and  $[O_2^*] = [O_2] + 0.5[O]$ . One of the authors (DDM) has developed a code, RAD\_REDOX, for calculating the redox potential in irradiated systems from similar codes that he developed for estimating ECP in nuclear reactor coolant circuits [32] and these calculations are described later in this review.

Using the methods outlined above, we show in Figure 4 the calculated evolution of the composition of irradiated water at  $25^{\circ}$ C for a  $\gamma$ -photon dose rate of 1 mGy/s, corresponding to the dose rate employed by Britto et.al. [24] in their experimental work on the effect of radiation dose rate on psychrotrophic bacteria, thiobarbituric acid reactive substances, and sensory characteristics of mechanically deboned chicken meat. The calculations are for a closed, liquid phase system (no gas phase) and it is seen that the system comes to a steady state within about 10s.

The concentrations of the species then remain invariant from about 10 ms up to the maximum simulation time of 104 s (2.8hrs). Radiolysis, at least at this dose rate, is predicted to have little impact on pH. The dominant species is predicted to be H2, followed closely by  ${\rm H_2O_2} > {\rm O_2} > {\rm OH} \approx {\rm O_2}^- > {\rm H} > {\it e}_{aq}^- \approx {\rm HO_2} > {\rm HO_2}^-$  in abundance. The redefined concentrations,  ${\rm [H_2^*]} = {\rm [H_2]} + 0.5{\rm [H]} + 0.5{\rm [}$   ${\it e}_{aq}^-$  ] +  ${\rm [O_2^*]}$  +  ${\rm [O_2^*]}$  ,  ${\rm [H_2O_2^*]}$  =  ${\rm [H_2O_2]}$  + 0.5[OH] and  ${\rm [O_2^*]}$  =  ${\rm [O_2]}$  + 0.5[O] +  ${\rm [O_2^*]}$  are calculated to be 2.546x10<sup>-7</sup> M (0.0005137 ppm), 1.327x10<sup>-7</sup> M (0.004514 ppm), and 5.570x10<sup>-8</sup> M (0.001782 ppm),

respectively. These concentrations result in  $E_{redox} = 0.0779 \, V_{she}$ . Had we used  $[H_2]$ ,  $[O_2]$ , and  $[H_2O_2]$  as inputs instead of  $[H_2^*]$ ,  $[O_2^*]$ , and  $[H_2O_2^*]$ ,  $E_{redox} = 0.0775$ , only  $0.0004 \, V_{she}$  lower than if the contributions of the highly energetic radiolysis products  $(H, \mathcal{C}_{aq}^-, O_2^-, \text{ and } OH)$  are ignored. This demonstrates the previous conclusion that only the most dominant redox species  $(H_2, O_2, \text{ and } H_2O_2, \text{ in this case, see Figure 4})$  need be considered in calculating the redox potential (and the ECP) [34].

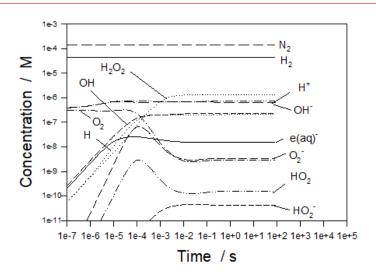


Figure 4: Predicted evolution of the radiolysis products of water irradiated with γ-photons at a dose rate of 1mGy/s at 25°c. Metal contaminants: 10 ppb  $Fe^{2+}$  and 10 ppb  $Cu^+$ .

From the value of the calculated redox potential (0. 0779  $V_{she}$ ), irradiation of the water with  $\gamma$ -photons at a low dose rate of 1mGy/s at 25 °Crenders the system moderately oxidizing, which is attributed primarily to the formation of  $H_2O_2$ . By calculating  $E_{redox}$ 

for various combinations of  $H_2^*$ ,  $O_2^*$ , and  $H_2O_2^*$ , we find that the two dominant species in determining the redox potential are  $H_2^*$  and  $H_2O_2^*$  (Table 4).

Table 4: Sensitivity of Eredox to  $[H_2^*]$ ,  $[O_2^*]$ , and  $[H_2O_2^*]$  for irradiation of water with γ-photons at a dose rate of 1mGy/s at 25°c.

| Case # \ Specie                     | [H <sub>2</sub> *] /ppm | [O <sub>2</sub> *] /ppm | [H <sub>2</sub> O <sub>2</sub> *] | $\mathbf{E}_{\mathrm{redox}}/\mathbf{V}_{\mathrm{she}}$ |
|-------------------------------------|-------------------------|-------------------------|-----------------------------------|---|
| Base                                | 0.0005132               | 0.0017824               | 0.004514                          | 0.0779  |
| Low H <sub>2</sub> *                | 1x10 <sup>-10</sup>     | 0.0017824               | 0.004514                          | 0.8629  |
| Low H <sub>2</sub> O <sub>2</sub> * | 0.0005132               | 0.0017824               | 1x10 <sup>-10</sup>               | -0.0085   |
| Low O <sub>2</sub> *                | 0.0005132               | 1x10 <sup>-10</sup>     | 0.004514                          | 0.0069  |

Thus, in the absence of  $\rm H_2$  (Low  $\rm H_2^*$  case),  $\rm E_{redox}$  is very high (0.8629  $\rm V_{she}$ ), which is attributed primarily to  $\rm H_2O_2^*$ , demonstrating the powerful impact of the reducing species ( $\rm H_2^*$ ) in the system. The impact of  $\rm H_2O_2^*$  is further illustrated by the Low  $\rm H_2O_2^*$  case where the redox potential is predicted to fall to -0.0085  $\rm V_{she}$ . Low  $\rm O_2^*$  results in a modest increase in  $\rm E_{redox}$  from the lor  $\rm H_2O_2^*$  case but the dominant roles played by  $\rm H_2^*$  and  $\rm H_2O_2^*$  are still evident.

In closing this discussion, we emphasize that the redox potential is only one (but an important one) of the factors that determine reactivity in irradiated aqueous system, such as food. Other factors controlling the rates of reactions between water radiolysis products and reactive moieties in substrates include the electron densi-

ty at the reaction site and the activation energy.

#### **Radiation Processing**

Radiation is a unique source of energy that can inaugurate chemical reactions at any temperature, including ambient, under any pressure, in any phase (gas, liquid or solid), without the use of catalysts. However, the temperature rise factor should be considered when the material is processed with high-dose irradiation [35].

#### **Materials Modification**

Polymers are quite often irradiated for modification or are the main component of radiation-sterilized medical products. There-

fore, the changes in their structure may be beneficial or undesirable. These facts are the reason why R&D concerning these materials is broad and most developments are foreseen in this area [36]. The application of radiation for the modification of synthetic materials, mostly curing and cross-linking, is a well-established technology. New applications are constantly being sought. Nevertheless, the procedures observed in the early years of technology implementation, when materials were irradiated just to see, should be avoided in this stage where knowledge of the phenomena is much more complete. On the contrary, new analytical techniques like NMR, FTIR, synchrotron radiation, etc. open new horizons for an understanding of processes such as cross-linking and scission in polymers in much more substantial detail than previously possible. New, unexpected discoveries are frequently being made [37,38]. In addition, very powerful accelerators with 700 kW output have made X-ray conversion a practical alternative to the historic use of radioisotopes, mainly cobalt-60, for applications such as medical device sterilization. New electron beam end-uses are emerging, such as the development of nanocomposites and nano-gels and the use of EB processing to facilitate biofuel production. These present new opportunities for future research and development. [39]. Radiation as a tool for product engineering, like sensors or membranes, is still not a fully exhausted area of application [40]. Gamma or e-beam irradiation of polymers allows good control of the chemistry at the micro/nano-scale with minimal recourse to toxic reactants and solvents. Another potential advantage is to obtain simultaneous sterilization when the absorbed doses are within the sterilization dose range [41].

The other field of possible applications is the processing of natural polymers. Processing of cellulosic materials for the pharmaceutical and cosmetic industry has already been implemented. Some chitosan derivatives are manufactured as well. New sorbent for various applications is also being developed [42]. The potential of combining radiation effects with nano-materials has been recognized from the very early stages of nano-science research. In the many uses of nano-structures, and nano-particles in particular, from catalysis, bio-sensing, nano-electronics, and magnetic applications including separations, mechano-chemical conversion, and to molecular computing, radiation can play a significant role. The use of radiation, UV beam, electron beam, or focused ion beam is clearly central to the fabrication of the nanostructured systems [43].

#### **Sterilization**

Radiation sterilization is a well-established technique and most of the strong and weak points were addressed. The observed tendency concerns the multi-technique offer of the service [44].

#### Irradiation of food

Food irradiation is a process that exposes food to ionizing radiation which is a form of electromagnetic energy. This involves the exposure of bulk or prepackaged food to ionizing radiations sourced from either accelerator that produce controlled amounts of X-rays, high-energy electron beams ( $\beta$  particles), or gamma ( $\gamma$ ) rays from radioactive isotopes of cobalt ( $^{60}$ Co) or cesium ( $^{137}$ Cs) in a controlled environment [11]. All three types of radiation result in the

excitation of the atoms in the target food product, but the energy is limited and does not interact with the nuclei to produce radioactive species. However, ionizing radiation has a pernicious impact on microorganisms in food if applied at a specific dose. The energy from ionizing radiation inactivates microorganisms by damaging the critical element in the cell, mostly the chromosomal DNA [45]. The damage prevents multiplication and arbitrarily terminates most cell functions. The damage to the DNA results from a direct collision between radiation energy and genetic material. As a result of the interaction between an adjacent molecule which in most situations is a water molecule and the radiation energy which then reacts with the DNA [46]. Though the use of ionizing radiation for food preservation began in the early 1920s, during the 1950s-1960s, the US Army conducted research into low-dose and high-dose irradiation of military rations. At the same time, similar studies of these experiments prompted in other countries and the interest in food irradiation have grown ever since. With proper application, irradiation can be an effective means of eliminating and reducing microbial and insect infestations along with the foodborne diseases they induce, thus improving the safety of many foods as well as extending shelf life [47].

#### **Safety for Consumption of Irradiated Foods**

The safety of irradiated foods for human consumption has been questioned because ionizing radiation can lead to chemical changes. The wholesomeness of irradiated foods has, therefore, been the subject of considerable and officially accepted by international organizations due to its effectiveness in food, and economic benefits [47]. Overall, the World Health Organization (WHO), the International Atomic Energy Agency (IAEA), and the Food and Agriculture Organization (FAO) have a less restrictive approach than the European Union. In 1981, 10 kGy was set as the maximum dose considered to be safe and wholesome by the Join Expert FAO/IAEA/WHO Committee on the wholesomeness of irradiated food [48]. According to a nutritional or microbiological viewpoint without substantial detrimental effects, the energy transferred up to 10 kGy did not show toxicological hazard. In 1997, a Join FAO/IAEA/WHO Study Group terminated that food irradiated to any dose appropriate to achieve the intended technological objective is both safe to consume and nutritionally adequate and, further, that no upper dose limit needs to be imposed [49]. In 2003, the Scientific Committee on Food of the European Commission did not accept the suggested removal of the upper limit of 10 kGy due to the very limited toxicological studies that had been carried out with irradiated food with doses above 10 kGy. The same Committee was of the viewpoint that it is appropriate to specify a maximum dose for the treatment by ionizing radiation of certain food products and that irradiated foodstuffs should continue to be evaluated individually considering the technological need and food safety [50]. A Codex general standard for irradiated foods and a recommended international code of practice for radiation processing of food has been developed by this time [51]. Therefore, the treatment carried out is authenticated by scientific evidence [52].

The assessment of the safety of radiation-treated foods involves:

- 1. Radioactivity safety
- 2. Toxicological safety
- 3. Microbiological safety
- 4. Impairment of sensory quality
- 5. Nutritional adequacy

Food irradiation is the most extensively studied processing method for enhancing the safety and quality of food. All scientific results regarding the safety and wholesomeness of irradiated foods have been considered by the experts of the World Health Organization (WHO), Food and Agricultural Organization (FAO), and the International Atomic Energy Agency (IAEA) and simultaneously by the governments of different countries [53,54].

#### Radioactivity safety

In a food irradiation facility, a set speed determines the amount of dose or energy absorbed by a food product. In a controlled environment the food itself never comes into direct contact with the source of radiation. In principle, all foods are radioactive to some degree as a result of exposure to natural background radiation. As no neutrons are emitted by 60Co, the food treated with the gamma rays does not become radioactive. As a result, no nuclear changes are produced in the nuclei of food molecules. There is no scientific evidence that irradiated food will contain levels of radioactivity higher than those in non-irradiated food. Researchers concluded in another study that energy beams emitted from food irradiated by doses below 60kGy, with gamma rays from c, 60Co and 137Cs were less than 5 MeV in energy and can be considered insignificant. Nevertheless, 60Co is the preferred source of radiation for food [53-55].

#### **Toxicological safety**

Irradiation of food under certain conditions is safe and that irradiation of any food commodity up to an overall average dose of 10kGy presents no toxicological hazard, hence toxicological testing of foods so treated is no longer required, which was reviewed by some independent reviews of the scientific toxicological studies on irradiated food [56]. Irradiation to high doses is essentially analogous to conventional thermal processing such as canning of low-acid foods, in that it eliminates biological hazards from foodstuffs intended for human consumption but does not result in the formation of physical or chemical entities that could constitute a hazard, which was studied by the Joint FAO/IAEA/WHO Study Group on High Dose Irradiation [49].

The formation of a series of radiation-induced cyclic ketones, namely, the 2-alkylcyclobutanones (2-ACBs), is the most important safety issue from the toxicological safety point of view concerning irradiated foods that are known as unique radiolytic products [57]. Several studies have been conducted over the past decade to determine the toxicology and mutagenic effects associated with the consumption of 2-ACBs. These studies have reported that 2-ACBs exhibit no mutagenic or genotoxic effects on mammalian cell lines at low concentrations. However, the consumption of these chemicals at higher doses has resulted in cytotoxicity and damage to the genetic material in rat and human colon cells [58]. Research-

ers have undertaken, mainly short-term investigations to study the toxicological potential of 2alkylcyclobutanones with respect to the health risks of the consumption of fat-containing irradiated foods. Recent results detected the toxic and colon carcinogenesis-promoting effects of 2-alkylcyclobutanones [59]. A team of French and German scientists carried out a detailed assessment of the toxicity of several 2alkylcyclobutanones. The studies reported toxic, genotoxic, and tumor-promoting activity of 2-dodecylcyclobutanone, 2-tetradecylcyclobutanone, and 2-(tetradic-5'-enyl)-cyclobutanone. On the other hand, it was identified to characterize the potential risk, hazards that need to be identified, the exposure, the exact dose-response relationship, and especially the kinetics and metabolism of 2-ACBs in the living organism should be expatriated. Based on recent data, it seems not appropriate to draw a conclusion concerning the risk associated with human consumption of raw irradiated fat-containing foods were concluded. The researchers deemed all these studies effective in gaining insight into the mechanisms of the toxic effects [60].

Reviewers observed the adverse effects and noted that based on these results it was not appropriate to assess the risk to human health associated with the consumption of 2-alkylcyclobutanones present in irradiated fatty foods. It was reported that toxic effects induced by certain 2-alkylcyclobutanones [61]. To evaluate the capacity of 2-DCB to induce mutations, E. coli tryptophan reverse mutation assay was used. *E. coli* tester strains WP2 (pkM101) and WP2 uvrA (pKM101), with and without exogenous metabolic activation, were exposed to 0, 0.05, 0.1, 0.5, and 1 mg/well 2DCB using the mini-screen version of the analysis. 2-DCB did not induce mutations in the E. coli tryptophan reverse mutation analysis which was the output of the analysis. These results are in agreement with negative results obtained in short-term and long-term genetic toxicology tests of irradiated food products [62].

In a study, the Salmonella mutagenicity test was conducted and the yeast DEL assay were used to evaluate the genotoxic potential of 2-DCB. The researchers concluded the absence of genotoxicity observed using purified 2-DCB agrees with the lack of genotoxic and teratogenic activity observed in previously conducted multigenerational feeding studies of laboratory animals (rats, mice, and rabbits) that used radiation-sterilized poultry containing 2DCB as a unique radiolytic product [63]. The amount of 2dodecylcyclobutanone in rat feces and adipose tissue was assessed to determine its metabolism. Up to 11% of the total administered 2-DCB was recovered intact in feces and adipose tissue. This finding indicates that most 2-DCB is metabolized and depleted quickly from the body or stored at sites other than adipose tissue [64].

Researchers found that the cytogenetic effects of 2-dodecyl-cyclobutanone in healthy human colon epithelial cells and in cells representing preneoplastic colon adenoma were clearly genotoxic. It was revealed that this compound may be regarded as a possible risk factor for the initiation and progression processes in colon carcinogenesis [65].

In another research, researchers explored the cytotoxic and genotoxic potentials of various highly pure synthetic 2alkylcyclobutanones in the lines of bacteria and human cell. They were observed

pronounced cytotoxicity in bacteria, however, no mutagenic activity has been revealed by the Ames test in Salmonella strains TA 97, TA 98, and TA 100. Genotoxicity was demonstrated in mammalian cells by the induction of DNA base lesions detected by the alkaline unwinding procedure. The researchers claimed that the cytotoxicity and genotoxicity were dependent on the fatty acid profile of the triacylglycerol composition [66]. The European Commission's Scientific Committee on Food conducted a review of various issues concerning irradiated foods. The committee considered that there is no sufficient evidence to pronounce that foods irradiated above 10 kGy are safe for human consumption [67]. Based on the published studies on the mutagenicity of irradiated foods, some scientists consider that there are no adequate long-term safety studies on the assessment of the overall health hazards posed by 2-ACBs, including different sensitivities to 2-ACBs among the human consumer population. Furthermore, additional in vitro and in vivo tests with regard to the tumor-promoting activities of unique radiolytic products should be conducted [68-70]. The lack of adequate information on the effect of long-term consumption of irradiated foods on human health and on the long-term health effects of eating a diet based on irradiated foods is still considered to be a problem, and thus, precautionary principles should be applied until such information are available. In this regard, WHO encourages further research in accordance with scientifically adopted protocols for assessing food safety to help resolve any remaining uncertainties regarding the toxicity or carcinogenicity of 2-alkylcyclobutanones. While the new evidence indicates potential public health risks, the WHO reiterates its previously stated willingness to reopen the risk assessment of irradiated foods [71].

# Microbiological safety

Food irradiation is an efficient processing method for destroying fungi, bacteria, viruses, or insects that may be present in food. Specifically, to reduce or eliminate pathogenic and spoilage microorganisms including L. monocytogenes, Salmonella and Staphylococcus aureus, E. coli, Campylobacter, Yersinia enterocolitica, yeast, and mold in meat products; irradiation is an effectual way. Therefore, to prevent possible public health hazards, irradiation can increase the hygienic quality of raw meats. Researchers observed that the differential sensitivity of microorganisms to ionizing radiation depends on the irradiation dose, the kind of species, the size of the organisms, different microenvironments, the presence of some chemical compounds such as proteins, sulfites, nitriles, sulfhydryl compounds, compounds containing the SH group, the presence or absence of oxygen, and the physical state of the food during irradiation etc. Spoilage and Gram-negative bacteria such as Enterobacteriaceae and Pseudomonas spp. can be inactivated by low and medium doses of radiation within 7 kGy as they are very sensitive to irradiation [67]. Irradiation doses below 1 kGy can be applied to prevent sprouting, slow the ripening process, and extend the fresh life of fruits and vegetables. Irradiation doses in the range of 1 to 10 kGy can be applied in the eradication or elimination of foodborne pathogens, reducing food spoilage. Dose rates are usually in the range of 20-30 kGy, and they are associated with the radiation sterilization of foods which eliminates some disease-causing viruses [72].

In the current study, three minimally processed vegetables, i.e., tomato, bell pepper, and white onion were studied for their post-processing microbiological profile; and to assess the effect of gamma radiation treatment (0.5-5 kGy) to maintain hygiene during extended storage at low temperature. Except for tomato, which did not contain any detectable Presumptive Coliform or Yeast and Mold on day 0, the other two cut vegetables were found to have higher load of microbes. In the radiation treated (2 kGy) vegetables, no Presumptive Coliform (PC) was detected; even after 20 days of storage; on the contrary, in the untreated bell pepper, tomato and onion samples PC counts were found [73]. In another study, the effect of ionizing radiation on the microbiological quality on minimally processed carrot and lettuce was studied. Minimally processed carrot and lettuce were analysed for total viable count, total coliform count and pathogenic organisms. The samples collected were treated and analysed for a 15day period. The predominant pathogenic organisms identified were Bacillus cereus, Cronobacter sakazakii, Staphylococcus aureus, and Klebsiella spp. It was concluded that 2 kGy was most effective for medium dose treatment of minimally processed carrot and lettuce [74].

Radiation treatment can play an important role in reducing post-harvest losses and use of chemical fumigants. Due to negative impact on human health and the environment, chemical fumigation using ethylene di-bromide, methyl bromide, and ethyl oxide for the control of insects has been banned. Insects cause enormous damage to food crops during their production in the field and after harvesting, especially during storage. They are relatively sensitive to irradiation; therefore, the lowest effective dose is applied. Irradiation with a dose of 1 kGy is recommended for the insect disinfection of cereals and oilseeds and 5 kGy for the reduction of microbial load [75]. In a study it was observed the effect of y-irradiation (0kGy, 5 kGy, and 10 kGy) on the microbial inactivation of selected pathogens (Bacillus cereus, Listeria monocytogenes, Staphylococcus aureus, Escherichia coli 0157: H7, and Salmonella typhimurium) of a liquid formulation and powdered infant formula. It was observed that y-irradiation treatment was effective in both types of samples to inactivate the pathogens and that their viability reduced exponentially with increasing the dosage. E. coli and B. cereus (under vegetative or spore forms) were observed more sensitive under frozen conditions than in powder. The radio-sensitivity was respectively 2 and 1.8 times for E. coli and B. cereus under vegetative form. B. cereus was found to be 2.6 times more sensitive under spore form in liquid formulation form as compared to powder form. Under powder and liquid formulation, S. Typhimurium, L. monocytogenes and B. cereus (vegetative and spore form) were the most resistant to irradiation treatment. B. cereus under the spore condition was found the most resistant bacteria. A dose of 14.2 and 36.8 kGy was needed to reduce by 6 Log (CFU/ml) B. cereus under the forms of liquid formulation and powder, respectively [76].

Egg products are another important item of food products under preservation technology. The effect of gamma irradiation on the presence of microorganisms in egg powder was investigated. Egg powder samples were exposed to several doses of irradiation: 0, 5, 10 and 15kGy and stored for up to 12 months at ambient temperature (25 °C). Results indicated that the total viable count (TVC), to-

tal coliform counts (TCC) and mold and yeast counts (MYC) in un-irradiated (control) samples of egg powder were higher than the maximum limits. Application of the higher doses (10 and 15 kGy) decreased the TVC, TCC and MYC of the egg powder samples to less than  $1 \log_{10}$  cfu g<sup>-1</sup> and the counts remained almost constant during storage for 12 months. D10 values for Escherichia coli and Salmonella typhimurium were 0.714 and 0.278 kGy, respectively. Gamma irradiation treatment could be chosen on the basis of preliminary microbiological tests including TVC, TCC and MYC and help improve the hygienic quality by killing and reducing the microorganisms that might be present inside of egg powder to meet national and international standards [77]. Researchers have evaluated the effect of irradiation followed by thermal treatment on the survival of the six Salmonella serovars inoculated into the liquid whole egg (LWE). It has been noted that irradiation in combination with heat treatment could be a promising pasteurization process to obtain Salmonella free stable LWE, reducing the effect of treatment on the quality of LWE. Furthermore, the existence of observed synergistic lethal effects for radiation doses lower than 1.5 kGy followed by heat treatment at 55°C and 57°C allowed thermal treatment times is reduced from 86% to 30% [78]. Properly processed irradiated foods are wholesome. To obtain the best results from food irradiation, only products of the highest quality should be treated. Irradiated products will not remove any off-odors or flavors already present nor will it improve the appearance, if food is rotten prior to irradiation. It cannot make 'spoiled' food 'fresh'. Food irradiation technology used for microbial inactivation is not intended to serve as a substitute for good food hygienic practices, and strict observation of Good Manufacturing Practices is required in order to provide the safety and quality of food [79].

#### Impairment of sensory and nutritional quality

Without adversely affecting the nutritional and sensory quality of food products, food irradiation is an efficient processing method. Impairment of sensory quality is not at all a relevant concern. Such food is just not consumed; the damage is exclusively with the producers. Nutritional quality is much more difficult to judge; however, reviewers have summed up the investigations on the nutritional adequacy of irradiated foods under diverse conditions in several reviews [80]. Generally, ionizing radiation does not affect macronutrients such as carbohydrates, proteins, and fats significantly, even at doses over 10 kGy. It is noteworthy that the effects of thermal energy and ionizing radiation on foods are similar. Animal and human studies have shown no effects of the consumption of a variety of irradiated foods on the metabolic power of the macronutrients [81].

The nutritional quality of essential amino acids, essential fatty acids, minerals, and trace elements undergo little changes. The impact of irradiation on the food's nutritional quality depends on the kind of food, radiation dose, the packaging atmosphere, the temperature during irradiation processing and post-irradiation storage, the presence or absence of oxygen, and the storage time. In principle, the radiolytic degradation of food components such as proteins, fats, and carbohydrates increase with increasing radiation dose. Foods with high lipid content (52-70%) and especially with high unsaturated fatty acid content are highly sensitive to irradiation as they form free radicals during irradiation, which enhanc-

es lipid oxidation [82,83]. The sensitivity to oxidation in radiation treatment increases with the increase of the degree of unsaturation. Oxidation products can be produced that can destroy nutrients and affect digestibility, if the food is stored in the air in a prolonged period [84].

Irradiation can lead to physical and chemical changes in some food products that can affect the sensory properties of the food. Because of the appearance of an off flavor even at irradiation with 0.1kGy, milk and dairy products are unsuitable for radiation processing. The most suitable foods for irradiation are roots and cereals, tubers, and legumes, poultry, meat products, fish and seafood, most fruits and vegetables, herbs, spices, and seasonings. Some fresh fruits and vegetables may cause softening as a result of damage to the wall cells because of the radiation treatment. High-dose radiation of meat products can induce unpleasant off-flavors. There is a special concern about the effect of irradiation on vitamins. Vitamins in pure solution are more radiation sensitive compared with those in a food matrix or in dehydrated foodstuffs. As a whole, their changes are similar to those appearing in food processing such as drying and/ or canning. Some vitamins in foods are quite unaffected even by high doses of radiation, whereas some other vitamins, particularly vitamin E and B1, are rather sensitive and are the most radiation sensitive of the water-soluble vitamins. The radiation sensitivity of vitamins decreases in the following sequences [85]:

Fat-soluble vitamins: vitamin E > carotene > vitamin A > vitamin D > vitamin K

Water-soluble vitamins: vitamin B1 > vitamin C > vitamin B6 > vitamin B2 > folate, niacin, vitamin B12.

In the current study, researchers found that cranberry juice and commercial citrus extract could be used in hurdle approaches in combined treatment with c-irradiation to assure food safety without a detrimental effect on nutritional value and maintain low processing cost [86]. The effects of dietary α-tocopherol supplementation and gamma-irradiation on α-tocopherol retention and lipid oxidation in cooked minced chicken during refrigerated storage were studied. Minced breast and thigh meat from broilers fed diets supplemented with 100, 200, or 400 mg α-tocopheryl acetate/kg feed was irradiated at 2.5 or 4.0 kGy. Cooked irradiated and unirradiated meat was stored at 4 °C for 5 days. α-Tocopherol concentrations increased with increasing dietary supplementation. The concentrations decreased during storage, but retention was not affected by irradiation. The results suggest that overall; irradiation had little effect on lipid stability in  $\alpha$ -tocopherol-supplemented meat following cooking and storage [87].

The characteristics of oils extracted from gamma-irradiated sunflower and maize seeds at absorbed doses of 2, 4, 6, 8, and 10 kGy were investigated. Gamma irradiation did not affect the lipid, protein, fiber, and ash contents of either sunflower or maize seeds significantly. A small decrease in the contents of  $\alpha$ -,  $\gamma$ -, and  $\delta$ -tocopherols of both sunflower and maize oils was noted by radiation treatment up to 6 kGy, however, the decline was more pronounced at higher dosages [88]. A recent review has summarized all the obtained results of the effects of irradiation technology on the fruits and vegetables quality and safety [89]. It was reported

that the yellowness of cashew nuts increased as a function of irradiation dose and storage period. Irradiation up to a dose of 7 kGy had no effect on the color parameter values of irradiated walnuts. Some differences in sensory properties of irradiated pine nuts at a dose of  $\leq 5.0$  kGy were noted while no significant changes occurred in flavor and aroma between unirradiated and irradiated walnuts after irradiation up to 1.5 kGy [75].

Effects of irradiation on the physicochemical properties of three rice genotypes with different colors were investigated where a dose of 2, 4, 6, 8 and 10 kGy were applied. The bound phenolic content in all the genotypes was significantly increased with the increase in dose of irradiation. Gamma irradiation at high doses significantly increased the free, bound, and total antioxidant activities of three rice genotypes except for the free antioxidant activities of red rice. Though the color parameters were slightly changed, these changes could not be visibly identified. It is suggested that gamma irradiation enhanced the antioxidant potential and eating quality of whole grain-rice [90]. In a study, three gamma irradiation doses (1, 2 and 3 kGy) were applied in refrigerated raw whole milk to investigate the bacteriological and sensory qualities of this milk. All of the irradiated samples, mainly 2 and 3 kGy, exhibited a lower bacterial load than the non-irradiated samples. This study indicated that the 2 kGy irradiation dose improved the bacteriological quality of raw whole milk and likely did not negatively affect sensory characteristics by maintaining a constant mesophilic count and titratable acidity over the 60 days of refrigerated storage. [91].

In another study, the effect of gamma irradiation on the physicochemical properties and nutrient contents including water activity, fatty acid value (FAV), peroxide value (PV), carbonyl value (CV), malon-di-aldehyde (MDA) content, and lipase activity of peanuts were treated with 0, 1, 3, 5, and 10 kGy was evaluated. This study showed that irradiation has no significant effect on the moisture and ash contents and total sugar of peanuts. Low-dose irradiation did not significantly alter the water activity and protein and fat contents, but high irradiation levels significantly decreased the fat and protein contents (10 kGy) and increased the water activity (5 and 10 kGy). Gamma irradiation accelerated the degree of lipid oxidation and consequently increased PV, CV, FAV, and MDA contents. The fatty acid and amino acid composition were changed after irradiation treatment. Moreover, with the increase of irradiation dose, the lipase activity decreased. In addition, irradiation of 1 kGy is suitable for peanut seed according to this study [92].

The efficacy and effect of gamma irradiation at different doses on the nutritional and sensorial characteristics of grape juice blends during storage in a study were verified. Grape juice blends (irradiated to 2.0 kGy) presented the highest antioxidant content and the highest vitamin C increase after 90 days of post-irradiation storage compared to the other treatments. At 120 days the soluble solids content and total phenolic values were higher in the blends subject to 1.0 kGy and 1.5 kGy, respectively, when compared to other treatments. At 2.0 dose of kGy, the sensory tests showed that the quality of the grape juice blends remained unchanged. The gamma irradiation technique can be considered a viable alternative for quality preservation of grape juice blends during storage, as well as

to replace the heat treatment methods [93]. Gamma irradiation up to 2.5 kGy had no significant effect on the concentration of malic, citric and succinic acids, while the level of ascorbic acid decreased significantly at all irradiation doses (0–5 kGy) in the jujube fruit. The vitamins C and  $\rm B_1$  content significantly decreased at all applied doses (0–5 kGy), whereas  $\rm B_2$  content at doses 2.5 kGy was not significantly affected. The results of this study indicate that gamma irradiation at doses below 2.5 kGy can be successfully used for improving the quality of the jujube fruit [94].

Microbiological status, sensory properties, and shelf-life of prepacked beef meat at ambient temperature were all improved after irradiation with a dose of 2.5 kGy. Researchers have found that irradiation causes no significant differences in the flavor, texture, and color of beef irradiated at less than 3 kGy [95]. The impact of irradiation on meat odor, flavor, and chemical composition as a function of irradiation dose, depends on the type of meat, temperature, pH, packaging, presence of oxygen during processing, and presence of antioxidants. Increasing irradiation dose leads to an increase in the production of carbonyl compounds originating from the lipid and protein fractions of meat and of volatile olefins. Reducing the temperature during radiation treatment reduces the effects on odor and flavor [96]. In a study, it was carried out to evaluate the impact of gamma irradiation on physicochemical quality (pH, Hunter's parameter, oxidative and microbial stabilities, haem pigment), stability, and antioxidant status of chicken meat. Higher sensory attribute scores for attributes like appearance, taste, texture, flavor, and overall acceptability were found in the 2 kGy-treated groups. It was concluded that chicken meat treated with 2 kGy was considered better for microbial and physicochemical quality, antioxidant activity as well as sensorial properties of chicken meat [97]. Electron-beam irradiation to a 2 kGy has great potential for extending the shelflife of meatballs without affecting sensory properties. The effect of electron-beam on color, taste, odor, and consistency characteristics showed no significant impact [98].

Researchers used selected natural essential oils (Chinese cinnamon, Spanish oregano, and mustard oils) in combination with irradiation at 1.5 kGy of ground beef to eliminate mesophilic aerobic and pathogenic bacteria and to prolong the shelf-life of meat. Irradiation alone completely inhibited the growth of these bacteria. The combination of irradiation and essential oils was better for reducing lactic acid bacteria and Pseudomonas. The best-combined treatment for extending the shelf-life of ground beef for up to 28 days was essential oils plus irradiation (1.5 kGy) and modified atmosphere packaging [99]. In another study, results showed that e-beam radiation to 3 kGy has significant effect on the microbial quality, physic-chemical parameters, and the profile of fatty acids of the frozen meat of duck with no effect on sensory attributes [100].

In a recent study, the effect of gamma radiation at doses of  $0-10~\rm kGy$  on antioxidant activity in faba beans was investigated which revealed that the incorporation of irradiated fava bean, especially at 9 kGy, in meatballs formulation, improved the antioxidant activity and oxidative stability in non-irradiated and irradiated samples and increased their refrigerated shelf-life through delaying of the appearance of mold growth on the samples [101]. The effect of ir-

radiation source (gamma-ray, electron-beam, and X-ray) and dose levels at 0, 2.5, 5, 7.5, and 10 kGy on the physicochemical, organoleptic and microbial properties of cooked beef patties were studied. The results of this study concluded that quality attributes of meat products, in a particular color, lipid oxidation, and microbial properties are significantly influenced by the irradiation sources [102].

Radiation could serve as a preservation method for liquid egg white. Reviewers studied in a recent study, the antioxidant activities of egg white protein (EWP) were improved after electron beam irradiation (EBI) treatment. Under the bombardment of a high-energy electron beam, the S–S bond broke into the –SH bond and the  $\alpha$ -helix turned into  $\beta$ -sheet and unfolded structures. In addition, the results suggest that EBI treatment unfolded EWP and exposed the buried hydrophobic amino acids, which are related to antioxidant activities. With the increased EBI dose, the surface was perforated into a "honeycomb", the particle size increased and the sample stability decreased; thus, aggregation or cross-linking of the EWP occurred. This study also provided data for studying the mechanism by which EBI influences the antioxidant activity of proteins and provides a theoretical basis for the application of EBI to improve the antioxidant properties of proteins [103].

To assess physicochemical and functional properties, low doses (2, 3, and 4 kGy) of electron beam irradiation were applied to shell eggs. It was found that electron beam irradiation proved to be an effective method for controlling microbial growth in shell eggs without adversely affecting physicochemical and functional properties. The protein and sulfhydryl contents of the egg white were unchanged by irradiation to a 5 kGy; only a slight degradation of high-molecular proteins was detected. After irradiation to 5 kGy, the color of the yolk was transformed into pale yellow, and the egg white was modified to a turbid yellow. The results indicated that increase of doses led to an increase of the yolk and a decrease of white egg viscosity [104]. Also, changes in nutritional and functional characteristics of irradiated eggs with a minimal dose of 1.5 kGy required for the inactivation of nonpathogenic bacteria could be still acceptable for at-risk population and some industrial use [105].

# **Identification and Detection of Irradiated Foods**

The ability to reliably differentiate between irradiated and non-irradiated foods or ingredients is in the interest of government agencies, food processors, and consumers. Moreover, detection tests can be used to enforce the labeling requirements for identifying irradiated foods [2]. Labeling will enhance consumer confidence by providing assurance of the consumer's right to choose. Furthermore, the knowledge of radiation-induced chemical changes in food provides the scientific basis for the safety evaluation of the consumption of irradiated food [39]. Several detection methods have been subjected to inter-laboratory collaborative studies including electron spin resonance (ESR), luminescence methods, physical methods, chemical methods, and biological methods [106,107]. ESR measures the concentration of free radicals in irradiated matter. The luminescence methods measure the presence of excited molecules such as light emission upon heating material (thermo-luminescence, TL). Essential requirements of a thermo-luminescence substance to be used for dosimetry are low hygroscopicity, high sensitivity, energy dependence, and large linearity for very low dose measurements [108]. The chemical methods are based on the measurement of radiolytic products, e.g., using gas chromatography (GC) to measure volatile radiolytic products such as alkanes, alkenes and 2-alkylcyclobutanones in fat-containing food, or to measure non-volatile compounds such as 6-ketocholesterol and o-tyrosine. The biological methods are based on measurements of changes in viable microorganisms or changes in plant germination because of irradiation. The most practical methods are ESR (for foods containing bones, shells, or other particles), TL (for foods containing mineral dust particles), and GC (for fat containing food) [109]. Continuing efforts to develop detection methods are focusing on the DNA comet assay [110-113], and the changes in protein molecular mass distribution measured by discontinuous SDS-polyacrylamide electrophoresis and quantified by laser scanning densitometry [114].

# Labeling

Like other forms of processing, irradiation can affect the characteristics of food. Consumer choice mandates that irradiated food be adequately labeled and under the general labeling requirements, it is necessary that the food processor inform the consumer that food has been irradiated. Labeling of irradiated foods, however, is undergoing reevaluation in the US. If whole foods have been irradiated, FDA requires that the label bear the radular symbol and the phrase "treated with radiation" or "treated by irradiation." Yet, if irradiated ingredients are added to foods that have not been irradiated, no special labeling is required on retail packages. Special labeling is required for foods not yet in the retail market that may undergo further processing in order to ensure that foods are not irradiated multiple times. In this regulation, FDA advises that other truthful statements, such as the reason for irradiating the food, may be included [115]. Because the words "radiation" and "irradiation" may have negative connotations, the labeling requirement has been viewed as an obstacle to consumer acceptance.

Many in the food industry believe that an alternative wording, e.g., "electronically pasteurized," would be helpful. In 1997, Congress attempted to resolve these issues in two ways. First, it mandated that the FDA could not require print size on a label statement to be larger than that required for ingredients and second, it directed the FDA to reconsider the label requirement and to seek public comment on possible changes. The FDA had not in fact mandated a type size but did require a statement that would be prominent and conspicuous. In response to this congressional directive, the FDA published an Advance Notice of Proposed Rulemaking (ANPR) in 1999 seeking public comment on the labeling of irradiated food, particularly on whether the current label may be misleading by implying a warning and invited suggestions of alternative labeling that would inform consumers without improperly alarming them. Thousands of comments were received, with a large number compiled into a categorical database for further examination by the CF-SAN's Office of Nutritional Products, Labeling, and Dietary Supplements. This leading office for labeling policy has not yet determined whether there will be a change in labeling requirements [2].

#### **Acceptance by Consumers**

For significant commercialization of irradiated food to proceed, it is necessary for food producers to adopt innovative technology for retailers to market the product and for consumers to purchase it [116]. It has generally been thought that the major barrier to food irradiation was reluctance by consumers to accept irradiated food. The food industry needs to be aware that this technology should never be used as a "clean-up" technology. The food industry should use this technology as a final step of a comprehensive pathogen reduction and elimination program so that only very high-quality food items are treated. By doing so, the doses that are employed can be significantly reduced to achieve significant improvements in public health [117,118]. The evidence of consumers' acceptance is now overwhelming that consumers will buy irradiated food; consumers make repeat purchases of labeled irradiated food in several countries. Although a significant minority of consumers may wish to avoid buying irradiated food, the evidence is clear that there is a market for irradiated food [119]. The greatest barrier to greater application of irradiation may be the persistent perception among food producers and retailers that consumers will not buy the product [116].

# **Food Irradiation Regulation**

Approximately 60 countries have approved the use of food irradiation in their health or food regulations for at least one, and usually more, food or food class. The IAEA keeps a Food Authorizations Database [120]. However, there is no obligation, only a request for countries to lodge approvals with the agency and it is doubtful whether it is completely up to date. Most countries would claim that their regulations are based on the Codex General Standard. However, the Standard does not name specific foods that may or may not be irradiated. All food may be irradiated to the maximum approved absorbed dose. Very few countries have a regulation which allows the irradiation of any food subject only to compliance with the Codex Standard. Within their regulations most countries adopt the Codex General Standard on Labeling of Pre-packaged Foods [121]. However, the interpretation and enforcement of labeling provisions is variable. Several countries allow a statement of irradiation purpose or verifiable benefit on the label. Indonesia requires the purpose to be included. In many countries, the regulation is silent on a statement of purpose or benefit [116].

#### **Conclusion**

Radiation technology is a well-established process. Radiation technology of food irradiation is gaining more and more attention around the world. In comparison with heat or chemical treatment, irradiation is a more effective and appropriate technology that can be used to ensure food safety by eliminating insects and pathogens to prolong the shelf life. The radiation technique makes the food safer to eat by destroying bacteria which is very much like the process of pasteurization. The process can be applied to fresh or frozen products without affecting the nutritional value. Many studies have shown that radiation technology for food irradiation in combination with other treatments can be used as an innovative and effective method to add value to food products. As previously detailed, people are still confused and fail to differentiate irradiated foods

from radioactive foods. When well informed, a reduced number of consumers will reject irradiated food. What a consumer is looking for is a product with good quality and a competitive price. When consumers are aware of the short- and long-term dangers of chemical additives, they accept more irradiation treatments being applied to food products. As radiation technology safely preserves food and controls pathogens, consumers and food processing companies will benefit from the commercialization of this process.

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#### **Conflict of Interest**

No conflict of interest.

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