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ARTICLE

Simulation of Gamma-Ray Irradiation of Lettuce Leaves in a ^{137}Cs Irradiator Using MCNP

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Ionizing radiation effectively reduces the number of common microbial pathogens in fresh produce. However, the efficacy of the process for pathogens internalized into produce tissue is unknown. The objective of this study was to understand gamma irradiation of lettuce leaf structure exposed in a ^{137}Cs irradiator using MCNP. The simulated ^{137}Cs irradiator is a self-shielded device, and its geometry and sources are described in the MCNP input file. When the irradiation chamber is filled with water, lower doses are found at the center of the irradiation volume and the dose uniformity ratio (maximum dose/minimum dose) is 1.76. For randomly oriented rectangular lettuce leaf segments in the irradiation chamber, the dose uniformity ratio is 1.25. It shows that dose uniformity in the Cs irradiator is strongly dependent of its filling density. To understand dose distribution inside the leaf, we divided a lettuce leaf into a low density (flat) region (0.72 g/cm^3) and high density (rib) region (0.86 g/cm^3). Calculated doses to the rib are 61% higher than doses to the flat region of the leaf. This indicates that microorganisms internalized in the rib can be inactivated more easily than organisms on the surface. This study shows that irradiation can effectively reduce viable microorganism internalized in lettuce.

KEYWORDS: food irradiation, Monte Carlo, MCNP, Cs irradiator, lettuce

I. Introduction

Consumption of fresh fruits and vegetables is important not only in a healthy diet, supplying much needed vitamins, minerals, and fiber, but also in the prevention of heart disease, cancer, and diabetes. Consumers now expect fresh produce year-round and its consumption has been increasing at a dramatic rate, more than doubling over the past two decades. Concomitantly, there has been an increase in the number of food-borne illness outbreaks linked to fresh produce consumption[1-3] (e.g. from fewer than 20 throughout the 1970s to more than 100 in the 1990s).

Washing and sanitizing treatments (chlorine, ozone, chlorine dioxide, peroxyacetic acid) of fresh produce are widely used methods in reducing the population of human pathogens,[4] such as *E. coli* and *Salmonella*. They have, however, limited efficacy in inactivating pathogens from fresh produce because of the strength of microbial attachment to produce and the location of the attached pathogens in inaccessible sites.

Higher bacterial incidence in the vein area was reported for *Salmonella* on cilantro leaves[5] and for *E. coli* on lettuce leaves.[6] The enhanced moisture of this part of the leaf and more abundant nutrients may allow for easier contact and attachment of pathogens to the plant. Researchers even have suggested that *E. coli* can be taken up by the roots and transported to the edible portion of a lettuce leaf.[7-9] These internalized pathogens cannot be washed off, and they are protected from environmental stresses. Furthermore, they are capable of growth during transport and storage, and thus present a human health risk. Therefore, it is necessary to develop alternative decontamination methods with better penetration and higher lethality.

Ionizing irradiation is a non-thermal treatment that has the ability to effectively eliminate human pathogens from fresh produce.[10] D_{10} values, the amount of radiation necessary to achieve a 90% (1-log) reduction, are in the range of 0.12 to 0.30 kGy for lettuce and baby spinach.[11-13] Also, their sensory attributes were not affected by irradiation up to 1 and 2 kGy, respectively. In August 2008, the Food and Drug Administration (FDA) allowed the use of ionizing radiation (up to 3 kGy) to make lettuce and spinach safer and delay spoilage.

In general, when individual electrons or photons interact with the food, the resulting dose distribution depends on the geometry and atomic density of the food. In the case of bagged spinach leaves,[13] doses are not uniform because of density variations of the surrounding product (the spinach leaf) due to the presence of air pockets and the random arrangement of the leaves inside the bag. Kim *et al.*[14] divided bagged Romaine lettuce into loosely-packed, densely-packed, and a root-closed area, and reported that the dose uniformity ratio (maximum dose/minimum dose) was 2.97, 1.69, and 1.73, respectively, for double electron beam treatment. Thus, dose distribution within the bag must be determined to assure proper treatment of the produce.

Furthermore, to ensure adequate treatment of fresh produce, radiation interactions at the surface and internal regions should be thoroughly understood. Published reports have

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shown irradiation to be an efficient method for eliminating pathogens from the internal spaces of lettuce and spinach leaf pieces[15)] and the D_{10} value in the internal regions is three times higher than the value at the surface,[11)] which means pathogens in the internal region could be more resistant than the ones on the surface. Even the radiation sensitivity of pathogens appeared to vary with the variety of fresh produce.[12, 16)]

Dosimeters, in radiation research and commercial processing, are used for quality and process control. In fresh produce, especially leafy vegetables, no dosimeters are available for measuring doses on the surface and internal regions. Up to now, for all radiation experiments with leafy vegetables, regardless of the radiation source, doses have been measured with alanine dosimeters or radiochromic film dosimeters. An alanine dosimeter is in the form of tablet (usually 5 mm in diameter and 3 mm thick), and its reading is the average of the absorbed dose over the volume of the tablet. Thus, it is almost impossible to measure doses at the thin surface area of leafy vegetables using alanine dosimeters. Radiochromic film dosimeters have been known to be less accurate or reproducible in their radiation response compared to reference dosimeters, such as alanine dosimeters or Fricke dosimeters.[17)] Inaccurate interpretation of dose measurement can result in misleading D_{10} values for a target pathogen in a particular produce.

Our objective in this study was to understand gamma irradiation of lettuce leaf structures exposed in a ^{137}Cs irradiator using MCNP so that the radiation treatment of fresh leafy vegetables can be optimized.

II. Materials and Methods

1. Cesium-137 Gamma Irradiator

Self-contained gamma-ray irradiators contain radioactive sources, such as ^{137}Cs or ^{60}Co, that emit ionizing electromagnetic radiation (gamma rays), under properly shielded conditions. These irradiators have an enclosed irradiator sample chamber connected to a sample positioning system. They can be used for many radiation processing applications: calibration of dosimeters, irradiation of small samples for radiation process validation, and batch irradiation of microbial samples.[18)]

In this study, a GammaCell[19)] type ^{137}Cs gamma irradiator was used as a radiation source for a Monte Carlo simulation. It contains sealed ^{137}Cs sources in an annular array around the irradiation chamber, resulting in a relatively uniform absorbed dose distribution.[18)] The radiation source material is contained in pellet form within 23 pairs of rectangular stainless steel tubes. These tubes are arranged vertically and concentrically around the internal perimeter of the irradiation chambers.[20)] **Figure 1** shows a schematic drawing of the irradiator. The energy level of the ^{137}Cs is 0.66 MeV.

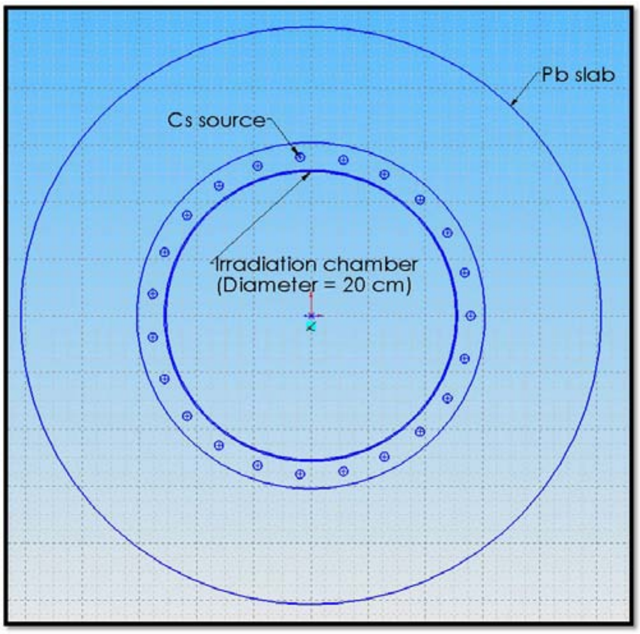


Fig. 1 Cross section of a Cesium-137 irradiator with the location of Cs sources

2. Monte Carlo Simulation

The MCNP5 (Monte Carlo N-Particle, Version 5) used in this study was developed at Los Alamos National Laboratory. In this Monte Carlo simulation, the incident photon and secondary particles produced in a target are tracked. Those electron tracks start off at a given positions with initial direction and energy. An electron path is broken into many steps to follow an electron through a significant energy loss. These steps are chosen not only to be long enough to include many collisions so that the multiple scattering theories are valid ("major steps"), but also to be short enough that mean energy loss is in any step is small ("sub-steps"). The energy loss and angular deflection of the electron during each steps can then be sampled from probability distributions on the appropriate multiple scattering theories. The electron tracks are finished either when they leave the material system or when the energy becomes smaller than an energy cutoff, which is the energy where particles are assumed to be effectively stopped and absorbed in the medium. The MCNP default cross sections for photon and electron transport in water were used. MCNP, along with EGS4, has been used extensively in medical physics.

To obtain the dose distribution in the irradiation chamber, a water phantom was used for simulation. The phantom was a circular cylinder with a radius of 8 cm and a height of 1 cm. The cylinder was divided into 10 degree sections, as indicated in **Fig. 2**, and the average dose was computed for each volume element. Twenty-three Cs point sources surround the phantom and are assumed to be mono-energetic and isotropic.

The lettuce leaf was assumed to be a simple rectangular-shape (0.2 cm x 0.8 cm x 3.0 cm) for the dose simulation. They were randomly oriented in the irradiation chamber, as seen in **Fig. 3**. Lettuce density (0.86 g/cm^3) was taken from the data of Han *et al.*[21)], and its atomic composition was calculated from data from the USDA National Nutrient Database

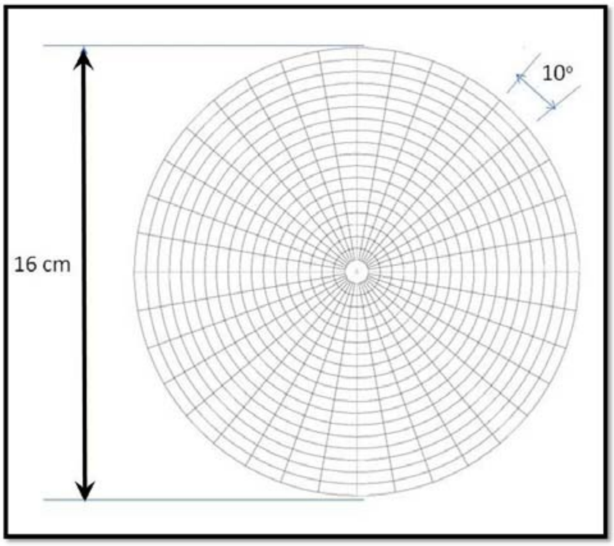


Fig. 2 The cross section of the cylindrical water phantom used in the Monte Carlo simulation

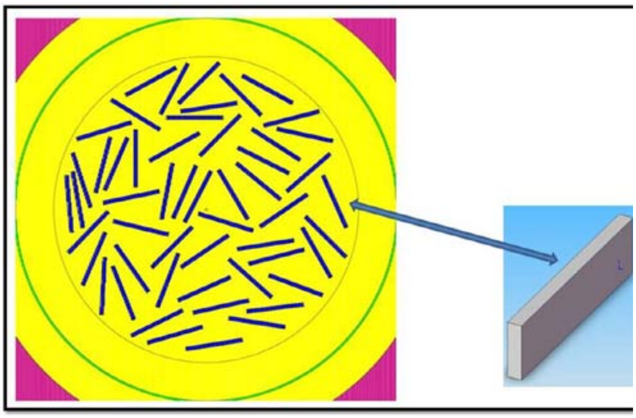


Fig. 3 Rectangular-shape (0.2 cm x 0.8 cm x 3.0 cm) lettuce leaves in the irradiation chamber

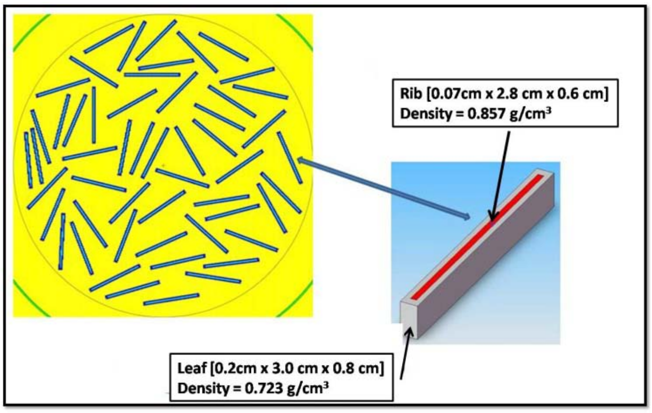


Fig. 4 Low density (flat) region and high density (rib) region of lettuce leaf structure

for Standard Reference, as seen in **Table 1**. In order to understand dose distribution inside the lettuce leaf, we divided a lettuce leaf into a low density (flat) region (0.2 cm x 0.8 cm x 3.0 cm, 0.72 g/cm^3) and high density (rib) region (0.07 cm x 0.6 cm x 2.8 cm, 0.86 g/cm^3). Like a whole lettuce simulation, these lettuce leaves are randomly oriented in the radiation chamber in **Fig. 4**. The rib region was considered as a passage of nutrition and water from root to leaf surface. It is also likely to be a location for attached microorganisms, and it is hard to control them with surface treatments.

Table 1 Elemental composition of lettuce leaf

	Elemental composition (wt%)
H	10.73
C	2.25
N	0.20
O	86.41
Ca	0.04
Mg	0.02
P	0.04
K	0.30
Na	0.01

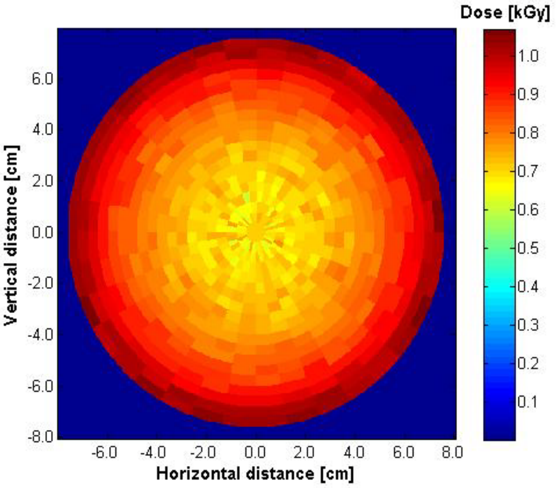
The pulse height tally (F8) was used for scoring dose deposition. This tally scores the energy (MeV) of a photon or electron as it enters or leaves a cell, which is analogous to a physical detector. A positive energy tally occurs from particles entering a cell and a negative energy tally occurs when particles exit a cell. The simulation was run in parallel computer platform (Dell Poweredge 6650) equipped with 4 of 1.5 GHz Intel Xeon processor, 8.0 GB of RAM, and a Red Hat Linux 8 operating system. The CPU time was approximately 120 hours for 10 x 10^6 histories. The estimated relative error in the Monte Carlo dose calculation, defined as one estimated standard deviation of the mean divided by estimated mean, was reduced to below 0.05.

III. Results and Discussion

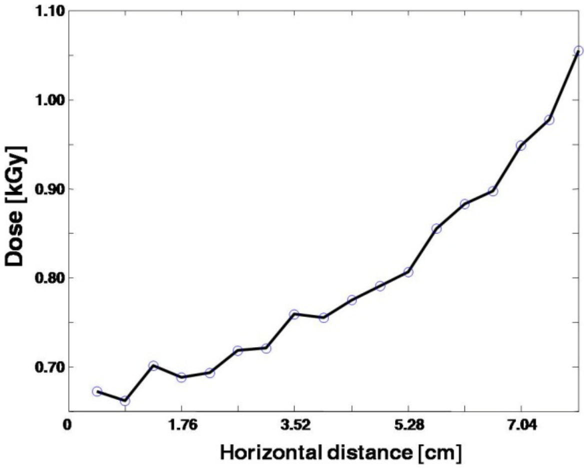
1. Absorbed Doses in Water in the Cs Irradiator

Figure 5 shows the calculation results of the absorbed doses in water. When the irradiation chamber is filled with water, the lower doses are found at the center of the irradiated volume. Dose uniformity ratios are 1.30 and 1.76 for up to 3.5 cm and 8.0 cm of radius, respectively, and are acceptable for irradiation treatment of leafy vegetables.

Shieh *et al.*[20] obtained depth-dose distribution for the Cs irradiator, placing radiochromic film dosimeters into the sample compartment at the desired position. In the horizontal positions the dose distribution was very homogenous about 1% to 2% higher at the peripheries of the sample compartments as compared to the center locations. These results showed that dose distribution in the Cs irradiator is strongly dependent of its filling density (total mass of sample/volume of irradiation compartment); Shieh *et al.*[20] measured doses inside empty cans (sample holders). Since its dose uniformity ratio is acceptable at an even fully packed volume, the Cs irradiator is good for irradiation treatment for food of various densities from leafy vegetables to frozen chicken carcasses.



Dose distribution of water (Average dose: 0.85 kGy)



Doses at a vertical position of 0 cm

Fig. 5 Dose distribution of water in the Cs irradiator

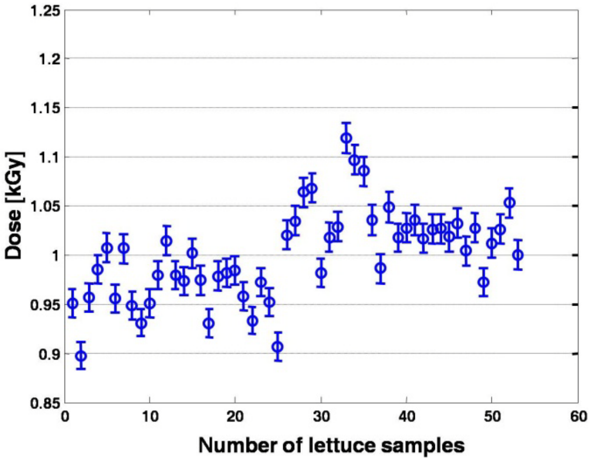


Fig. 6 Doses in lettuce leaf segments in the Cs irradiator

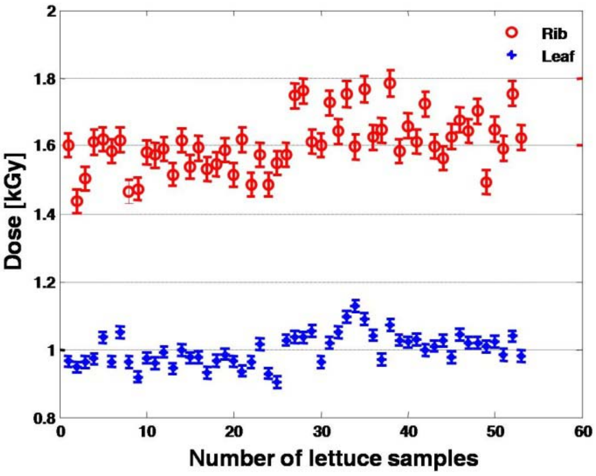


Fig. 7 Doses in the lettuce structure: low density (flat) region and high density (rib) region

2. Distribution of Dose to Lettuce in the Cs Irradiator

For the simulation with rectangular lettuce leaf segments, the dose uniformity ratio is 1.25, which is better than the water simulation, as seen in **Fig. 6**. The standard deviation of these doses is only 4.61% of the average dose. To compare doses from the radiation source, lettuce leaf segments were divided into two groups: (1) those for which the distance between the center position of each lettuce segment and the center of the radiation chamber was less than 4 cm, and (2) those for which the distance was between 4 cm and 8 cm. Doses in group two are 8.0% higher than those in group one. The orientation of the lettuce leaf segments does not affect their absorbed dose. Thus, the closer to the radiation source, the higher the dose to the lettuce leaf segments.

Gamma ray photons lose their kinetic energy in large interactions and have no limiting range through matter (exponential attenuation). In this Cs irradiator, where radiation source materials are arranged concentrically around the irradiation chamber, doses to the sample are much more uniform than in a narrow beam geometry where doses to group two is 38.5% higher than group one in the water sample from the Monte Carlo simulation. These results show that the doses to the lettuce leaf segments are very uniform and the uniformity ratio is dependent on the filling density of the Cs irradiator.

The uniformity ratio of the dose distribution in the flat and rib regions inside the leaf are 1.24 and 1.25, which are similar to the ones for the whole lettuce leaf segments in **Fig. 7**. It should be noted that a high dose for a flat region does not necessarily result in a high dose at the rib. The variation might be due to the location and orientation of the lettuce samples.

Calculated doses to the rib are significantly higher (60.7%) than doses to the flat portion of the leaf. This indicates that microorganisms internalized in the rib can be inactivated more easily than organisms on the surface.

IV. Conclusion

In this study, we simulated the exposure of lettuce leaves to gamma irradiation in a ^{137}Cs irradiator. For randomly oriented rectangular lettuce leaves, the absorbed doses are very uniform with a uniformity ratio of 1.25. Dose uniformity is strongly dependent on the filling density of the Cs irradiator and calculated doses to the rib are 61% higher than doses to the rest of the leaf. These simulation results could be validated with cylinder or box-shaped phantoms and radiochromic film dosimeters. This is the first study of dose distribution in leafy vegetables using a Cs irradiator, targeting internal microorganisms. These results provide valuable information for optimizing irradiation treatment for leafy vegetables. However, in this study we simplified the lettuce structure into a rectangular shape and the only radiation source was gamma rays from ^{137}Cs. Geometry extracted from CT or MRI images and various radiation sources (industrial-type 10 MeV electrons, 5 MeV X-rays, and gamma rays from ^{60}Co) would improve the validity of the irradiation treatment of leafy vegetables. In conclusion, this study shows that irradiation can effectively reduce viable microorganisms internalized in lettuce and could be used as an effective killing step to mitigate the risk of food-borne disease outbreaks.

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