



Surface Treatment of Eggshells with Low-Energy Electron Beam

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ABSTRACT

Background: *Salmonella enteritidis* (SE) was the main cause of the pandemic of foodborne salmonellosis. The surface of eggs' shells can be contaminated with this bacterium; however, washing them with sodium hypochlorite solution not only reduces their flavor but also heavily impacts the environment. An alternative to this is surface sterilization using low-energy electron beam. It is known that irradiation with 1 kGy resulted in a significant 3.9 log reduction (reduction factor of 10,000) in detectable SE on the shell. FAO/IAEA/WHO indicates irradiation of any food commodity up to an overall average dose of 10 kGy presents no toxicological hazard. On the other hand, the Food and Drug Administration has deemed a dose of up to 3 kGy is allowable for eggs. However, the maximum dose permitted to be absorbed by an edible part (i.e., internal dose) is 0.1 Gy in Japan and 0.5 Gy in European Union.

Materials and Methods: The electron beam (EB) depth dose distribution in the eggshell was calculated by the Monte Carlo method. The internal dose was also estimated by Monte Carlo simulation and experimentation.

Results and Discussion: The EB depth dose distribution for the eggshells indicated that acceleration voltages between 80 and 200 kV were optimal for eggshell sterilization. It was also found that acceleration voltages between 80 and 150 kV were suitable for reducing the internal dose to ≤ 0.10 Gy.


Conclusion: The optimum irradiative conditions for sterilizing only eggshells with an EB were between 80 and 150 kV.

Keywords: Food Irradiation, Low-Energy Electron Beam, Sterilization, Internal Dose, Monte Carlo Simulation

Original Research

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Introduction

Foodborne illness occurs around the world. For example, a *Salmonella enteritidis* (SE) outbreak caused by infected eggs sickened 38 people in seven states in the United States [1]. Japanese people have traditionally enjoyed raw eggs, resulting in salmonellosis becoming prevalent in the 1960s [2]. In 2007, it was reported that SE was detected in the shells of 5 out of 2,030 (0.25%) packs of eggs in Japan despite shells had been washed before packing [3]. Therefore, the Japanese government mandated the following management and operation standards for washing the eggshells [4].

- (1) The washing water should consist of a sodium hypochlorite solution (≥ 150 ppm), or the fungicide should possess the same sterilization ability against *Salmonella* (1/1,000).

(2) The water for washing eggs should be 30°C and must be 5°C warmer than the eggs.

(3) Brushes used for washing eggs should be clean and hygienic.

Washing eggs with sodium hypochlorite solution reduces their flavor and heavily impacts the environment. Therefore, this study explored the use of low-energy electron beam (LEEB) as an alternative method for sterilizing the eggs' surfaces. LEEB is generally defined the energy of electron is up to 300 kV. With the relatively recent development of reliable, compact, cost-effective, LEEBs, a new class of in-line applications is now possible. The benefits of high-speed, high efficacy treatments, with no chemicals and at room temperature, are now realized across a variety of packaging applications.

The penetration depth of an electron beam (EB) can be controlled by adjusting its energy. The penetration depth of electrons from 50 kV to 10 MV is 0.025–50 mm in water [5, 6]. Considering that the shells of the egg are typically 0.26–0.50 mm thickness, LEEB (≤ 300 kV) is therefore useful for sterilizing surface contamination only [7]. The D_{10} value (radiation dose needed to reduce 90% of the *Salmonella* population) is 0.30 kGy, and the value required for a 4-log reduction of *Salmonella* is 1 kGy [8, 9]. Furthermore, SE can no longer be detected at 2–3 kGy [8]. On the other hand, the Food and Drug Administration indicated that the irradiation of eggs is not to exceed 3.0 kGy. In addition, irradiation of eggshells up to 2.5 kGy had little negative impact on the physiochemical and functional properties of liquid egg while [10]. The use of food irradiation is prohibited in Japan except for the inhibition of potato sprouts. However, irradiation of up to 0.10 Gy is permitted in food, as X-rays are used for foreign matter inspection in Japan [11]. In this case, 0.10 Gy is defined at the surface of the edible part. Therefore, if the absorbed dose of the egg's edible tissue dose not exceeds 0.10 Gy, they can be irradiated with EB without the permission from the government. The electrons accelerated in the EB generator pass through a window made of titanium or aluminum foil which maintains the internal vacuum and applies a high voltage to accelerate the electrons. X-rays are emitted when EB interact with titanium foil. X-rays are similarly emitted upon the interaction of eggshells and EB, and the X-rays' energy and dosage increase with the EB energy and dosage [12]. X-rays have much higher penetrative power than electrons, travel several tens to hundreds of centimeters in water (depending on their energy), and gradually lose their energy as they collide with atoms in water. Because X-rays also irradiate an egg's edible

tissues during EB exposure, it is necessary to ensure that the egg's absorbed dose does not exceed 0.10 Gy. Therefore, the following two conditions are necessary: (1) the irradiating EB's depth dose distribution in the given eggshell and (2) absorbed dose of the edible tissue does not exceed 0.10 Gy.

These requirements have led to a new approach to food irradiation using LEEB, which utilizes relatively small devices that generate ionizing radiation with energies of hundreds of kilovolts. In this study, the eggshells were irradiated with LEEB, and the irradiation conditions were examined via experimentation and Monte Carlo simulation. The purpose of this study is to measure the internal dose distribution regardless of the 0.10 Gy limit.

Materials and Methods

1. Simulation Methods

1) Depth dose distribution with EB by Monte Carlo simulation

The depth dose distribution in the eggshells was calculated using Monte Carlo simulation (Particle and Heavy Ion Transport code System [PHITS] ver. 3.02) to irradiate only the shell [13]. Fig. 1 illustrates irradiation model. Eggshell must be thick to estimate the depth dose distribution because the commercial eggs have different shell thicknesses depending on egg size. The thickness of the eggshell was 0.60 mm, and the eggshell's absorbed dose was calculated at every 0.02 mm.

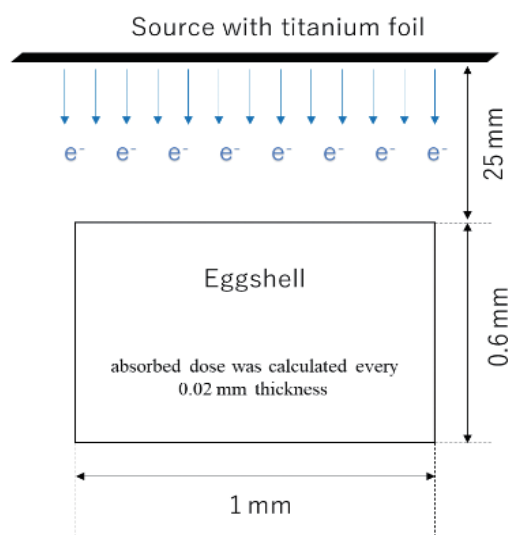


Fig. 1. Depth dose distribution model with electron beam. There was a titanium foil (10 μm thickness) under the source. The thickness of the eggshell is 0.60 mm, where the absorbed dose in the eggshell was calculated at every 0.02 mm thickness.

Eggshells are generally composed of 96% calcium carbonate (CaCO_3) and 4% organic substances. The density of the eggshell is lower than that of pure CaCO_3 (2.7 g/cm^3) because of its high porosity. Therefore, the eggshell used for the simulation was set to 100% CaCO_3 with a density of 2.0 g/cm^3 [14]. The EB were emitted from a plane source ($6 \text{ cm} \times 6 \text{ cm}$), and the distance from the source to the egg was 2.5 cm. Titanium foil (thickness = $10 \text{ }\mu\text{m}$) was placed under the source. The EB energies were 80, 100, 150, 200, and 250 keV, and the number of emitted electrons was 6.25×10^{12} . The energy spectrum of the electrons irradiated at 80 kV was calculated.

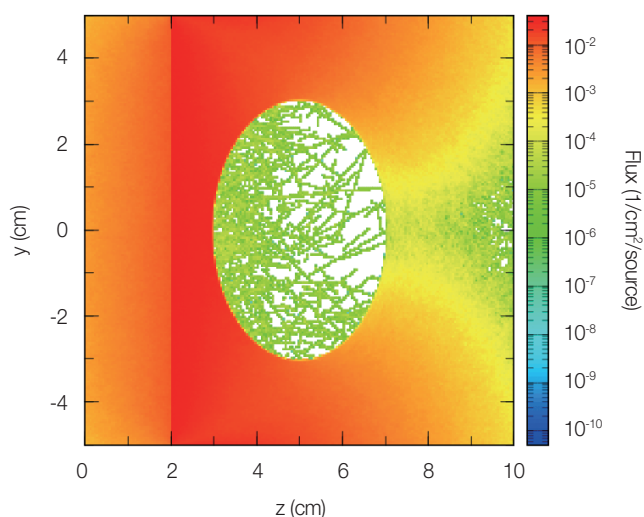


Fig. 2. The simulation diagram to estimate dose of the edible part in eggs. The flux is electron and photon.

2) Internal dose from EB-associated X-rays calculated by Monte Carlo simulation

Fig. 2 shows the simulation diagram; the length of the egg's major axis was 60 mm, and its minor axis was 40 mm. The eggshell's thickness was 0.40 mm. The edible area was set to 100% H_2O with a density of 1.0 g/cm^3 , and the shell was set to 100% CaCO_3 with a density of 2.0 g/cm^3 [14]. The EB were emitted from a plane source ($10 \text{ cm} \times 10 \text{ cm}$), and the distance from the source to the egg was 1.0 cm. Titanium foil (thickness = $6 \text{ }\mu\text{m}$) was placed at a distance of $20 \text{ }\mu\text{m}$ from the source. The internal dose was calculated at the center of the egg, with a size of 1 cm cubic meter. The eggshell's absorbed dose was calculated simultaneously. Therefore, the internal dose was estimated while the eggshell was irradiated at 3 kGy. The energies of the EB were 80, 100, 150, 200, and 250 keV.

2. Experiment Methods

1) Egg sample and dosimetry

The dosimeter which is measured in egg should be small to measure the dose distribution in egg. It was necessary to detect at 0.10 Gy and to be low fading effect. TLD-100 (Thermo Fisher Scientific, Waltham, MA, USA; chip $3.2 \text{ mm} \times 3.2 \text{ mm} \times 0.9 \text{ mm}$) satisfied the above requirement and was very useful in this work. The contents of the raw eggs were first removed and dried. TLD-100 was wrapped with polyethylene film to keep it dry. This TLD-100 was then placed as shown in Fig. 3 and solidified with agar. The internal doses of the egg were

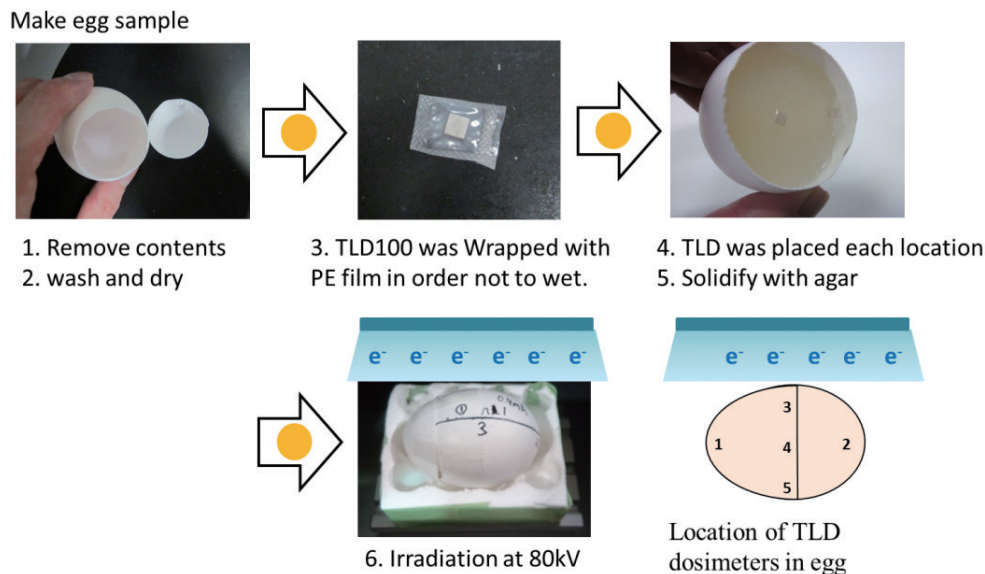


Fig. 3. TLD-100 dosimeters were inserted to eggs and located to measure the internal dose of eggs.

measured with TLD-100. The amount of light emitted from the TLD-100 was measured with a TLD reader (Harshaw 3500; Thermo Fisher Scientific).

2) EB irradiation

EB irradiation was carried out at 80 kV with an Eye Compact (EC90/10/50L; Iwasaki Co. Ltd., Tokyo, Japan). A titanium window (thickness = 6 μm) was under the source. On a conveyor, the egg sample was passed through the scan area at a constant speed. The current value and the conveyor speed were 0.1 mA and 5.0 m/min, respectively. The distance from the source to the egg was 1.0 cm. At that time, the absorbed dose at the shell's upper side (Location #3 in Fig. 3) was 2.76 kGy. In this experiment, the egg sample was irradiated with EB from one direction. Afterwards, the egg sample was turned in the opposite direction and irradiated with EB again.

Results and Discussion

1. Depth Dose Distribution of Eggshell with EB by PHITS

The eggshells (0.60 mm) were irradiated with EB (80, 100, 150, 200, and 250 keV); according to Monte Carlo simulation, the number of emitted electrons was 6.25×10^{12} . Fig. 4 shows the results of the eggshell's depth dose distribution obtained PHITS code. When the eggshell was irradiated at 200 kV, the EB penetrated up to 0.2 mm. After 0.2 mm, bremsstrahlung

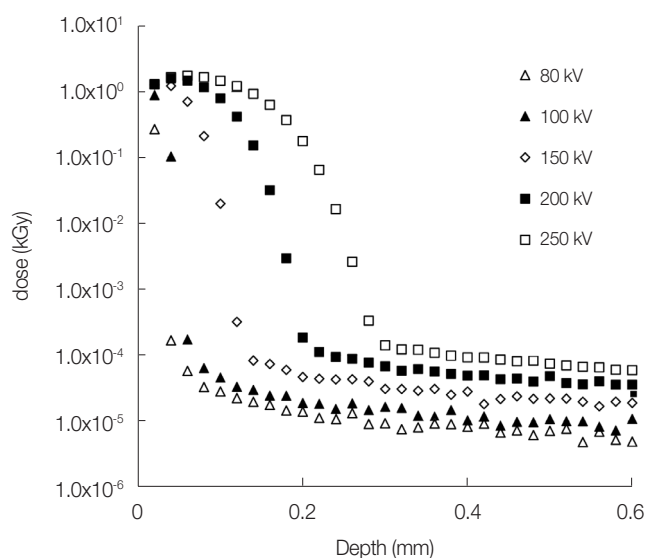


Fig. 4. Depth dose distributions of eggshell irradiated with low-energy electron beam by the Particle and Heavy Ion Transport code System (PHITS) code. Maximum value of uncertainty is 10% (2σ).

was contributed to the eggshell because the dose remained constant after 0.2 mm. Therefore, the EB penetrated the eggshell to a depth of 0.02, 0.04, 0.12, 0.20, and 0.30 mm for energy levels 80, 100, 150, 200, and 250 keV, respectively. This means that, if LEEB irradiated an eggshell at 250 kV, they would reach the egg's edible tissue because the minimum thickness of an eggshell is 0.26 mm. When this edible tissue was irradiated at EB, the permitted dose of 0.10 Gy was exceeded. Thus, the energy of the EB was high enough to pass through the eggshell, reach the edible tissue, and exceed the dosage limit (≤ 0.10 Gy). However, such high energy is needed to sterilize the *Salmonella* that can subsist in eggshells' pores. The optimal energies were between 80 and 200 kV. Fig. 5 shows the energy spectrum of the electrons that irradiated the eggshell at 80 kV. Their maximum energy was 73 keV because the electrons were scattered by the titanium foil and air on the way to the eggshell.

2. Internal Dose by PHITS

EB at each energy level were directed to the eggs as shown in Fig. 2, and the edible tissues' absorbed doses were calculated by Monte Carlo simulation. Fig. 6 shows the absorbed doses of the edible tissues irradiated with EB at each energy. The radiation that reached the edible tissues was bremsstrahlung rather than electron. The vertical axis in Fig. 6 represents the edible tissue's absorbed dose when the eggshell was irradiated at 3 kGy. The edible tissue's absorbed doses

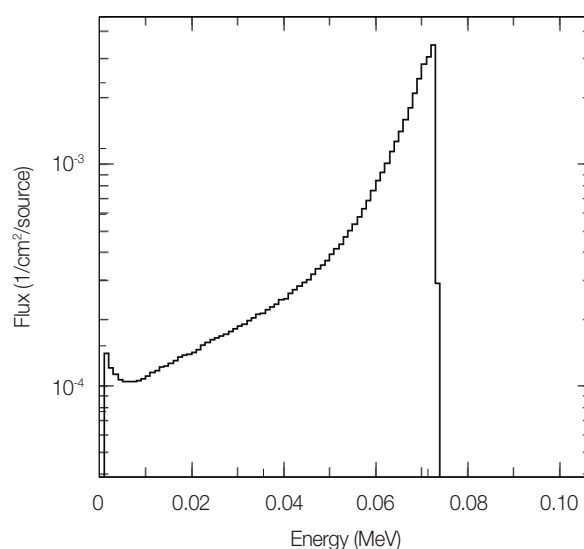


Fig. 5. Energy spectrum irradiated with electrons at 80 kV on the eggshell.

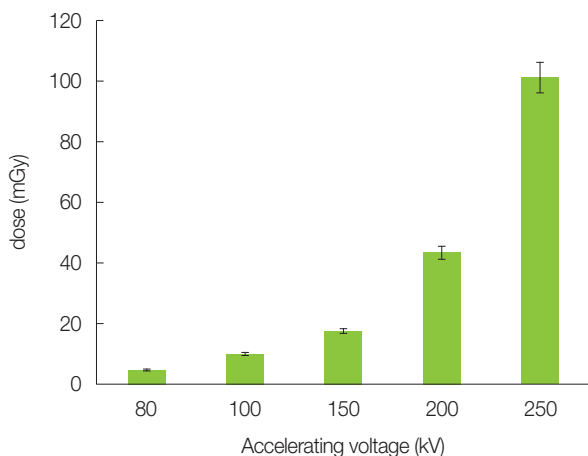


Fig. 6. Absorbed doses of edible part in eggs irradiated with electron beam at each energy were calculated with the Particle and Heavy Ion Transport code System (PHITS) code.

increased along with the energy, exceeding 0.10 Gy at 250 kV. In this simulation, EB irradiation was emitted in one direction only. However, it is necessary to irradiate the entire eggshell. In such a case, irradiation at energies between 200 and 250 kV could exceed 0.10 Gy. Therefore, the optimal energies were between 80 and 150 kV.

3. Internal Dose by Experimentation

The egg sample was irradiated with an EB at 80 kV from one direction. After the egg sample was turned in the opposite direction, it was irradiated again. Each thermoluminescent dosimeter (TLD) in the egg was taken out, and the internal dose was estimated. Table 1 presents the internal doses at each location. The internal doses were estimated when the eggshell was irradiated at 3 kGy; they were much lower than 0.10 Gy in all locations. From the unidirectional irradiation, it was found that the internal dose increased at Location #3. Therefore, the internal dose was higher on the side facing the source. Following bilateral irradiation, the internal dose at Location #3 was close to that of Location #5. These results demonstrate that the internal dose can be made uniform by irradiating the entire egg.

4. Comparison between Experiment and Simulation

In a previous study, the comparison of the internal dose of egg between the experimental and the simulated values was a very good relationship [14]. In this study, the experimental and simulated values when irradiated at 80 kV were 3.18 mGy (Location #3) and 4.80 mGy, respectively. The reason for the difference is that the eggs were irradiated with being conveyed

Table 1. Internal Dose of Each TLD Location at Unilateral Irradiation and Bilateral Irradiation

Location no.	Internal dose (mGy)	
	One side irradiation	Both side irradiation
1	1.76 ± 0.35	2.46 ± 0.32
2	1.44 ± 0.29	2.64 ± 0.34
3	3.18 ± 0.32	4.27 ± 0.50
4	1.45 ± 0.29	2.18 ± 0.28
5	1.12 ± 0.22	4.06 ± 0.46

Values are presented as mean ± standard deviation. These average doses in three times were estimated when the eggshell was irradiated at 3 kGy. TLD, thermoluminescent dosimeter.

in the experiment. In the simulation, the internal dose of egg was evaluated without moving it. In the future, we plan to conduct experiments and simulations to evaluate the dose when the eggs are rotated and conveyed.

Conclusion

Food irradiation is prohibited in Japan except in potato sprout inhibition and where the dosage does not exceed 0.10 Gy. We examined the absorbed dose of the edible tissues in eggs when sterilizing their shells with LEEB. The irradiation conditions with these EB were evaluated by experimentation and Monte Carlo simulation. The resulting eggshell depth dose distributions indicate that the acceleration voltages between 80 and 200 kV were optimal for eggshell sterilization. It was also found that the acceleration voltages between 80 and 150 kV were suitable for the reduction of dosage to the egg's edible tissues (≤ 0.10 Gy). Furthermore, the contribution of bremsstrahlung to the edible tissues decreased as the energy of the EB was lowered.

Conflict of Interest

No potential conflict of interest relevant to this article was reported.

Author Contribution

Conceptualization: Kataoka N, Kawahara D. Data curation: Kataoka N, Kawahara D, Sekiguchi M. Formal analysis:

Kataoka N, Kawahara D, Sekiguchi M. Funding acquisition: Kataoka N. Methodology: Kataoka N. Project administration: Kataoka N. Writing - original draft: Kataoka N. Writing - review & editing: Kataoka N. Investigation: Kataoka N, Kawahara D, Sekiguchi M. Resources: Kataoka N. Supervision: Kataoka N, Sekiguchi M.

References

1. Roberts K. *Salmonella* outbreak from eggs sickens 38 in 7 states [Internet]. New York, NY: NBC News; 2018 [cited 2021 Feb 1]. Available from: <https://www.nbcnews.com/health/health-news/salmonella-outbreak-egg-sickens-38-7-states-n916446>.
2. Ministry of Health, Labor and Welfare. Number of incidents and patients from food poisoning by causative food and preparing facility [Internet]. Tokyo, Japan: Ministry of Health, Labor and Welfare; 2007 [cited 2021 Feb 1]. Available from: https://www.mhlw.go.jp/stf/seisakunitsuite/bunya/kenkou_iryuu/shokuhin/syokuchu/04.html.
3. Ministry of Agriculture, Forestry and Fisheries. Handbook of egg production and hygiene management (reference): director version [Internet]. Tokyo, Japan: Ministry of Agriculture, Forestry and Fisheries; 2012 [cited 2021 Feb 1]. Available from: <https://www.maff.go.jp/j/syouan/seisaku/handbook/pdf/sairanshido-sha.pdf>.
4. Ministry of Health, Labor and Welfare. Safety management of grading and packing center for egg [Internet]. Tokyo, Japan: Ministry of Health, Labor and Welfare; 1998 [cited 2021 Feb 1]. Available from: https://www.jz-tamago.co.jp/wp/wp-content/uploads/2020/03/E05_3_3.pdf.
5. Tabata T, Andreo P, Shinoda K. An algorithm for depth-dose curves of electrons fitted to Monte Carlo data. *Radiat Phys Chem*. 1998; 53:205–215.
6. Rogers DW, Bielajew AF. Differences in electron depth-dose curves calculated with EGS and ETRAN and improved energy-range relationships. *Med Phys*. 1986;13(5):687–694.
7. Hertwig C, Meneses N, Mathys A. Cold atmospheric pressure plasma and low energy electron beam as alternative nonthermal decontamination technologies for dry food surfaces: a review. *Trends Food Sci Technol*. 2018;77:131–142.
8. Federal Register Volume 65, Number 141 (Friday, July 21, 2000) page 45280 [Internet]. Washington, DC: National Archives and Records Administration; 2000 [cited 2021 Feb 1]. Available from: <https://www.govinfo.gov/content/pkg/FR-2000-07-21/pdf/FR-2000-07-21.pdf>.
9. Tellez IG, Trejo RM, Sanchez RE, Cenicerros RM, Luna QP, Zazua P, et al. Effect of gamma irradiation on commercial eggs experimentally inoculated with *Salmonella* enteritidis. *Radiat Phys Chem*. 1995;46:789–792.
10. Ministry of Health, Labor and Welfare. Standards for food and food additives, etc. (Part 1. Food) [Internet]. Tokyo, Japan: Ministry of Health, Labor and Welfare; c2020 [cited 2021 Feb 1]. Available from: https://www.mhlw.go.jp/stf/seisakunitsuite/bunya/kenkou_iryuu/shokuhin/jigyousya/shokuhin_kikaku/370b.html.
11. Min B, Nam KC, Jo C, Ahn DU. Irradiation of shell egg on the physicochemical and functional properties of liquid egg white. *Poult Sci*. 2012;91:2649–2657.
12. Pages L, Bertel E, Joffre H, Sklavenitis L. Energy loss, range, and bremsstrahlung yield for 10-keV to 100-MeV electrons in various elements and chemical compounds. *At Data Nucl Data Tables* 1972;4:1–27.
13. Sato T, Iwamoto Y, Hashimoto S, Ogawa T, Furuta T, Abe SI, et al. Features of particle and heavy ion transport code system (PHITS) version 3.02. *J Nucl Sci Technol*. 2018;55:684–690.
14. Kataoka N, Sekiguchi M, Kawahara D. Dose evaluation for sterilization of shell egg using low energy electron beam. *Radioisotopes*. 2020;69:163–170.