RECENT DEVELOPMENTS IN RADIATION STERILIZATION OF FOODS[1]

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Research efforts during the past decade indicate that ionizing radiations may be used in certain industrial processes. Unique among these processes is that of cold sterilization of foods, drugs, and pharmaceuticals. Discussion of the applicability of the various types of ionizing radiations available for such purposes and a brief discussion of the theory of the mode of action of ionizing radiations are presented. Major problems involved in this method of processing are discussed. The results of some of the more pertinent research studies are summarized.

The results of research efforts in the past ten years indicate that certain industrial processes may be carried out by utilizing high-voltage X-rays or cathode rays. Unique among these processes is that of cold sterilization of foods, drugs, and pharmaceuticals. For over fifty years it has been known that ionizing radiations have bactericidal effects. In fact, research in this direction was begun shortly after the discovery of X-rays by Roentgen. However, only within the past ten years, when large sources of ionizing radiations have been developed, has intensive research in this field been carried out. The increased interest in this subject is indicated by the fact that in 1948 only two or three laboratories were engaged in research on sterilization by ionizing radiations, whereas now, in 1954, ten or twelve laboratories are actively engaged in studies of this nature.

The purpose of this paper is to describe briefly the means by which microbes are destroyed by ionizing radiations and to discuss the present-day status of these studies.

Relative Radiosensitivities of Various Forms of Life

During the past few years it has been shown that all kinds and types of microorganisms can be destroyed by ionizing radiations. Perhaps the most important single factor in the energy requirements in radiation sterilization is the radiation sensitivity of the particular species of microorganisms under consideration. The resistance of a given species of microorganism to ionizing radiations parallels, in general, its resistance to conventional heat processing.

The lethal doses of cathode rays for various microorganisms are indicated in Figure 1[8]. It will be noted that spore-forming bacteria are more resistant to ionizing radiations than non-spore-forming types and that mold spores are less resistant than bacterial spores.

Figure 2, based on examination of results reported in the literature, also presents some information on the relative radiosensitivities of various forms of life, indicating the dose levels of ionizing radiations required for various biological effects. In general, the lower the order in the plant and animal kingdom, the greater is the resistance of the organism to ionizing radiations.

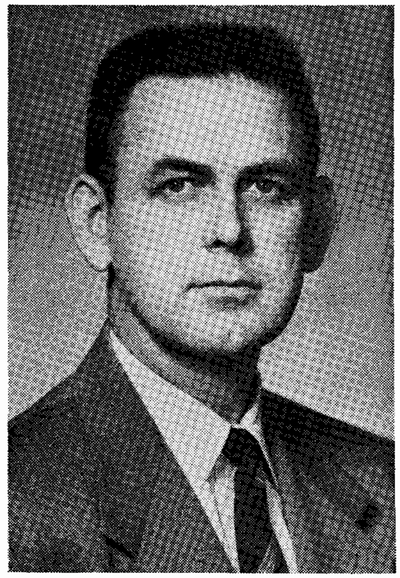
Mode of Action of Ionizing Radiations

As in the case of conventional heat processing, the destruction of microorganisms by ionizing radiations appears to be a first-order reaction. A number of investigators have shown that destruction of bacteria or of other single-celled organisms is effected by the passage of a single ionizing particle through or near the cell.[4] Such passage, which is called a "hit", causes ionization and the subsequent death of the organism.

With increase in the dose of ionizing radiations, the number of viable organisms decreases in a geometric progression. In other words, the survival curve is exponential, that is, the number of organisms that withstand a given exposure to ionizing radiations is an exponential function of dose. Accordingly, the number of organisms surviving a given dose may be calculated according to Eq. 1:

$$n = n_0 e^{-D/D_0} \qquad (1)$$

where the fraction surviving dose D is n/n_0 and where n_0 is the initial number of organisms, n is the number of organisms surviving dose D, and D_0 is the dose required to score an average of one hit per organism[4]. D_0 is conveniently defined as the mean lethal dose, the inactivation



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From 1940 to 1943, Mr. Miller engaged in county health work in Columbus County, North Carolina. In 1943, he was commissioned into the United States Public Health Service and assigned to milk sanitation activities in Tennessee. In 1944-1946 he was with UNRRA and saw duty in Egypt and Greece. After serving as Milk and Food Consultant in the Chicago Regional Office of the Public Health Service from 1946 to 1949, he was assigned to the Interstate Carrier Branch, Division of Sanitation, Public Health Service, Washington, D.C., to participate in the development of the series of handbooks relating to sanitation on interstate carriers. In 1951, he became a member of the staff of the Milk and Food Branch, Public Health Service.

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dose, or the 37 percent dose ($e^{-1} = 0.368$).

This theory regarding the mode of action of ionizing radiations in the destruction of bacteria is known as the "direct hit" or target theory. This direct mechanism of destruction is recognized by the following criteria:

(a) the survival curve is exponential;

(b) destruction is independent of dose rate, that is, the effect of a given dose is the same whether the dose is administered rapidly or over a longer period of time;

(c) destruction is independent of temperature; and

(d) the concentration of organisms does not affect the percentage survival.

FREE RADICALS

Although the "direct hit" or target theory is thought to account for most bacterial destruction, some lethal action may be attributed to chemical effects caused by free radicals produced in the solvents that may be present[10]. For example, the reaction of high energy radiation in water may be illustrated as follows.

When water is bombarded by ionizing radiations, a positive water ion is formed and an electron is liberated.

$$H_2O \rightarrow H_2O^+ + e^- \quad (2)$$

The electron reacts with another water molecule and produces a negative water ion.

$$e^- + H_2O \rightarrow H_2O^- \quad (3)$$

The positive water ion dissociates into a hydrogen ion and a free hydroxyl radical.

$$H_2O^+ \rightarrow H^+ + OH^\circ \quad (4)$$

The negative water ion dissociates into a hydroxyl ion and a hydrogen atom.

$$H_2O^- \rightarrow OH^- + H \quad (5)$$

The hydroxyl radical is a strong oxidizing agent and will oxidize any oxidizable solute. The hydrogen atom is a strong reducing agent and will reduce any reducible solute. If no oxidizable or reducible solute is present, these free radicals may recombine to form water again.

$$H + OH^\circ \rightarrow H_2O \quad (6)$$

Knowledge of these reactions is of principal importance in an understanding of the side-effects that may be produced on the sterilized product.

TYPES OF IONIZING RADIATION

A number of different types of ionizing radiation and radiations that cause excitation are available for consideration. All types have destructive effects on microorganisms, but each has its own characteristics and limitations.

Beta rays, which are streams of electrons emitted from a radioactive material, and cathode rays, which are streams of artificially accelerated electrons, have a somewhat limited power of penetration. However, the efficiency of cathode ray production is relatively high, for approximately 75 percent of the energy of the electron beam can be utilized, thereby effecting sterilization in a matter of seconds or fractions thereof.* Although cathode rays penetrate matter less deeply than X-rays of corresponding voltage, their range of penetration is of sufficient magnitude to warrant giving consideration to this type of radiation. The penetration of cathode rays into matter of unit density is approximately 0.5 cm per 1 m.e.v. of energy. Figure 3 indicates the ranges of penetration of cathode rays of different accelerating voltages into matter of unit density[15].

Gamma rays and X-rays, on the other hand, have a relatively great power of penetration into matter. X-rays are produced by the bombardment of a heavy metal target with cathode rays. However, only 3 to 5 percent of the electron energy is used in the production of X-rays. The remainder of the electron energy produces heat in the target. As a result, the sterilization of a No. 2 can of food, for example, would require from 10 to 20 minutes, even with a beam of high current. Hence it becomes questionable whether, with the present types of available equipment, the use of X-rays as a means of sterilizing foods is practicable.

The biological and chemical effects of X-rays are similar to those of cathode rays, and in neither case is any induced radioactivity produced except at voltages in excess of 15 million volts.[5]

Alpha particles are heavy particles consisting of helium nuclei. They produce dense ionization along their paths in matter and readily kill bacteria. However, alpha particles have little power of penetration into matter. In fact, they may be stopped by the thick-

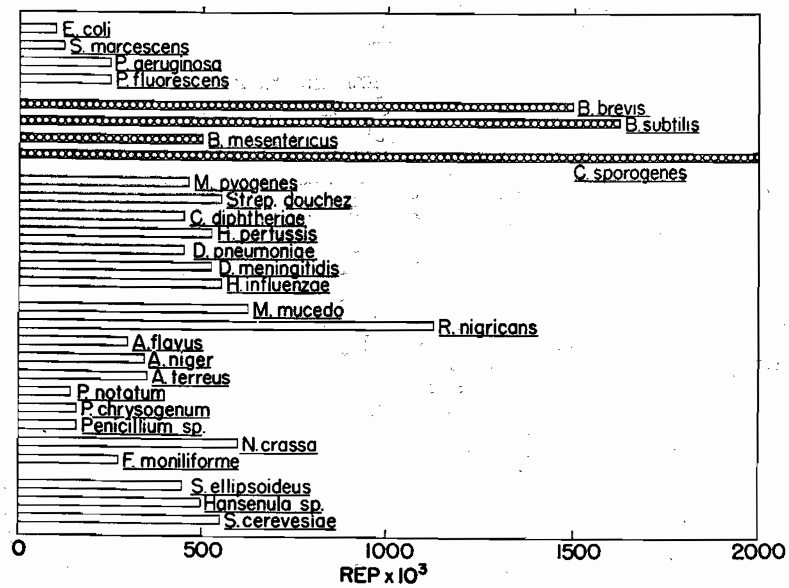

Fig. 1. Doses of cathode rays lethal to bacteria, molds, and yeasts. (Proctor and Goldblith, **8**; reprinted by permission of *Food Technology*.)

*For greater efficiency in the use of cathode rays, solid food products may be treated by "cross firing", that is, by exposing the product to beams of electrons coming from opposite directions through two ports. This can be accomplished by splitting the electron beam and bending it electromagnetically or by using two accelerators located opposite one another. By "cross firing" with present-day machines, samples approximately one inch thick can be sterilized effectively.

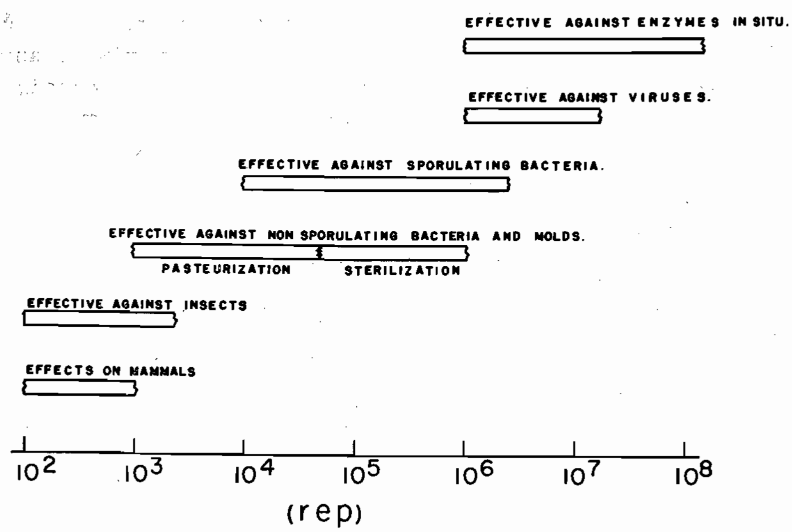

Fig. 2. Dose levels of ionizing radiation required for various biological effects. (Nickerson *et al.*, 7; Reprinted by permission of *Am. J. Pub. Health.*)

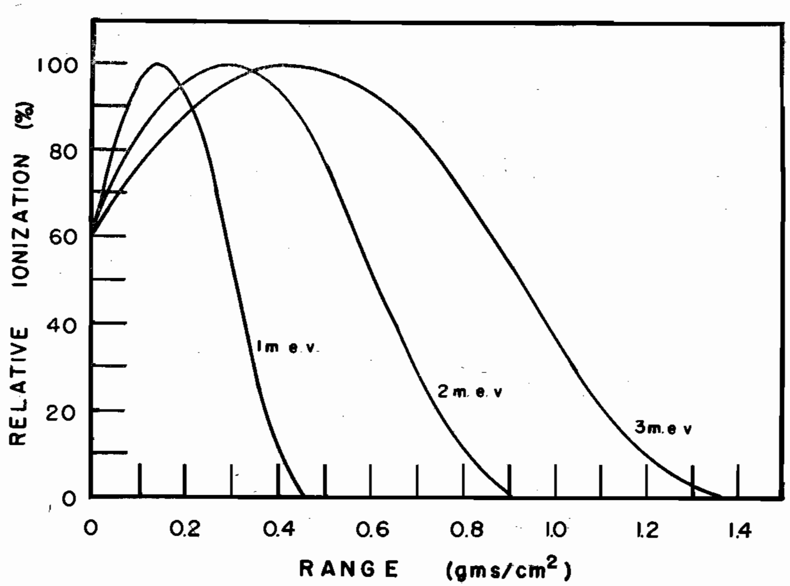

Fig. 3. Ranges of penetration of cathode rays of various voltages into matter of unit density. (Trump, Wright, and Clarke, 15; courtesy of Prof. J. G. Trump and *J. Appl. Physics.*)

ness of a sheet of paper and, therefore, should not be considered for the sterilization of foods.

Neutrons are unchanged subatomic particles. They have a relatively great power of penetration into matter. Because neutron production is extremely inefficient and because neutrons create induced radioactivity, this type of radiation does not appear feasible for food sterilization.

The inability of ultraviolet light to penetrate matter to any appreciable depth makes this type of radiation unsuitable for food sterilization under present-day considerations.

On the basis of the above considerations as well as on the basis of the present availability of sources of ionizing energy in the quantities required for sterilization, it appears that cathode rays are the only type of ionizing radiation possessing the three characteristics of efficiency, safety, and practicability. Gamma rays may hold promise for use in sterilization at some later date, dependent on the availability of adequate quantities of radioactive fission products.

SOURCES OF IONIZING RADIATIONS

The sources of ionizing radiations include both machines and radioactive isotopes. Among the cathode ray generators are the Van de Graaff Electrostatic Accelerator[14], the Capacitron[2], and the Resonant Transformer[3]. Generators with accelerating voltages of 3 m.e.v. are available today, which make possible the sterilization of solid food packs 1 inch in thickness. Generators of 4 or 5 m.e.v. may be available in the near future.

Other potential sources of ionizing radiations are the by-products of atomic fission that emit gamma rays. These fission products or isotopes retain a portion of the energy of fission and release this energy by radioactive decay. Until adequate technology for processing can be developed, these fission products have been stored in underground tanks as a part of the warborn expediency.

PROBLEMS TO BE SOLVED

Present-day problems in the use of ionizing radiations for food processing are concerned both with the source of such radiations and with the effects of these radiations on the food product. All these problems are being subjected to intensive investigation. In any consideration of the industrial utilization of particle accelerators for sterilization, the reliability of the equipment as it relates to power output must be given considerable attention. Furthermore, accelerating voltages of the order of magnitude required to handle reliably conventional containers of foods have not as yet been achieved. In the case of isotopic sources of energy, there are problems to be solved in the preparation of these materials in megacurie quantities, in the obtaining of such materials in the appropriate condition of decay (or age) required for sterilization, and in the packaging of these materials so as to facilitate their safe use.

When sterility in food products is effected by ionizing radiations, undesirable flavor changes occur in many instances. The theory has been postulated that the flavor molecules in food are true chemical compounds. As indicated previously, when foods are subjected to ionizing radiations of high energy, hydroxyl radicals and free hydrogen atoms are produced. If a flavor molecule in a food is oxidizable, it can react with the hydroxyl radical present and produce an oxidized off-flavor. If, however, a compound is introduced into the food and such a compound shows a greater affinity for the particular radical involved than do the compounds already present in the food, the development of off-flavor may be obviated to a great extent.[10, 11] This is illustrated by the following formula, in which FRA means free-radical acceptor:

$$\text{Flavor molecule} + OH^{\circ} + \text{FRA (reduced)} \rightarrow \text{Flavor molecule} + \text{FRA (oxidized)} + OH^{-} \quad (7)$$

Research has been instituted in which so-called free-radical acceptors have been used as a means of obviating flavor changes. Some of the results obtained have been encouraging. It has been found, for example, that ascorbic acid or vitamin C and its analogues will, in many cases, act as free-radical acceptors and prevent flavor changes in foods during and after irradiation.[11] Ascorbic acid is an illustration of one type of compound that, when added to food, would in itself have no toxic effect.

Studies are also being conducted on other methods of minimizing undesirable changes in irradiated products. These studies include observations on the effects of irradiation of foods in the frozen state and at low oxygen tension.[9] Studies have also been made on the use of extremely short pulses of cathode rays as a possible means of overcoming these changes.

Figure 4 shows the protective effect of D-iso-ascorbic acid in low concentration when the amino acid histidine is irradiated with cathode rays in the presence of this free-radical acceptor. Figure 5 shows the protective effect of sodium D-iso-ascorbate when an enzyme, crystalline pepsin, in acetate buffer is irradiated in its presence. Both pepsin and histidine are biochemical substances typical in foods.

A number of other factors must be studied before ionizing radiations may be used commercially for food processing. Toxicological studies must be made, preferably in collaboration with the various

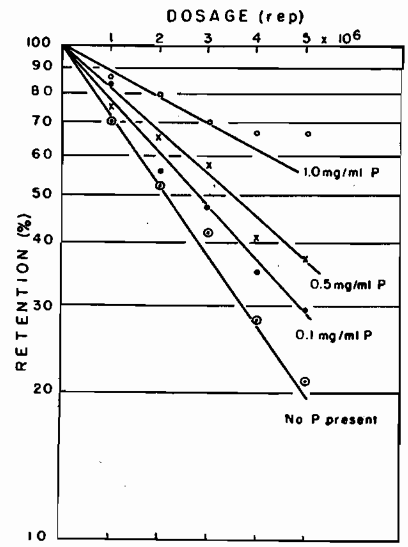

Fig. 4. Effects of cathode rays on aqueous solutions of histidine HCl (1.0 mg/ml) in the presence of varying concentrations of D-isoascorbic acid. (P is protector) (Proctor *et al.*, 11; Reprinted by permission of *Food Technology.*)

government regulatory agencies that would be concerned with the particular application. Studies must also be made to determine whether irradiation causes losses of the nutrient elements of foods and the effect that various storage temperatures have on the keeping qualities of irradiated foods.

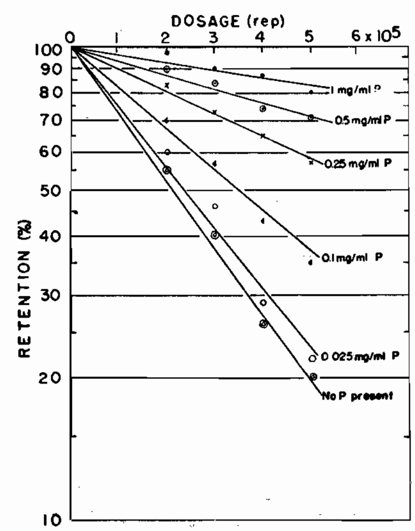

Figure 5. Effects of cathode rays on pepsin (1.0 mg/ml) in acetate buffer (pH 4.3) in the presence of different amounts of sodium D-isoascorbate. (P is protector) (Proctor *et al.*, 10; reprinted from *Nucleonics* April 1952; copyright McGraw Hill Publishing Co., 1952).

Although some studies on these factors have been made and are being made, the results obtained thus far do not offer conclusive answers to the problems mentioned in respect to all types of foods.

SOME DEVELOPMENTS IN RADIATION STERILIZATION

Investigations carried out at the Massachusetts Institute of Technology and elsewhere have shown that ionizing radiations are capable of killing any kind of microorganism in any medium or menstruum, and in any type of container, provided the physical dimensions of container and contents are within the practical limits of penetration of the particular type of radiation used.[7]

Successful sterilization of meat, fish products, vegetables, and spices[12] has been effected with ionizing radiations.[9] As mentioned earlier, however, a number of problems associated with the sterilization of these foods by ionizing radiations are still to be solved.

Liquefied whole eggs inoculated with three species of *Salmonella* have been successfully sterilized with ionizing radiations at a dose level of 300,000 rep.* No organoleptic preference was found between liquid eggs irradiated at this level and then spray-dried, and untreated liquid eggs that had been spray-dried.[13]

A number of investigators have shown that milk can be pasteurized by comparatively low doses of high-energy cathode rays.[9] However, milk and milk products appear to be extremely sensitive to ionizing radiations, and irradiation of these products is frequently accompanied by undesirable changes in flavor.

Insects are relatively sensitive to ionizing radiations. They can be killed by doses of less than 100,000 rep, and their reproduction is prevented with even lower doses.[7] The elimination of insect infestation in grain by application of ionizing radiations may prove economically feasible.

Trichinae in pork can be destroyed with low doses of high-voltage cathode rays.[1]

In addition to these applications related to foods, investigations indicate that ionizing radiations may find service in the cold sterilization of heat-labile drugs and biologicals, in the sterilization of blood, and in the sterilization of human organs for subsequent transplant. The Department of Food Technology at the Massachusetts Institute of Technology has been cooperating with surgeons at the Harvard Medical School with respect to this latter application.[6] Aortae sterilized in this manner have now been successfully transplanted into human beings. Today approximately twenty persons have had sections of their aortae replaced with tissues that were removed from fresh cadavers and sterilized with ionizing radiations.

If the present-day knowledge of radiation sterilization of foods is compared with what was known five years ago, it is apparent that considerable progress has been made. Much research is yet to be done, however, before food processing with ionizing radiations becomes an industrial reality. Studies on the fundamental mechanism of the action of ionizing radiations on foods and food components are being continued, in an effort to determine the cause of undesirable side effects and the means of preventing them.

*This expression "rep" is the density of energy, equivalent to 93 ergs absorbed by 1 gram of ordinary tissue.

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ABSTRACTS OF PAPERS

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and efficient for the 20-year period. To get this type of service from a hardening room it must be constructed correctly from the very beginning. Detail description of the building of a hardening room to last not only 20 years but longer was given. First described was the construction of the outer walls—the thickness, type of materials to use, and how to build them. Similar details followed for the floor and other parts of the hardening room. The heaving of floors in hardening rooms was discussed and also the three different types of construction that may be used to avoid heaving. After the details of constructing the shell were given, the best type of insulation for the floors, walls, and ceiling were given and finally a discussion of the different types of doors available and the ones that give the most satisfactory service.

A bound book containing mimeographed copies of the original papers reported in these abstracts may be obtained by sending a check, money order, or draft for $1.50 to Dr. Carl W. Hall, Dept. of Agricultural Engineering, Michigan State College, East Lansing, Michigan.

MARKET MILK CONFERENCE PURDUE UNIVERSITY

Lafayette, Ind., April 13. About 95 percent of all fluid milk sold in Indiana today is Grade A pasteurized milk, John Taylor, dairy division director of the Indiana State Board of Health, said Tuesday at Purdue University.

He told more than 85 dairy plant operators and dairy products manufacturers at a Market Milk Conference that this top quality milk is being inspected and supervised by cities operating under Grade A milk ordinances.

Taylor went on to say that current proposals in Indiana call for dairy farm and dairy plant sanitarians to be licensed so they can offer relief to dairies outside the jurisdiction of cities having Grade A milk ordinances. They would help dairies to qualify their supplies and label their products Grade A. He also thought the time was drawing near when other products such as ice cream may be made from inspected products and qualified for labeling as Grade A.

G. P. Gundlach, president of G. P. Gundlach and Company, Cincinnati, Ohio, said that dairy products offer one of the greatest food bargains today for the 41 million housewives who are trying to buy food for their families at the lowest possible price.

Speaking on the preparation of cultured buttermilk, Dr. C. E. Parmelee, of Purdue's dairy department, told dairy plant operators that the incubation temperature for this product should be maintained at 70 to 72 degrees F. to keep the culture organisms in balance and to produce a fine flavor.

Other speakers on the Tuesday program were Dewey Shaw, of Kraft Foods Company, Chicago; C. E. French, Purdue agricultural economist; and members of the Purdue dairy department staff.

The milk conference was held in cooperation with the Indiana Dairy Products Association.