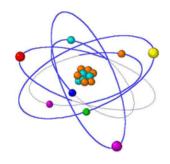


Radiation Biology, Protection and Applications (FS2018)



Food Irradiation, Radioisotope Batteries (Week 11)

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26.11. 2018



Food Irradiation, Radioisotope Batteries: Outline

- Food Irradiation
 - Advantages
 - Sources
 - Dose Requirements
 - Biological Effects
 - Some Practical Applications
- Sterilization
- Radionuclide Batteries (RB):
 - Sources
 - Radioisotope Thermoelectric Generator
 - Examples of Applications
 - Summary



Food Irradiation: Introduction

- ☐ Food irradiation (FI) is the process of exposing food, either prepackaged or in bulk, to controlled levels of certain types of ionizing radiation to:
 - increase storage life of food
 - reduce postharvest food losses
 - inactivate specific food-borne pathogenic organisms
- □FI is one of the most thoroughly and intensively investigated methods of food preservation. Nevertheless there is some controversy regarding its safety.
- The ionizing radiation applied in FI is limited to high-energy EM-radiation (γ -rays or X-rays) with energies up to 5 MeV or high-energy electrons up to 10 MeV. These radiations are chosen because:
 - They produce the desired effects with respect to the food.
 - They do not induce radioactivity in foods or their packaging materials.
 - They are available in quantities and at costs that allow practical uses of the process.



Food Irradiation: Advantages

- □ Radiation treatment can be considered as nonthermal processing, because even at largest absorbed doses (~50 kGy), the amount of energy is equivalent to 50 J. Thus frozen food can be treated.
- ☐ Irradiation can be applied through any packaging materials including those that cannot withstand heat. Thus recontamination or reinfestation of the product can be avoided.
- ☐ The useful effects of ionizing radiation are summarized in the table. In some cases even improvement of certain functional or sensory quality characteristics of food can be achieved with irradiation.

Useful Effects of Irradiation as a Food Processing Treatment

Effects	Results
Inhibition of sprouting of tubers and bulbs	Increased storability
Decrease of after-ripening and delaying senescence of some fruits and vegetables	Increased shelf life
Killing or sterilizing stored product insects	Insect disinfestation of food
Inactivation of parasites transmissible by food	Prevention of food-borne parasitic diseases
Inactivation of food-borne microorganisms	Microbial decontamination of food: Increased shelf life and/or prevention of food poisoning



Food Irradiation: Sources

- ☐ Two basic types of radiation sources are used in FI:
 - High-voltage X-ray tubes or e⁻ accelerators; e⁻ have low penetrability (4cm for 10MeV e⁻).
 - γ-ray emitting radionuclides: mainly ⁶⁰Co, also ¹³⁷Cs.
- Typical FI facilities consist of a process chamber, a conveyor system, and control/safety systems.

Comparison of Typical Processing Parameters

	Gamma	X-ray	E-beam		
Typical source power	3.5 MCi	25 kW	35 kW		
Typical processing speed	12 tonnes/ hr at 4 kGy	10 tonnes/ hr at 4 kGy	10 tonnes/ hr at 4 kGy		
Source energy	1.33 MeV	5 MeV	5–10 MeV		
Penetration depth	80-100 cm	80-100 cm	8-10 cm		
Dose homogeneity	High	High	Low		
Dose rate	Low	High	Higher		
Best application	Bulk processing of large boxes or palletized product in shipping cartons in a warehouse environment	Bulk processing large boxes or palletized product in shipping cartons in a warehouse environment	Sequential processing of primary or secondary packaged product in-line or at-line		



Food Irradiation: Typical Dose Requirements

Application	Dose Requirement (kGy)
Inhibition of sprouting of potatoes and onions	0.03-0.12
Insect disinfestation of seed products, flours, fresh and dried fruits, etc.	0.2-0.8
Parasite disinfestation of meat and other foods	0.1–3.0
Radurization of perishable food items (fruits, vegetables, meat, poultry, fish)	0.5–10
Radicidation of frozen meat, poultry, eggs and other foods and feeds	3.0–10
Reduction or elimination of microbial population in dry food ingredients (spices, starch, enzyme preparations, etc.)	3.0–10
Radappertization of meat, poultry, and fishery products	25–60

- The technological feasibility of a FI treatment depends on how much irradiation the food withstands without adversely changing its qualities.
- Not wanted are changes to:
 - the chemical composition
 - the nutritional value
 - sensory properties of the product
- Generally there is a minimum dose requirement (see table).
- Not every mass element of a food must be irradiated (sometimes irradiation of the surface will suffice).



Terms in Food Irradiation

Radappertization:

- The application to foods of a dose of ionizing radiation sufficient to reduce the number and/or
 activity of viable microorganisms to such an extent that very few, if any, are detectable in the
 treated food by any recognized method (viruses being excepted). No microbial spoilage or
 toxicity should become detectable in a food so treated, no matter how long or under what
 conditions it is stored, provided the package remains undamaged. The required dose is
 usually in the range of 25–45 kGy.
- Radappertization is derived from the combination of radiation and Appert, the name of the French scientist and engineer who invented sterilized food for the troops of Napoleon.

■ Radicidation:

 The application to foods of a dose of ionizing radiation sufficient to reduce the number of viable specific non-spore-forming pathogenic bacteria to such a level that none are detectable when the treated food is examined by any recognized method. The required dose is in the range of 2–8 kGy.

■ Radurization:

 The application to foods of a dose of ionizing radiation sufficient to enhance its keeping quality by causing a substantial decrease in numbers of viable specific spoilage microorganisms. The required dose is in the range of 0.4–10 kGy.



Food Irradiation: Biological Effects of Ionizing Radiation (Supplement!)

D₁₀ Values (kGy) of Some Nonsporeforming Bacteria

Bacteria	Nonfrozen Food	Frozen Food	
Vibrio spp.	0.02-0.14	0.04-0.44	
Yersinia enterocolitica	0.04-0.21	0.20-0.39	
Campylobacter jejuni	0.08-0.20	0.18 - 0.32	
Aeromonas hydrophila	0.11-0.19	0.21 - 0.34	
Shigella spp.	0.22 - 0.40	0.22-0.41	
Escherichia coli (incl. O157:H7)	0.24-0.43	0.30-0.98	
Staphylococcus aureus	0.26-0.57	0.29-0.45	
Salmonella spp.	0.18 - 0.92	0.37-1.28	
Listeria monocytogenes	0.20-1.0	0.52 - 1.4	

- □ Primary target of biological effects is the DNA.
- ☐ The radiation dose to kill stored product insects depends on the species and a number of other factors such as age, sex, and stage of development.
- Radiation effects on food-borne parasitic protozoa and helminths are associated with loss of infectivity, loss of pathogenicity, interruption or prevention of completion of life cycle, and death of parasites.
- The actual percentage of cells or microbial population that will be killed depends on various factors:
 - inherent resistance of particular organism,
 - growth stage,
 - environmental factors (temperature, oxygen presence, water content).
- □ The table shows ranges of decimal reduction doses (D₁₀ values) of the most important pathogens.



Food Irradiation: Radiation Induced Chemical Changes

- As water is present in almost all foods, water radiolysis takes place producing:
 - the very reactive transient species: •OH (oxidizing agent), e_{ag} (reducing agent), •H
 - stable end-products: H₂, H₂O₂
- □ The presence or absence of oxygen can have an important influence on the course of radiation induced changes of food components: oxygen can add to some of the radicals to form •RO₂ (peroxy radicals).
- The radiolysis of water is pH-dependent.
- Also the temperature during irradiation influences the chemical changes.
- Other major constituents of food are carbohydrates, proteins, and lipids.
 - Irradiation of sugars and polysaccharides can cause changes in the physical properties.
 - Irradiation effects on amino acids (proteins) and consequently enzymes are rather small.
 - Irradiation of fats can lead to a multitude of products.



Food Irradiation: Types of Food That are Being Irradiated

Types of Food	Radiation Dose in kGy	Effect of Treatment
Meat, poultry, fish, shellfish, some vegetables, baked goods, prepared foods	20 - 71	Sterilization. The treated product can be stored at room temperature without spoilage. The treated product is safe for hospital patients who require microbiologically sterile diets.
Spices and other seasonings	Up to maximum of 30	Reduces number of microorganisms and insects. Replaces chemicals used for this purpose.
Meat, poultry, fish	0.1 - 10	Delays spoilage by reducing the number of microorganisms in the fresh, refrigerated product. Kills some types of food poisoning bacteria and renders harmless disease-causing parasites (e.g. trichinae).
Strawberries and some other fruits	1 - 5	Extends shelf life by delaying mold growth.
Grain, fruit, vegetables, and other foods subject to insect infestation	0.1 - 2	Kills insects or prevents them from reproducing. Could partially replace post-harvest fumigants used for this purpose.
Bananas, avocados, mangos, papayas, guavas and certain other non-citrus fruits	1.0 maximum	Delays ripening.
Potatoes, onions, garlic, ginger	0.05 - 0.15	Inhibits sprouting.
Grain, dehydrated vegetables, other foods	Various doses	Desirable physical changes (e.g. reduced rehydration times).



Food Irradiation: Some Practical Applications

- Control of sprouting and germination of vegetable crops.
- Insect control in stored foods.
- Irradiation as a quarantine treatment.
- Parasite disinfection.
- Extension of shelf life of fresh fruits and vegetables.
- Irradiation of fresh meat and poultry.
- Irradiation of fish and shellfish products.
- ☐ Irradiation of minimally processed or ready-to-eat foods.
- Radiation decontamination of dry food ingredients.
- □ Radiation sterilisation of food (radappertisation).



Combination Processes in Food Irradiation

- When irradiation is used with other preservative or antimicrobial factors, the global efficiency is reinforced through additive or synergetic action.
- The combination of irradiation with mild heat treatment has a number of advantageous effects, which may be due to the inability of cells to repair radiation damage because heating might inactivate repair enzymes.
- In the field of muscle foods, the use of marination before irradiation reduced the dose necessary to eliminate Salmonella in poultry.
- Some antimicrobial additives, especially the natural ones and GRAS (generally recognized as safe) preservatives can be usefully combined with irradiation to reduce dose requirements.
- Some antioxidants have also been used to prevent the undesirable oxidative effects in irradiated foods.



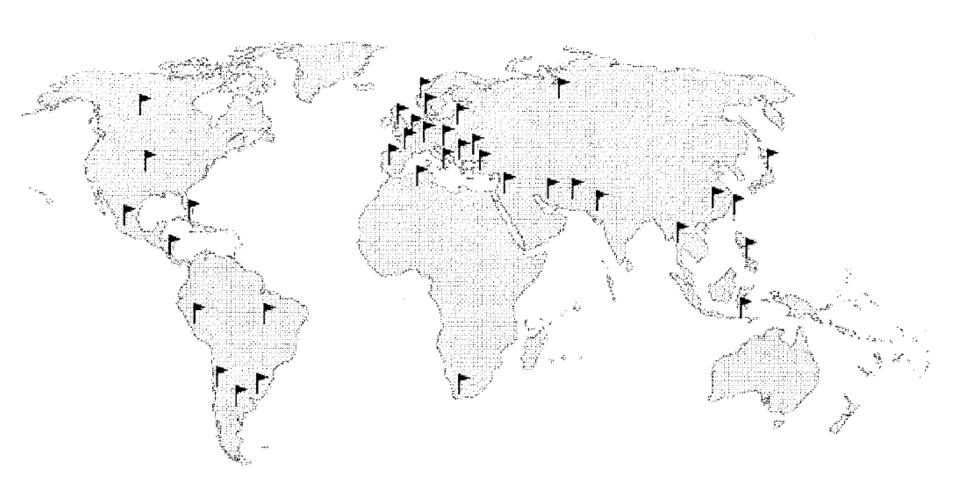
Legislation of Food Irradiation



- Legislatory authorities require that irradiated food products be labeled. Generally the international food irradiation symbol, the so-called Radura logo is required with a statement that the product has been intentionally subjected to radiation.
- In 1984 the International Consultative Group on Food Irradiation (ICGFI) was established under the aegis of FAO, IAEA, and WHO. The ICGFI:
 - promulgates harmonized regulations on food irradiation in developing countries,
 - issues numerous publications relating to food irradiation including codes of good irradiation practice for various classes of foods, and compilations of technical data for authorization and control of food irradiation.



Countries with Approval of Food Irradiation





Sterilization

- As you want to kill the cell, sterilising doses are of the order of 10 kGy to 30 kGy delivered over a period of hours to days.
- In industry sterilisation is mainly applied to disposable medical products and has the following advantages:
 - In a well designed plant, the radiation reaches the whole of the product at a dose rate which can be controlled and accurately monitored.
 - Unlike other sterilisation processes, radiation technology is well adapted to continuous operation and can be undertaken at normal temperature and after final packing.
- Three types of ionising radiation are used: ⁶⁰Co γ-radiation, electron beams and bremsstrahlung from high energy electron accelerators (3 to 6 MeV).
- The irradiated products include medical gloves, syringes and a range of pharmaceutical products.
- Other potential applications of radiation disinfection that have been demonstrated are the treatment of industrial waste and sewage sludge.



Radionuclide Batteries: Introduction

- ☐ In radionuclide batteries (also referred to as "Isotope Batteries") the energy from the decay of the radionuclide is transformed to electric energy within one or several steps.
- ☐ The advantages of radionuclide batteries are:
 - Energy is produced over longer periods of time without a need for maintenance.
 - They have a relatively high energy output related to mass and volume of the radionuclide.
- □ Radionuclide batteries are mainly used as maintenance-free energy sources in satellites, remote meteorological stations and oceanography.
- ☐ There are several radionuclides that can be used as sources for batteries.
- For the conversion of the decay energy to electric energy several methods exist (overview):
 - Direct conversion by use of charging potentials or by betavoltaic conversion.
 - Indirect conversion is mostly based on **thermoelectric conversion** consisting of 2 steps: decay energy is transformed to heat, which is transformed to electric energy by a thermoelement (SEEBECK-effect).
 - Other indirect methods involving two steps are: thermionic, thermophotovoltaic or photoelectric conversion.
 - Dynamic converters apply 3 steps (radiation energy → heat → mechanical energy → electricity).



Radionuclide Batteries: Sources

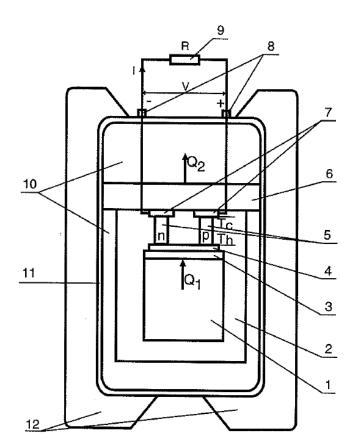
Radionuclides for application in radionuclide batteries.

Radionuclide	Half-life [y]	Radiation	Production
 ³ Н	12.323	β-	$^{6}\mathrm{Li}(\mathrm{n},\alpha)^{3}\mathrm{H}$
¹⁴ C	5730	β-	$^{14}N(n,p)^{14}C$
⁶⁰ Co	5.272	β^- , γ	59 Co(n, γ) 60 Co
⁶³ Ni	100	β^-	62 Ni $(n,\gamma)^{63}$ Ni
⁸⁵ Kr	10.76	β-, γ	Fission product
⁹⁰ Sr	28.64	β^-, γ (90 Y)	Fission product
¹⁰⁶ Ru	1.02	β^- , γ (¹⁰⁶ Rh)	Fission product
¹³⁷ Cs	30.17	β^- , γ	Fission product
¹⁴⁴ Ce	0.78	β^- , γ	Fission product
¹⁴⁷ Pm	2.62	β^-, γ	Fission product
¹⁷⁰ Tm	0.35	β^- , (ε) , γ , e^-	$^{169}{ m Tm}({ m n},\gamma)^{170}{ m Tm}$
¹⁷¹ Tm	1.92	β^- , γ	$^{170}\mathrm{Er}(\mathrm{n},\gamma)^{171}\mathrm{Er}\stackrel{\beta-}{\rightarrow}\ ^{171}\mathrm{Tm}$
²⁰⁴ Tl	3.78	$\beta^-, (\varepsilon)$	$^{203}\text{Tl}(n,\gamma)^{204}\text{Tl}$
²¹⁰ Po	0.38	α,γ	Decay product of ²³⁸ U
²²⁸ Th	1.913	α, γ	Decay product of ²³² Th
$^{232}{ m U}$	68.9	α ,(sf), γ	230 Th $(n,y)^{231}$ Th $\overset{\beta-}{\longrightarrow}$ 231 Pa; 231 Pa $(n,y)^{232}$ Pa $\overset{\beta-}{\longrightarrow}$ 232 U
²³⁸ Pu	87.74	α ,(sf), γ	$\begin{cases} {}^{237}\mathrm{Np}(\mathrm{n},\gamma)^{238}\mathrm{Np} \stackrel{\beta-}{\to} {}^{238}\mathrm{Pu} \\ \mathrm{Decay product of } {}^{242}\mathrm{Cm} \end{cases}$
²⁴¹ Am	432.2	α ,(sf), γ	240 Pu $(n,\gamma)^{241}$ Pu $\stackrel{\beta-}{\longrightarrow}$ 241 Am
²⁴² Cm	0.45	α ,(sf), γ	241 Am $(n,\gamma)^{242}$ Am $\stackrel{\beta-}{\longrightarrow}$ 242 Cr
²⁴⁴ Cm	18.10	α ,(sf), γ	243 Am $(n,\gamma)^{244}$ Am $\stackrel{\beta-}{\rightarrow}$ 244 Cr

- ☐ The table gives a survey of radionuclides applicable in radionuclide batteries.
- ☐ The following selection criteria are important:
 - The half life should be long compared to the desired operation time (usually ≥10y).
 - The power output per mass (specific power) should be as high as possible, which is achieved if:
 - the half life is not too long ($<10^3$ y)
 - the energy of the radiation is high
- \Box Alpha emitters have the advantage that the decayenergy is relatively high and that α-particles are effectively absorbed.
- \Box Radionuclides decaying by subsequent emission of several α particles, such as ²³⁸Pu and ²³²U, are most favourable:
 - 238 Pu: $T_{1/2}$ =87.74a, E_{α} =5.50, 5.46, ... MeV, specific power = 0.56 W_{therm} /g
 - ²³²U: T_{1/2}=68.9a, E_q=5.32, 5.26, ... MeV



Radioisotope Thermoelectric Generator (RTG or RITEG)



Principal scheme of RITEG: (1) RHS, (2) thermal insulation, (3) hot heat conductor, (4) commutating plate of the hot junctions, (5) semiconductor branches with different types of conductivity, (6) cold heat conductor, (7) commutating plates of cold junctions, (8) power points, (9) external electric resistance, (10) biological shield, (11) casing, and (12) cooling ribs. Designations: T_h , T_c are the temperatures of hot and cold junctions, respectively; Q_1 , Q_2 are heat power emitting by RHS and dissipated heat power, respectively.

- ☐ The radioisotope fuel is held in hermetically sealed container or ampoule called radionuclide heat source (RHS).
- ☐ The conversion of heat to electricity is based on the SEEBECK-effect:
 - a temperature gradient between two branches of an electric circuit composed of different conductors (or semiconductors) will lead to a thermoelectric force, i.e., an electric current will flow in the loop.
- As a source typically the ceramic form of ²³⁸PuO₂ is used.



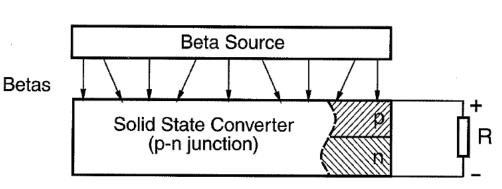
Other Conversion Methods (1)

- The principle of thermionic conversion is that of a (classical) diode:
 - A cathode (emitter) emits electrons that are collected at the anode (collector).
 - Alloys of W, Re, Mo, Ni or Ta are used as emitters.
 - The diodes operate at a temperature of about 2200 K.
 - The efficiency varies between about 1 and 10% (depending on the power).
 - Radionuclides of high specific power are needed, such as ²³⁸Pu, ²³²U, ²²⁷Ac, ²⁴²Cm.
 - Prototypes of 0.1 to 1kW have been developed.
- The principles of thermophotovoltaic conversion are:
 - Heat is converted to electric energy by means of a infrared-sensitive photoelement (e.g. Ge diodes).
 - The device must be cooled effectively because the efficiency decreases drastically as the temperature rises.
 - Adequate for power levels between 10 W and 1 kW.
 - The efficiency is relatively low (up to about 5%).



Other Conversion Methods (2)

- ☐ The principles of photoelectric (or radiophotovoltaic) conversion are:
 - First radiation energy is converted to light by means of luminescent substances, then to electric energy by means of photoelements.
 - Radiative decomposition of the luminophore limits the number of radionuclides applicable.
 - Alpha emitters are unsuitable, the most suitable β emitter is ¹⁴⁷Pm.
 - Construction: radionuclide and luminophore are mixed in a ratio of about 1:1 and brought between two photoelements in form of a thin layer.
 - Powers of the order of 10μW per cm² are obtained.
 - The efficiency is very low (0.1 to 0.5%). Therefore this type has no technical significance.
- ☐ The principles of **betavoltaic conversion** are (**direct method**):
 - In a semiconductor incident radiation generates free charge carriers, that are separated in the p,n-barrier of the semiconductor.
 - Sources must be low E β-emitters.
 - Suitable are: ¹⁴⁷Pm, ¹⁴C, ⁶³Ni, T.
 - Efficiencies of about 4% are obtained.



Scheme of direct-conversion betavoltaic.



Other Conversion Methods (3)

- ☐ The principles of alphavoltaic conversion are (direct method):
- An alpha voltaic battery includes at least one layer of a semiconductor material comprising at least one p/n junction, at least one absorption and conversion layer on the at least one layer of semiconductor layer, and at least one alpha particle emitter.
- The absorption and conversion layer prevents at least a portion of alpha particles from the alpha particle emitter from damaging the p/n junction in the layer of semiconductor material. The absorption and conversion layer also converts at least a portion of energy from the alpha particles into electron-hole pairs for collection by the one p/n junction in the layer of semiconductor material.



Application Examples (1): Characteristics of RITEGs

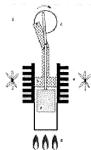
- □ RITEGS are used in heart pacemakers, autonomous power sources for optical and radio beacons, meteorological stations, deep-sea buoys, and spacecraft electronics.
- ☐ In several Apollo missions the SNAP-27 device was used (Systems Nuclear Auxiliary Power).
- □ SNAP-7B, e.g., is for use in shore light stations.

Characterization of RITEGs

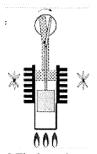
		Power, W					_	
Designation, Country	Thermal	Electrical	Voltage, V	Efficiency, %	Radionuclide	Fuel Loading, Ci (g)	Service Life, Years	Mass, kg
SNAP-3B7, U.S.	52	2.7	3.5	5.2	²³⁸ Pu	1,600	5	2.1
SNAP-7B, U.S.	1440	68	12	4.7	90Sr	225,000	10	2090
SNAP-7C, U.S.	256	11.6	5	4.5	90Sr	40,000	10	850
SNAP-11, U.S.	396	19	3	4.8	242Cm	(6.2)	0.5	7.55
,						. ,		(without protection)
SNAP-17, U.S.		30	 .		90Sr	· —	5-10	11.4
SNAP-27, U.S.	_	63			²³⁸ Pu		1	14
RTG-3, U.S.		1	_	_	· 238Pu	_	20	4.4
RIPPLE-1, GB		0.075	_	1.71	90Sr	_		600
Beta-3, USSR	265	12	12	4.5	90Sr	40,000	10	250
Beta-h, USSR	208	10	6	4.8	90Sr	31,000	10	156
G-90-60/40, USSR	1650	60	40	3.6	90Sr	250,000	10	1200
Ritm, USSR	0.2	10⁻³	1	0.5	²³⁸ Pu	_	10	0.050



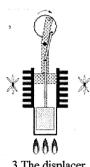
Application Examples (2): Stirling Radioisotope Generator developed at NASA



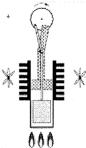
1 Power piston (hatched) has compressed the gas, the displacer piston (grey) has moved so that most of the gas is adjacent to the hot heat exchanger.



2 The heated gas increases its pressure and pushes the power piston along the cylinder. This is the power stroke.



3 The displacer piston now moves to shunt the gas to the cold end of the cylinder.



4 The cooled gas is now compressed by the flywheel momentum. This takes less energy since when it cooled its pressure also dropped.

- The Stirling Radioisotope Generator (SRG) is one of the technologies being developed to provide spacecraft onboard electric power for potential use on future NASA missions.
- ☐ Its principle is that of a dynamic converter:
 - radiation energy → heat
 - heat → mechanical energy
 - mechanical energy → electric energy
- The RHS contains ~ 600 grams of ²³⁸PuO₂ and produces ~250 W of thermal power.
- ☐ The hot-end operating temperature is 650°C.
- A Stirling engine (shown in the figure) transforms the heat into reciprocating motion.
- An alternator produces an AC electrical power output of 60 to 62 W.
- The efficiency was demonstrated to be in the mid 20% range.



Summary

- ⁶⁰Co γ-radiation, electron beams and bremsstrahlung are used to sterilize disposable medical products.
- Gamma rays, X-rays and electron beams are used to irradiate food with the benefits:
 - Increase of the storage life of food.
 - Reduction of postharvest food losses.
 - Inactivation of specific food-borne pathogenic organisms.
- □ Radioisotopes are further applied in radioisotope batteries to produce electricity over longer periods of time without a need for maintenance.



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