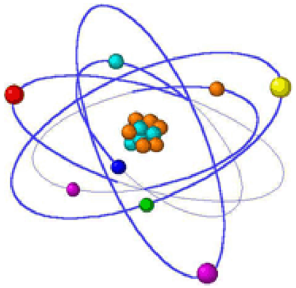
Radioisotope and Radiation Applications (FS2013)



Polymerization, Food Irradiation, Radioisotope Batteries (Week 5b)

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Polymerisation, Food Irradiation, Radioisotope Batteries: Outline

- Introduction to Radiation Chemistry
- Basics:
  - Radiation Induced Reactions
  - Time Scale of Events
  - Chemical Yield
  - Chain Reactions
  - Examples of Radiation Induced Reactions
- Radiation Induced Polymerization
- Food Irradiation
  - Advantages
  - Sources
  - Dose Requirements
  - Biological Effects
  - Some Practical Applications
- Sterilization
- Radionuclide Batteries (RB):
  - Sources
  - Radioisotope Thermoelectric Generator
  - Examples of Applications
- Summary

Introduction: Radiation Chemistry

- Chemical reactions (effects) induced by ionizing radiation are the field of radiation chemistry, which must be separated from radiochemistry (the study of radioactive nuclei).
- Radiation Chemistry and Photochemistry are closely related:
  - Photochemistry: chemical reactions induced by light (energy in the range 1–10eV or wavelength λ in the range 1240–124nm, ($E=hv=hc/\lambda$, $hc=6.62 \cdot 10^{-34}$ Js × $2.9979 \cdot 10^{8}$ m/s = $1.2398 \cdot 10^{-6}$ eVm = 1240 eVnm)).
  - Radiation Chemistry: chemical reactions induced by γ-rays, β and α-particles (energy in the range 100eV–10MeV).
- Photochemistry can be considered the low-energy branch of Radiation Chemistry. In both fields the chemical (and biological effects) of radiation occur through two mechanisms:
  - direct interaction of the radiation (**primary reactions**)
  - indirect action from radiolytic products, especially free radicals (**secondary reactions**)
- Radiation Chemistry should be distinguished from radiation damage, which refers to structural transformation (particularly in the solid state) induced by radiation.

Basics: Radiation Induced Reactions

- **Primary reactions** set off by ionizing radiation (M=atom or molecule, "$\rightarrow$ "= radiation induced reaction):
  - Ionization processes: $M \rightarrow M^{+} + e^{-}$
  - Production of excited states: $M \rightarrow M^{*}$
- The minimum energy needed for both processes is of the order of several eV.
- The ions M^{+} and excited atoms or molecules M^{*} give rise to (further) **secondary reactions**:
  - $M^{+} \rightarrow R^{+} + R\bullet$ (dissociation, production of a **free radical**)
  - $M^{+} + e^{-} \rightarrow M^{*}$ (recombination)
  - $M^{+} + X \rightarrow Y^{+}$ (chemical reaction)
  - $M^{+} + X \rightarrow M + X^{+}$ (charge transfer)
  - $M^{+} \rightarrow M^{n+} + (n-1)e^{-}$ (emission of Auger electrons)
  - $M^{*} \rightarrow M + h\nu$ (fluorescence)
  - $M^{*} \rightarrow 2R\bullet$ (dissociation into **radicals**)
  - $M^{*} \rightarrow R^{+} + R^{-}$ (dissociation into ions)
  - $M^{*} + X \rightarrow Y$ (chemical reaction)
  - $M^{*} + X \rightarrow M + X^{*}$ (transfer of excitation energy)
- The concentration of the reaction products in the track is proportional to the LET.

Approximate Time Scale of Events in Radiation Chemistry

(Supplement!)

Stage	Process	Duration
Physical	Energy absorption, ionization.	10^{-18} to 10^{-12} s
Physico-Chemical	Interaction of ions with molecules, formation of free radicals.	10^{-14} to 10^{-6} s
Chemical	Interaction of free radicals with molecules, (cells and DNA).	10^{-12} to seconds
Biochemical / Biological	Cell death, change in genetic data in cell, mutations, cancer.	10^{-3} seconds to tens of years

Chemical Yield of a Reaction / First Examples

- Exploitation of the chemical effects of radiation on an industrial scale requires doses of ~1Gy to 1MGy.
- The G-value is used as a measure of the chemical yield. It is defined by the number of molecules formed or decomposed per 100eV energy absorbed in the system. E.g.: $G(H_2)=3$, $G(-H_2O)=11$.

Examples of radiation-induced reactions.

Reactions	*G*-values
Production of O_3 by irradiation of O_2	6–10
Production of NO_2 by irradiation of N_2/O_2 mixtures (by-products NO and N_2O)	1–7
Production of C_2H_5Br by irradiation of a mixture of C_2H_4 and HBr	$>10^5$
Production of C_2H_5Cl by irradiation of a mixture of C_2H_4 and HCl	$\approx 10^4$
Chlorination (similar to the photochemical reaction by UV), e.g. by irradiation of a mixture of C_6H_6 and Cl_2	10^4–10^5
Oxidation of carbohydrates; production of phenol by irradiation of a mixture of C_6H_6 and O_2	10^4–10^5
Sulfochlorination (similar to the photochemical reaction by UV), e.g. production of sulfonic acid chlorides by irradiation of mixtures of carbohydrates, SO_2 and Cl_2	$\approx 10^7$
Production of alkylsulfonic acids by irradiation of mixtures of carbohydrates, SO_2 and O_2	10^3–10^4

Applications of Radiation Chemistry

Advantages:

- Radiation induced reactions can be carried out at lower temperatures than those that depend on thermal initiation. This may eliminate unwanted side reactions.
- Many free-radical processes require the addition of a chemical initiator whose presence may lead to contamination of the end product. Thus purer products may be obtained by radiation induced reactions.
- While photoinduced reactions must be carried out in vessels transparent to the radiation (glass or silica), steel vessels can be used for γ-initiated reactions.

Disadvantages:

- The capital cost of building suitable protective housing is relatively high. Especially stringent safety precautions need to be taken.
- The yield of primary products in most irradiated systems is low: G = 1 to 10.

Chain Reactions / Examples of Radiation-Induced Reactions (1)

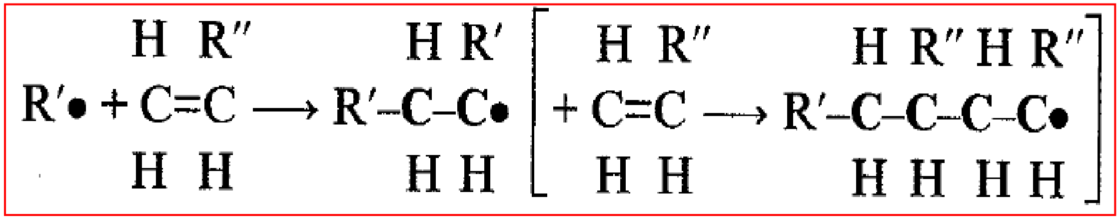
- **In chain reactions a sequence of radical-substrate reactions is repeated a number of times before the radical center is lost** (M=substrate/monomer, P=product/polymer)**:**
  - Initiation: M → R•
  - Propagation: R• + M → R• + P
  - Termination: 2R• → unreactive products
- **Production of ethyl bromide:**
  - Ethylene reacts with hydrogen bromide when irradiated with γ-rays at room temperature: $C_2H_4 + HBr \rightarrow C_2H_5Br$
  - The reaction is a chain process with G (C_2H_5Br) ~ 10^5.
  - A very pure product (> 99.5% purity) is obtained.
- **Production of gammexane:**
  - The reaction of chlorine with benzene to give gammexane (γ-benzene hexachloride) is a chain reaction that can be conveniently initiated by radiation: $C_6H_6 + 3Cl_2 \rightarrow C_6H_6Cl_6$
  - The G-value for this process is about 10^5.
  - Gammexane is widely used as an insecticide.

Examples of Radiation-Induced Reactions (2)

- **Production of detergents:**
  - The sulphoxation of alkanes (an alkane is a long chain of carbon linked together by single bonds, general formula C_nH_{2n+2}; examples: methane CH_4, ethane C_2H_6) can be initiated in high yields by ionizing radiation: $RH + SO_2 + \frac{1}{2}O_2 \rightarrow RSO_3H$
  - The reaction product is used for detergents.
  - In comparison to non-radiation processes the radiation process products are more easily biodegradable.
- **Nitrogen fixation:**
  - It has been shown that nitrogen and oxygen can be made to combine directly by irradiation to form nitrous oxide (laughing gas):
    - $N_2 + O_2 \rightarrow 2NO\bullet$
    - $2N_2 + O_2 \rightarrow 2N_2O$
  - As a by product also nitrogen dioxide NO_2 can be made.
  - The G-value for this process is low (about 4).

Radiation-Induced Polymerization (1): Basics

- **In polymers chain reactions can be triggered by the radicals created by irradiation:**
  - Initiation: Abstraction of atoms leading to a new free radical R'• on a polymer chain:
    R• + R'H ⟶ RH + R'•
  - Propagation: Addition to double bonds in olefin (olefin or alkene is an unsaturated chemical compound containing at least one carbon-to-carbon double bond; e.g., the simplest alkenes with only one double bond are the series of hydrocarbons with the general formula C_nH_{2n}.) to form another radical:



  - Termination: Radicals combine to an unreactive product: $2R\bullet \rightarrow R_2$
- **Although high-energy radiation can be used to initiate polymerisation, it is not used on an industrial scale for the production of bulk polymers as it cannot compete economically with catalytic methods.**
- **Radiation induced polymerisation has found specialist applications in the curing of surface coatings and the printing industry.**
- **Radiation effects on polymers are complex and may lead either to the enhancement of the polymer structure (cross-linking) or to its degradation. Although both processes occur simultaneously, polymers may be classified according to which one dominates.**

Radiation-Induced Polymerization (2): Cross-Linking/Degradation

Radiation interlacing or cross-linking:

- leads to an increase in hardness, while elasticity and solubility decrease
- active sites along the polymer chain are formed by the ionizing radiation
- chemical bonds are established between adjacent polymer chains leading to complex networks
- cross-linking dominates for polyethylene, polystyrene, PVC (polyvinyl chloride), and in particular heat shrink plastics
- heat shrink plastics are industrially produced by radiation cross-linking

Radiation induced degradation:

- caused by bond scission of the carbon chain
- scission polymers are Teflon, cellulose and polypropylene
- only limited industrial applications like the degradation of Teflon:
  - with doses of ~1MGy Teflon can be sufficiently degraded to allow grinding into small particles
  - thus scrap Teflon can be recycled into powders fine enough (~3μm) for use in printing ink and as coating additives

Radiation-Induced Reactions in Chemical Dosimetry

- Dosimetry systems have different properties and hence different areas of applications.
- Chemical dosimetry systems are important, because they are widely used as transfer dosimeters in industry, e.g., **Fricke dosimeter**: absorbed dose calculated from readily measured concentration of ferric ions.

Dosimetry systems (after McLaughlin, 1989).

Dosimeter	Classification	Range*	Comments
Calorimetry	Primary dosimeters and reference dosimeters	0.2 to 2 Gy/s**	Radiation energy absorbed as heat in a thermally isolated mass and measured
Ionisation chambers	Reference and field dosimeters	300 to 20,000 Gy/s	Radiation induces secondary elecrons in the chamber walls and ionisation in the gas
Standard ferrous sulphate (Fricke dosimeter)	Chemical dosimeters, reference dosimeters	2 to 200 Gy	The radiation induced yield of ferric species from acidified ferrous sulphate is measured
Cerous ceric dosimeter	Chemical dosimeters, transfer dosimeter	10^3–5×10^4 Gy (up to 10^6 Gy possible)	The formation of ceric ions induced by the irradiation of acidified cerous or cerous/ceric solutions
Alanine dosimeter	Transfer dosimeter	1 to 10^5 Gy	Alanine mixed with paraffin, cellulose, polystyrene or other binders produces free radicals on irradiation which are measured by (ESR)
Plastic dosimeters	Routine dosimeters	Varies, but generally in the range 10^3 to 10^6 Gy	Changes in the optical absorption of a range of plastics or dyed polymers (e.g. red, amber or clear perspex). Colour changes in radiation sensitive films
Thermoluminescent (TLD) dosimeters	Routine dosimeters	refer comments	Usable dose range depends on the material: $Li_2B_4O_7$: 10^{-2} to 10^5 Gy; CaF_2;Mn: 10^{-6} to 10^3 Gy; $CaSO_4$;Dy: 10^{-1} to 5×10^6 Gy

* Calorimetry and ionisation chambers expressed in terms of the absorbed dose rate (Gy/s); chemical dosimeters expressed as total dose absorbed (Gy)
** A water/polystyrene calorimeter (US National Institute of Science and Technology)

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.

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 **^{60}Co, also ^{137}Cs**.
- 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

Dose Requirements of Various Applications of Food Irradiation

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!)

D_{10} 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 (D_{10} 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^{-}_{aq} (reducing agent), •H
  - stable end-products: H_2, H_2O_2
- 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 $•RO_2$ (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).

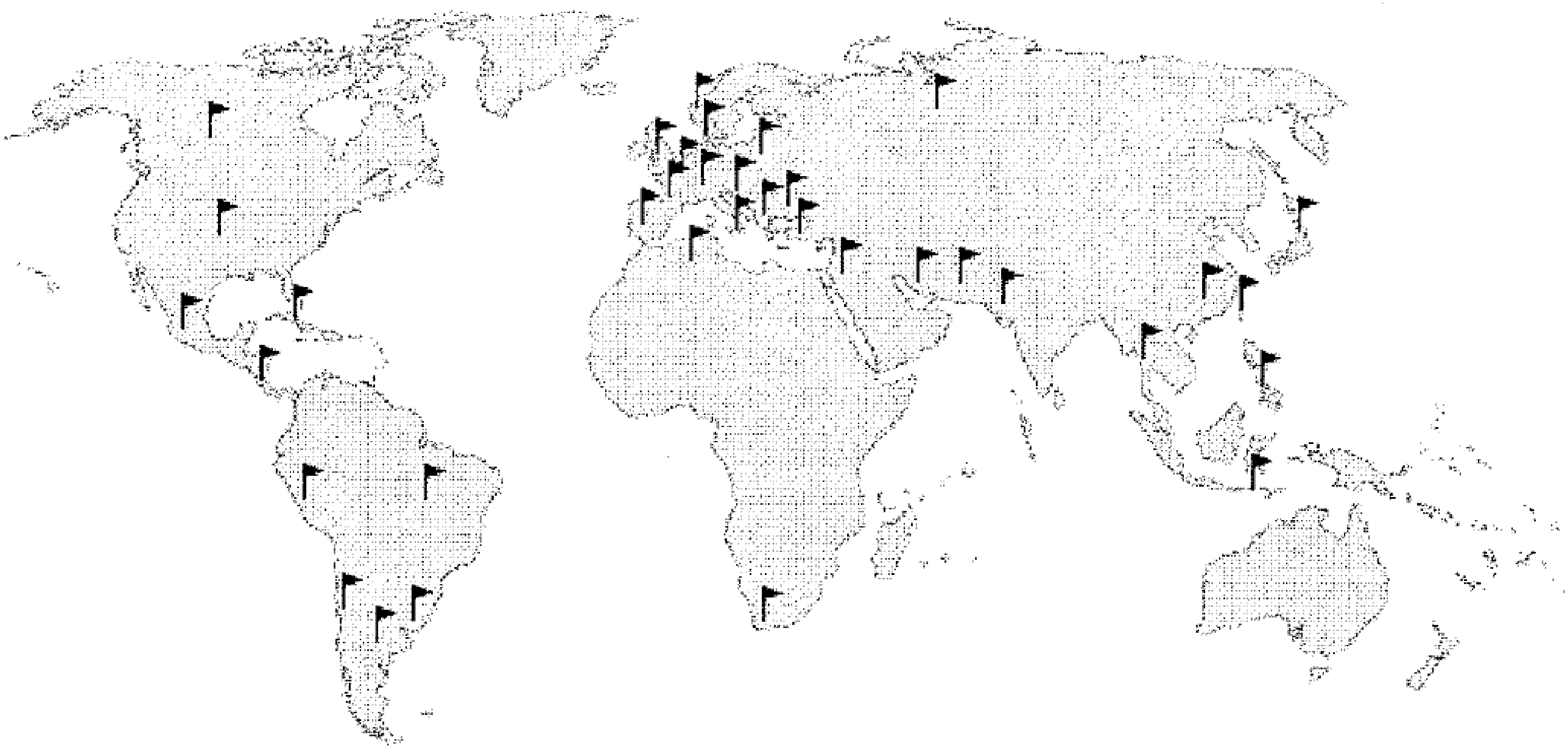
Combination Processes in Food Irradiation

- 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: ^{60}Co 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).

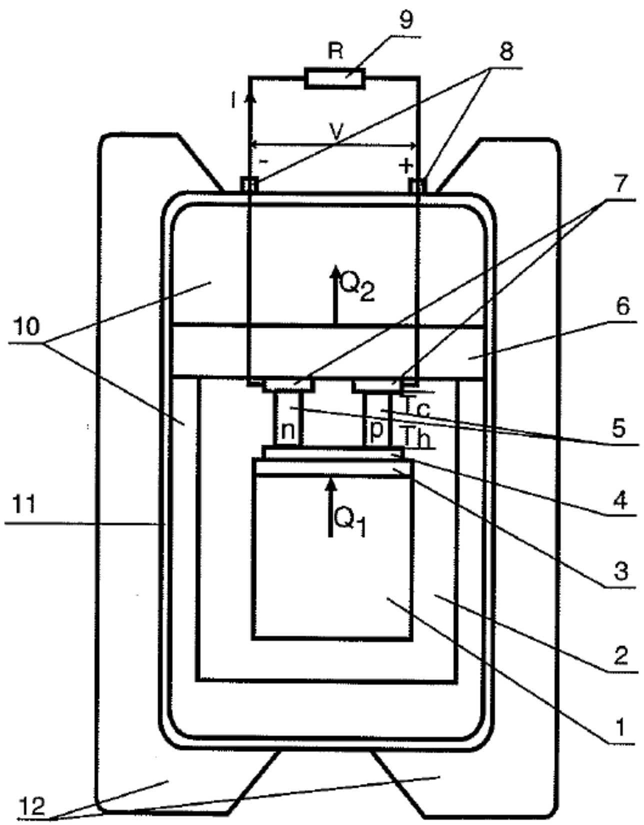
Radionuclide Batteries: Sources

Radionuclides for application in radionuclide batteries.

Radionuclide	Half-life [y]	Radiation	Production
^{3}H	12.323	β^-	^{6}Li$(n,\alpha)^{3}$H
^{14}C	5730	β^-	^{14}N$(n,p)^{14}$C
^{60}Co	5.272	β^-, γ	^{59}Co$(n,\gamma)^{60}$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	$\beta^-, (\varepsilon), \gamma, e^-$	^{169}Tm$(n,\gamma)^{170}$Tm
^{171}Tm	1.92	β^-, γ	^{170}Er$(n,\gamma)^{171}$Er $\xrightarrow{\beta^-}$ ^{171}Tm
^{204}Tl	3.78	$\beta^-, (\varepsilon)$	^{203}Tl$(n,\gamma)^{204}$Tl
^{210}Po	0.38	α, γ	Decay product of ^{238}U
^{228}Th	1.913	α, γ	Decay product of ^{232}Th
^{232}U	68.9	$\alpha, (sf), \gamma$	^{230}Th$(n,\gamma)^{231}$Th $\xrightarrow{\beta^-}$ ^{231}Pa; ^{231}Pa$(n,\gamma)^{232}$Pa $\xrightarrow{\beta^-}$ ^{232}U
^{238}Pu	87.74	$\alpha, (sf), \gamma$	^{237}Np$(n,\gamma)^{238}$Np $\xrightarrow{\beta^-}$ ^{238}Pu Decay product of ^{242}Cm
^{241}Am	432.2	$\alpha, (sf), \gamma$	^{240}Pu$(n,\gamma)^{241}$Pu $\xrightarrow{\beta^-}$ ^{241}Am
^{242}Cm	0.45	$\alpha, (sf), \gamma$	^{241}Am$(n,\gamma)^{242}$Am $\xrightarrow{\beta^-}$ ^{242}Cm
^{244}Cm	18.10	$\alpha, (sf), \gamma$	^{243}Am$(n,\gamma)^{244}$Am $\xrightarrow{\beta^-}$ ^{244}Cm

- 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
- Alpha emitters have the advantage that the decay-energy is relatively high and that α-particles are effectively absorbed.
- Radionuclides decaying by subsequent emission of several α particles, such as ^{238}Pu and ^{232}U, are most favourable:
  - **^{238}Pu**: $T_{1/2}$=87.74a, E_α=5.50, 5.46, ... MeV, specific power = 0.56W_{therm}/g
  - **^{232}U**: $T_{1/2}$=68.9a, E_α=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.

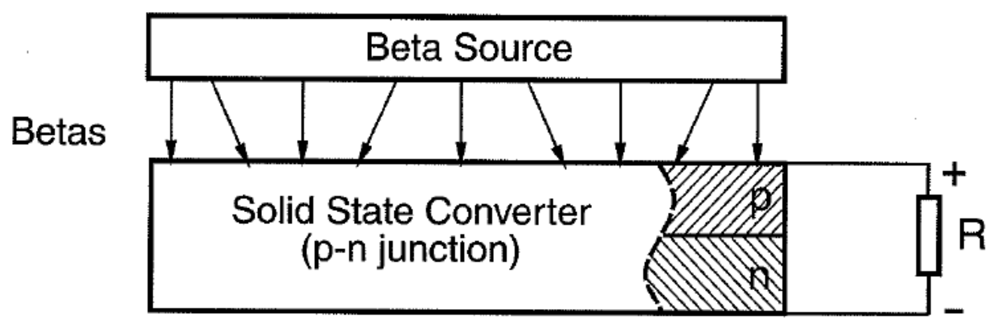
- The radioisotope fuel is held in hermetically sealed container or ampoule called radionuclide heat source (RHS).
- The conversion of heat to electricity is based on the **SEEBECK-effect**:
  - a temperature gradient between two branches of an electric circuit composed of different conductors (or semiconductors) will lead to a thermoelectric force, i.e., an electric current will flow in the loop.
- As a source typically the ceramic form of $^{238}PuO_2$ 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 ^{238}Pu, ^{232}U, ^{227}Ac, ^{242}Cm.
  - 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 ^{147}Pm.
  - 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 cm^2 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: ^{147}Pm, ^{14}C, ^{63}Ni, T.
  - Efficiencies of about 4% are obtained.



Scheme of direct-conversion betavoltaic.

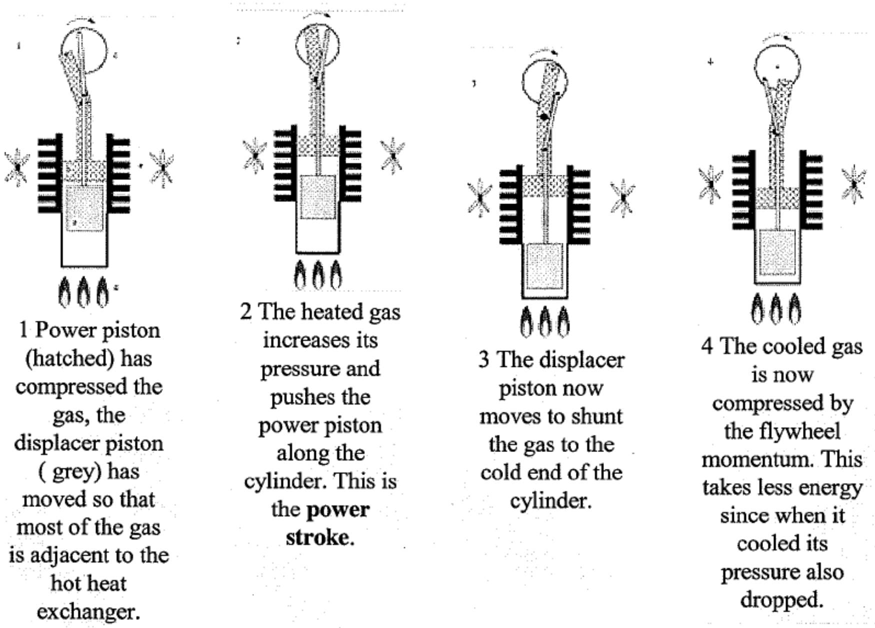
Application Examples (1): Characteristics of RITEGs

- 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

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



- 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 $^{238}PuO_2$ 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

- Ionizing radiation is successfully applied to induce chemical reactions:
  - Especially polymerisation can be induced by γ-radiation and leads to cross-linking and degradation.
  - Teflon can be recycled.
- ^{60}Co γ-radiation, electron beams and bremsstrahlung are used to sterilise 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 pathogenetic 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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