RADIATION PROCESSING

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Summary

Irradiation of a substrate with ionizing radiation produces free radicals through excitation and ionization processes. Subsequently, these free radicals will induce chemical or biological changes in the substrate. These events can be controlled, and are used by industry as a substitute for conventional processing techniques based on heating or addition of chemicals: collectively, these techniques are known as **radiation processing**. Radiation processing is used widely for cross-linking various polymer-based products such as wire and cable, tube, pipe, tires, packaging film, and molded products. It is also used in purification or treatment of flue gases and industrial wastewater, and as an alternative method for sterilizing medical devices. The advantages of radiation processing include the creation of novel products with desirable material properties, the absence of any chemical residues (since no chemical additives are required to initiate the reaction), and environmental friendliness. In most cases radiation-induced reactions are independent of temperature, allow high throughput processing, and are homogenous processes that are easy to control.

1. Radiation Processing

Radiation takes the form of either electromagnetic waves or particles, that travel at or near the speed of light respectively. Radiation processing utilizes radiation energy to treat a material that can be in the form of gas, liquid or solid. Radiation treatment is a means of initiating chemical and biological changes in a material. The radiation must have sufficient energy, and this type of radiation is known as **ionizing radiation**.

Ionizing radiation is defined as radiation that has sufficient energy to dislodge electrons from atoms and molecules and convert them to electrically-charged particles called **ions**. Further reactions of ions and electrons lead to the formation of **free radicals**. These are usually highly reactive, and eventually lead to chemical reactions. The studies of the chemical and biological changes in the system produced by absorbing ionizing radiation are known as **radiation chemistry** and **radiation biology**, respectively. These phenomena are all exploited during radiation processing.

1.1. Types of Ionizing Radiation

1.1.1. Electromagnetic Radiation

There are two types of ionizing radiation: **electromagnetic radiation** and **high-energy charged particles**. Figure 1 shows an electromagnetic radiation series that comprises radio waves, microwaves, visible light, ultraviolet light, X-rays, and gamma rays. However, only X-rays and gamma rays, having short wavelengths and energy \( E = \frac{hc}{\lambda} \) higher than 50 eV, are capable of ionizing atoms and molecules. Therefore, X-rays and gamma rays have the same properties and similar effects on materials, yet their origins are different. Industrial X-rays are generated by machines, while gamma rays are emitted by radioisotopes.
Electromagnetic radiation of longer wavelength (for example, ultraviolet radiation) may initiate chemical changes in the system not through direct ionization but through the process of photochemistry via the formation of excited species.

1.1.2. High-Energy Charged Particles

*High-energy charged particles* can be negatively or positively charged. Negatively-charged particles such as electrons can be generated by a machine such as an electron accelerator. On the other hand, positively-charged particles, such as H, He, Ar, C, and positrons, are generated by an ion beam accelerator. However, high-energy charged particles, such as beta particles (\(\beta\)—electron) and alpha particles (\(\alpha\)—helium), can also be obtained from radioisotopes. Unlike X-rays and gamma rays, charged particles have limited penetrating power, and can be stopped by a thin substrate such as paper. However, increasing the energy of the particles from a few keV to several MeV can increase their penetration power. Beam currents of charged particles can vary from a few \(\mu\)A to several mA. A high current will increase the number of charged particles, and hence dose rate. Since charged particles have sufficient energy to ionize atoms and molecules, their chemical effects are the same as those of X-rays or gamma rays.

1.2. Sources of Ionizing Radiation

Ionizing radiation can be obtained from two different sources: radioisotopes and electrical discharge machinery. The most common radioisotopes used commercially are cobalt-60 and cesium-137, both of them gamma (\(\gamma\)) emitters. Other sources of ionizing radiation are electron accelerators, X-ray machines, and positively-charged particle or ion-beam accelerators.

1.2.1. Radioisotopes

*Radioisotopes*, also known as radioactive isotopes or radionuclides, occur naturally, and can also be produced artificially in a nuclear reactor. They are unstable elements with an excess of neutrons or protons in their nuclei, and emit alpha, beta, and gamma (\(\alpha, \beta, \gamma\)) radiation as they spontaneously disintegrate (or decay) to a stable state. The time taken by radioisotopes to decay to the level of radioactivity originally present is known as their *half-life*, and these values are specific for each radioisotope of a particular element. The decay scheme of the radioisotope cobalt-60 is shown in Figure
2.

Figure 2. Decay scheme of the radioisotope cobalt-60

<table>
<thead>
<tr>
<th>Source</th>
<th>Half life (Year)</th>
<th>Type of radiation</th>
<th>Energy of radiation (MeV)</th>
</tr>
</thead>
<tbody>
<tr>
<td>60Co</td>
<td>5.27</td>
<td>β</td>
<td></td>
</tr>
<tr>
<td></td>
<td></td>
<td>γ</td>
<td>0.314</td>
</tr>
<tr>
<td></td>
<td></td>
<td>γ</td>
<td>1.332</td>
</tr>
<tr>
<td>137Cs</td>
<td>30</td>
<td>β</td>
<td>0.520</td>
</tr>
<tr>
<td></td>
<td></td>
<td>γ</td>
<td>0.662</td>
</tr>
</tbody>
</table>

Table 1. Radioisotopes: radiation sources

Cobalt-60 and cesium-137 are the industrial sources of gamma radiation: they have different half-lives, as shown in Table 1. They are generally housed in a lead cell known as a gamma cell, or in a concrete cell with walls a few meters thick. Cobalt-60 is the most popular industrial source of gamma radiation, because its radiation energy is higher than that of cesium-137. It is used for:

- sterilizing medical products, herbs, cosmetic items, and pharmaceutical raw materials;
- food preservation;
- decontamination of sewage sludge and wastewater;
- vulcanization of rubber latex; and
- other applications where the greater penetration of gamma radiation offers an advantage.

1.2.2. Electron Accelerator

An electron accelerator is generally the preferred radiation source for curing or cross-linking polymer-based materials such as plastic films, heat-shrink film, heat-shrink tubing, hot water pipes, wire, cable, and for many other applications requiring high-speed processing of materials that are not too thick.
Electron accelerators are manufactured in various types, and can be designed to produce electron beams with energies ranging from 80 keV to 10 MeV for commercial applications. Types with beam powers ranging from a few kW to 300 kW are also currently available. A more powerful accelerator with beam power of up to 500 kW is anticipated for future production.

Electrons have lower penetrating power than gamma radiation. The energy of the electron can be controlled using the electron accelerator to offer a penetration power appropriate to the application in question. For example, electron energy ranges of 80–350 keV are used to cure printing inks, coatings, and adhesives on various types of substrate, while an energy range of 500 keV–5.0 MeV is used to cross-link polyolefins such as the insulations on wires, tubes, and plastic films. Medical products are sterilized using electron energy ranges of 5.0–10 MeV.

1.3. Radiation Units

Radioactivity is the property of a material to emit radiation. Its value also indicates the strength of the radiation source. The Standard International unit of radioactivity is the Becquerel (Bq), although the old Curie (Ci) is still in use.

The energy of ionizing radiation is as follows:

The energy unit of the electron is measured in volts (eV, keV, MeV) as

\[ 1 \text{ eV} = 1.6021 \times 10^{-19} \text{ Joule} \quad \text{or} \quad 1 \text{ J} = 6.2418 \times 10^{18} \text{ eV} \quad (1) \]

Gamma radiation from a cobalt-60 source has two energies (1.17 MeV and 1.33 MeV), whereas energies of X-rays and electron beams may vary from a few keV to 10 MeV.

Radiation dose is defined as follows:

The absorbed dose (Grays/Gy) is the amount of energy absorbed per unit mass of the irradiated product. The Gray, an SI unit, has replaced an older unit, the rad.

\[ 1.0 \text{ Gy} = 1.0 \text{ J/kg} (=100\text{rad}) \]

\[ = 6.2418 \times 10^{18} \text{ eV/kg} \quad (2) \]

\[ = 6.2418 \times 10^{18} \text{ eV/dm}^3 \text{ for water (dm}^3\text{.liter)} \]

The absorbed dose rate is the absorbed dose per unit time (for example, Gy sec\(^{-1}\), kGy min\(^{-1}\), or kGy hr\(^{-1}\)).

The relationship of radiation doses to calories is as follows:

\[ 1.0 \text{ cal} = 4.185 \text{ Joule} \quad 1 \text{ J} = 0.2389 \text{ cal} \]
1.0 kGy = 1 x 10^3 J/kg = 238.9 cal kg⁻¹ (about 0.24 °C increase) 

G is the number of molecules/radicals produced per 100 eV absorbed energy.

Radiation from a Co-60 source measuring 1 Ci means that 37 thousand million (3.7 x 10^{10}) of its radioactive atoms are disintegrated in one second. The newer unit, the Becquerel, measures this with reference to disintegrations per second (1.0 Bq = one disintegration per second).

2. Interaction of Radiation with Matter

2.1. Electromagnetic Radiation (Gamma Rays)

Appreciating the interaction between radiation and matter is essential to any understanding of radiation-induced chemical changes arising directly from the absorption of radiation energy by matter. When electromagnetic radiation such as gamma rays passes through matter, the absorption of gamma rays by matter obeys the fundamental Lambert-Beer law 

\[ I = I_0 e^{-\mu x} \]

where \( I \) and \( I_0 \) are the intensities of the transmitted and incident radiation respectively, \( x \) is the thickness of the absorber, and \( \mu \) the linear absorption coefficient. If the thickness of the absorber is expressed in centimeters, \( \mu \) is measured in cm⁻¹.

The linear absorption coefficient depends on the density of the absorber and is defined as the mass absorption coefficient by 

\[ \mu_{\text{mass}} = \frac{\mu_{\text{linear}}}{\rho} \]

where \( \rho \) is the density of the absorber.

The total absorption coefficient is the sum of three separate coefficients representing the three main processes of energy absorption by gamma rays: the photoelectric effect, the Compton effect, and pair production.

2.1.1. Photoelectric Effect

In this process, the photon or electromagnetic radiation (gamma rays) is completely absorbed by an atom. The photon energy is transferred to the inner orbit of an electron (usually from the K-shell), which is then ejected from the atom. An electron from the outer atomic orbit subsequently fills the inner orbital space, with consequent liberation of energy. This energy may manifest as X-rays. The ejected electron will travel with energy equivalent to the energy of the photon less that binding energy of the electron to the atom, and will undergo further interactions with other atoms. The photoelectric effect is greatest in the case of low photon energies of <0.1 MeV, or in matters with high atomic numbers.

2.1.2. Compton Effect (Compton Scattering)

Interaction between a high-energy photon or electromagnetic radiation and a free or loosely-bound electron will lead to diversion of the photon, and a consequent loss of energy. The lost energy is transferred to the electron which will travel with an energy
equivalent to the energy loss by the photon. The scattered photon may then undergo subsequent absorption by either the photoelectric effect or the Compton effect. Compton absorption is most important in the cases of photons with higher energies of 0.1–10 MeV. The Compton effect is the dominant process in the interaction of Co-60 gamma rays with water.

2.1.3. Pair Production

Pair production is the simultaneous formation of a positive electron (positron) and an electron as a result of interaction between electromagnetic radiation with sufficient energy (≥1.02 MeV) and the field of an atomic nucleus of the atom. After being slowed down the electron and positron recombine, resulting in the production of two 0.51 MeV gamma rays (annihilation radiation).

Whatever the mechanism by which the gamma radiation loses energy, secondary electrons with considerable kinetic energy are produced. Subsequent energy absorption processes, accounting for most of the energy of the incident radiation, will be those characteristic of electrons. These electrons are similar to those generated by an electron accelerator.

2.2. Electrons

Electrons interact with matter in several ways, principally:
- emission of electromagnetic radiation
- inelastic collision
- elastic collision.

2.2.1. Emission of Electromagnetic Radiation

Electrons passing close to the nucleus of an atom are decelerated, with subsequent emission of energy in the form of X-rays. This radiation is known as Bremsstrahlung radiation. Bremsstrahlung radiation can be absorbed by matter in a similar manner to electromagnetic radiation, and this may lead to chemical changes. It is the predominantly mode of energy loss for electron energies in the range 10–100 MeV, but it is negligible below 0.1 MeV.

2.2.2. Inelastic Collisions

At lower energy levels electrons lose their energies as a result of inelastic collisions with electrons of the target material(s), resulting in ionization and excitation. The rate of energy loss with distance is known as the linear energy transfer (LET). The LET of all particles increases as their energy decreases: in other words, as the electrons are slowed down in the medium (Table 2).

<table>
<thead>
<tr>
<th>Energy (MeV)</th>
<th>Range in air (cm, 15 °C, 760mmHg)</th>
<th>Range in aluminum (mm)</th>
<th>Range in water (mm)</th>
<th>Average LET in water (keV μ m⁻¹)</th>
</tr>
</thead>
</table>

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Table 2. Ranges and LET values for electrons

<p>| | | | | |</p>
<table>
<thead>
<tr>
<th></th>
<th></th>
<th></th>
<th></th>
<th></th>
</tr>
</thead>
<tbody>
<tr>
<td>1</td>
<td>405</td>
<td>1.5</td>
<td>4.1</td>
<td>0.24</td>
</tr>
<tr>
<td>3</td>
<td>1400</td>
<td>5.5</td>
<td>15</td>
<td>0.20</td>
</tr>
<tr>
<td>10</td>
<td>4200</td>
<td>19.5</td>
<td>52</td>
<td>0.19</td>
</tr>
</tbody>
</table>

2.2.3. Elastic Collisions

The coulomb field of a nucleus readily deflects electrons because of their small mass. There is no energy loss in elastic collision, but these collisions will result in nonlinear passage of electrons through the medium. Elastic collisions are important in the case of low-energy electrons in materials of high atomic number.

Bibliography


Applications.


Biographical Sketch

Dr. Khairul Zaman Haji Mohd Dahlan is currently a Director of the Radiation Processing Technology Division of the Malaysian Institute for Nuclear Technology Research (MINT). MINT is the research institute operated by the Malaysian government under the Ministry of Science, Technology,
and the Environment. Dr. Khairul Zaman obtained his M.Sc. in Nuclear and Radiation Chemistry, and subsequently his Ph.D. in Radiation Chemistry, from the University of Salford, UK in 1978 and 1981 respectively. He has been involved in radiation chemistry and radiation-processing research in the field of radiation curing and radiation cross-linking of polymer blends, and in recent years has worked on radiation cross-linking of agro-fiber plastic composites.

He has been instrumental in establishing radiation processing technology facilities and capabilities in Malaysia, and in particular the establishment of electron beam technology at MINT. He is the National Coordinator in Radiation Technology, and represents Malaysia in many regional programs and international meetings organized by the International Atomic Energy Agency (IAEA). He has served as an IAEA expert under the technical assistant project mission to several countries in the Asia/Pacific region.

He co-supervised seven Ph.D. and seven M.Sc. students attached to MINT during 1993–2001 in various field of research related to radiation processing. He was also elected an external examiner for an M.Sc. and a Ph.D. student during this period. He is currently a visiting scientist at the Institute of Advanced Technology (ITMA), University Putra Malaysia (UPM).