

RADIATION CHEMISTRY

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Summary

Radiation chemistry implies the chemical effects of ionizing radiations whose energies are several orders of magnitude higher than the energies of the chemical bonds that are typical of the matter they interact with. The ionizing radiation penetrating the matter causes excitation and ionization of the molecules in the medium. The primary energy deposition is relatively little dependent on the properties and composition of the medium. On the contrary the reactions of the reactive intermediates formed (excited molecules, excess electrons, ions, radicals, etc.), strongly depend on the properties and composition of the medium. The ejected electron in low-permittivity condensed phases (e.g. in hydrocarbons) after losing energy in collisions with the surrounding molecules returns to the positive ion and in this geminate recombination excited molecule forms. (Nevertheless, some of the ejected electrons are present as excess electrons in a localized or quasi-free state.) In high-permittivity liquids, like water and lower alcohols, the electron stabilizes in the form of solvated (in water hydrated, e_{aq}^-) electron. Radiation chemistry of water in dilute solutions reduces essentially to the chemistry of three intermediates e_{aq}^- , $\cdot\text{OH}$ and $\cdot\text{H}$. The yield of radiolytic decomposition of organic molecules is largely determined by the relation between the first excitation energy and the energy of chemical bonds: when the excitation energy is much higher than the bond energy, decomposition is highly probable. In alkanes, all ionized and excited molecules undergo chemical decomposition. In aromatic molecules, however, where the relationship between excitation energy and bond energy is just the opposite, the decomposition yield is very low. In this chapter after describing the characteristics of ionizing energy absorption, radiation sources and techniques applied in radiation chemistry research the radiolysis of different classes of inorganic and organic substances is discussed in concise form. The chapter introduces also some of the established radiation technologies.

1. Introduction, Short History

Radiation chemistry implies the chemical effects of interactions of ionizing radiation with materials. The term ionizing radiation in a wider sense is also applied to photons or particles having sufficient energy to ionize the molecules of the medium: it involves photons with energies ranging from the first ionization energy of the medium (~ 10 eV) up to several million eV (MeV), as well as energetic charged particles, electrons, positrons, accelerated heavy ions, etc. In a narrower sense, only those radiations are considered in radiation chemistry, whose energies are several orders of magnitude higher than the energies of the chemical bonds. The result of the energy absorption is breaking or rearrangement of chemical bonds; i.e. decomposition of some of the initial molecules and formation of new ones.

High-energy photons, charged species with sufficiently high energy and neutrons might be absorbed by the nuclei and cause nuclear reactions. This is the field of nuclear physics and nuclear chemistry or radiochemistry and these effects are not considered in radiation chemistry.

Radiation chemistry traces its origin to the discovery of X-rays (Roentgen rays) by Wilhelm C. Röntgen in 1895 and of radioactivity by Antoine Henri Becquerel in 1896. X-rays and uranium salts were both shown to fog packed photographic films and they were shown to produce electrically charged ions in air and other gases. It was also shown that by decomposition of water a mixture of hydrogen and oxygen was produced. These results clearly demonstrated that the radiation exerts physical and chemical effects on the material irradiated. The biological effects were also discovered very shortly after discovery of ionizing radiation.

Most early studies on the chemical effects of ionizing radiation were carried out with the weak radiation sources available in early 20th century. Even then, using long exposure, it was possible to convert a significant proportion of the starting material to products for chemical analysis. The development of powerful X-ray machines for industrial and medical purposes in the 1930s, the gradual development of accelerator technology, the industrialized production of radionuclides in nuclear reactors in the 1950s provided new high-intensity radiation sources that were used not only in scientific research, but also in industrial research laboratories for radiation technological purposes.

The chemical effects are due to the absorption of radiation energy in the material. Therefore, for practical applications the amount of energy absorbed should be precisely known. For this purpose a number of physical and chemical dosimeter systems were developed. The most well known chemical dosimeter is the Fricke dosimeter (discovered in 1936), which is based on the oxidation of Fe^{2+} ions to Fe^{3+} ions in 0.4 mol dm^{-3} aqueous H_2SO_4 solution.

In the early period, chemical changes in gases were mostly studied. In gaseous systems, using electric measurements, it was relatively easy to measure the number of ion-pairs produced. For that reason the radiation chemical yield (the so-called ion-pair yield) was originally defined as the number of molecules produced (or decomposed) divided by the number ion pairs created by the radiation. Since in liquids and solids it is difficult to measure the number of ion pairs, later a new quantity was introduced that refers to the amount of energy absorbed rather than to the number of ion pairs created. This quantity was the *G*-value, defined as the number of product molecules formed (or initial molecules changed) for every 100 eV of energy absorbed. Using SI units, while retaining the old symbol, the *G*-value is given in units of mol J^{-1} or $\mu\text{mol J}^{-1}$. The conversion between the “molecular” *G*-values and their “molar” SI equivalents is given as follows: $(1 \text{ molecule})/(100 \text{ eV}) \Leftrightarrow (0.1036 \mu\text{mol}/\text{J})$.

2. Absorption of Radiation Energy

2.1. Energy Absorption

In the radiation-chemical practice, accelerated electron irradiation and γ -ray irradiation are mostly applied. Therefore a brief summary will be given here on the energy absorption of these particles in matter. When they penetrate matter, the high-energy photons or electrons interact with the molecules of the medium.

High-energy electrons interact either with the Coulomb field of the nucleus of the atom or with the electron shell. When passing through the electric field of an atomic nucleus, the electron can be decelerated leading to continuous bremsstrahlung. In interactions with the electron shell characteristic X-rays may form. The absorption of the energy of X-rays and high-energy bremsstrahlung is similar to the absorption of γ -radiation energy. In collisions with the electrons of the medium the high-energy electron loses energy and slows down, and ionization or excitation of the absorber material results.

High-energy photons interact with the medium by the photoelectric effect, Compton scattering, and by electron-positron pair production. The threshold energy of the pair production is twice the rest-mass energy of electron (2×0.51 MeV). This interaction type has importance at energies considerably higher than the threshold energy and especially in materials containing heavy elements. The photoelectric effect is predominant at low energies. In the latter interaction, an electron is ejected from the atom or molecule with a kinetic energy equal to the energy of photon minus the binding energy of the electron. In the Compton effect only a part of the incident photon energy is used to eject an electron from the atom or molecule. The lower-energy photon thus produced may take part in further Compton or photoelectric interactions. As a consequence of the energy degradation process of a high-energy photon, a large number of energetic electrons are produced. Activated (electron-excited or ionized) molecules that may undergo chemical decomposition mostly form in the interactions of these fast electrons with the atoms or molecules in the medium.

In the interactions of energetic electrons (or other charged particles), a large amount of energy is transferred to the electron of the interacting molecule (*hard collisions*). In this case, the classical model, which treats the problem in terms of a fast-moving charged particle passing along a free electron initially at rest, gives a good description. As a result of the Coulomb interaction, the free electron is set in motion at the expense of the kinetic energy of the fast-moving charged particle.

When the energy of the bombarding electron is comparable to the binding energy of the target electron, the picture of a collision with a free electron at rest is not correct any more. For such *soft collisions* a model is applied which involves resonance-like absorption of energy by the molecules. According to Platzman's optical approximation theory the fast electron passing by the molecule induces a strongly time-dependent electric polarization in it. This time-dependent polarization is experienced by the molecule as interaction with electromagnetic waves with a range of frequencies. Some of the electrons may start oscillating and a resonant excitation or ionization may occur.

The total energy lost per unit path length by the primary charged particle is obtained by summing up all losses occurring in hard and soft collisions. The total energy lost per unit path length is called the linear energy transfer (LET). In water the mean LET for energetic γ -rays or fast electrons is ~ 0.2 eV nm⁻¹. When 5.3 MeV α -particles are applied for irradiation, the LET-value is much higher: 43 eV nm⁻¹. LET is proportional to the square of the charge of the particle, and inversely proportional to the square of the particle speed.

In radiation chemistry, the intermediates form with a strongly non-homogeneous distribution in the matrix. At the beginning of an electron track, when the energy is still high, the intermediates are sparsely populated. When the electron slows down and the energy is low, the local intermediate density is high. The energetic electrons along the track also produce secondary electrons (δ -electrons) of lower energy that also cause ionization and excitation in the medium. However, due to the lower energy, their tracks are shorter. When the energy of the electron is ~ 100 eV, its range in condensed phase is very short and there is a high probability that it transfers excitation or ionization energy to a molecule that is close to the original site of ionization. In this way a small cluster or *spur* of ionized or excited molecules forms. In the spur the intermediates mutually influence each other's reaction possibilities. According to a more strict definition even a single ionization producing a geminate ion pair creates a spur: a spur is a grouping of reactive intermediates in which there is a significant probability that some of the intermediates will react with each other during the course of their diffusion into the bulk of the medium.

In water vapor, 44% of the spurs contain one ion pair, 22% of them two and 13% three ion pairs. In condensed phase there are 2-4 activated molecules in one spur corresponding to ~ 100 eV of energy loss. The δ -electrons in the 100-500 eV range produce larger spurs that may be called blobs. The δ -electrons in the 500 eV-5 keV range can generate short tracks, or above 5 keV branching tracks.

If a mixture of compounds is irradiated, then the fraction of the total absorbed energy transferred to each component of the mixture is determined by the mole fraction (x) of the component and by its energy absorption characteristics. For instance, to a first approximation, the yield of a product P , $G(P)$, formed in a binary mixture of A and B is given by the equations:

$$G(P) = G(P)_A \varepsilon_A + G(P)_B \varepsilon_B, \quad (1)$$

$$\varepsilon_A = \frac{(1-x) \cdot n_A}{(1-x) \cdot n_A + x \cdot n_B}, \quad (2)$$

where $G(P)_A$ and $G(P)_B$ are the yields of product P from A and B , and ε_A and ε_B are the electron fractions of A and B , respectively. The electron fraction in a binary mixture is defined by Eq. (2), where n_A and n_B are total numbers of electrons in molecules A and B , respectively, and x is the mole fraction of B .

2.2. Formation and Decay of Ion Pairs

The energy of high-energy particles absorbed in the medium mostly causes ionization of molecules. In gas phase the ejected electron can easily escape the Coulomb field of the positive ion. The charges become homogeneously distributed in the medium: charge recombination occurs randomly (homogeneous recombination). In contrast to the gas phase, in condensed phases the fate of the ion pairs and the nature of the recombination are strongly dependent on the properties of the medium. In the low-permittivity hydrocarbons (low dielectric constant) most of the ejected electrons lose the kinetic

energy in interactions with the surrounding molecules and return to the geminate ion. Such recombination is called geminate recombination.

Molecule	Ionization potential, eV	W-value, eV
<i>Inorganic molecules</i>		
H ₂	15.4	36.5
O ₂	12.1	30.8
CO ₂	13.8	33.0
N ₂ O	12.7	32.6
H ₂ O	12.6	29.6
NH ₃	10.2	26.6
<i>Hydrocarbons</i>		
Methane	12.7	27.3
Ethane	11.7	25.0
<i>n</i> -Hexane	10.17	22.61
3-Methylpentane	10.05	22.67
Cyclohexane	9.88	25.05
Ethylene	10.52	24.6
Benzene	9.25	20.9
<i>Other organic molecules</i>		
Methanol	10.08	
Ethanol	10.5	24.8
Acetone	9.7	

Table 1: Ionization potentials and the energy values of one ion pair production (W-value) in the gas phase.

According to Onsager's theory the probability, $P(r)$, that an electron escapes the Coulomb field of the positive ion depends on the permittivity of the solution:

$$P(r) = \exp\left(-\frac{e^2}{4\pi\epsilon_0\epsilon_r r kT}\right), \quad (3)$$

where e is the elementary charge, ϵ_r is the relative permittivity, ϵ_0 is the vacuum permittivity and r is the distance between the ion and the thermalized electron. The critical distance $r_c = e^2 / (4\pi\epsilon_0\epsilon_r kT)$ – where the energy of Coulomb interaction between electron and positive ion is equal to the thermal energy kT – is 25-32 nm in hydrocarbons. When $r > r_c$, the fate of the electron mainly depends on the thermal diffusion. When $r < r_c$, the Coulomb interactions are predominant. In alkanes 2-30% of the ejected electrons escape the positive ion and undergo homogeneous recombination. The free ion yields strongly depend on the molecular structure; in linear alkanes the yields are low, the yields increase with the increasing branching.

In liquid water at room temperature the critical distance is only $r_c = 0.7$ nm. Most of the ejected electrons (dry electrons) have sufficient kinetic energy to get away from the positive ions. After thermalization the electron orients the surrounding water molecules and forms the so-called hydrated electron.

In Table 1 the ionization potentials and the mean energies needed for ionization in the gas phase (W -values) are collected for a few molecules. The W -values are between 21 and 36 eV, which are much higher than the ionization potential values. The high difference is partly due to the fact that a considerable part of energy absorbed is used for excitation and also to the fact that ionization does not occur solely at the lowest-energy electron.

3. Radiation Sources, Dosimeters

3.1. Radiation Sources

The sources of ionizing radiation used for research or industrial irradiation purposes can be divided into two groups: sources applying radioactive nuclides, such as ^{60}Co , ^{137}Cs or the $^{90}\text{Sr}/^{90}\text{Y}$ pair, and machine sources of radiation, such as X-ray equipment or electron accelerators. Without going into details, a few characteristics of some radioactive sources and accelerators will be mentioned, which are most relevant to using them for irradiation.

^{60}Co is the most widely used γ -radiation source for experimental and industrial purposes. It has a half-life of 5.27 years and emits two high-energy photons, 1.17 MeV and 1.33 MeV per disintegration. The isotope is produced by neutron irradiation in $^{59}\text{Co}(n,\gamma)^{60}\text{Co}$ reaction in nuclear reactors. ^{59}Co is usually irradiated in the form of pellets and then the pellets are encapsulated in stainless-steel containers. For research purposes, sources of 10^3 – 10^4 Ci (3.7×10^{13} – 3.7×10^{14} Bq) activity are generally used. The commercially available irradiation facilities are often self-shielded and portable although they may weigh several tons. In the industrial irradiation facilities the activity is generally in the range 5×10^4 – 10^6 Ci (1.8×10^{15} – 3.7×10^{16} Bq). These facilities have irradiation chambers of the size of a room, 100–500 m³. Shielding is provided by massive concrete walls, and the operators enter the irradiation chamber through a labyrinth. In the industrial irradiation facilities many tons of goods, e.g. medical products, agricultural products etc, are treated every day. There are about 200 industrial γ -facilities worldwide (2005). The irradiation facilities used in cancer therapy usually have activities around 10^4 Ci (3.7×10^{14} Bq).

Less frequently, the fission product ^{137}Cs is used for γ -irradiation. (Gamma energy: 0.66 MeV, half-life: 30 years.) ^{137}Cs is separated from spent fuel by reprocessing. It is usually applied in the form of cesium chloride sealed in stainless steel capsules.

From the β^- -ray emitters ^{90}Sr should be mentioned. This radionuclide is obtained from spent fuel by reprocessing. It emits β^- -rays with 0.545 MeV maximum (end-point) energy and a half-life of 28 years. ^{90}Sr disintegrates to its daughter ^{90}Y , which is also β^- -emitter with a half-life of 64 h, maximum energy 2.25 MeV.

In the pioneering period of radiation chemical research the radiation emitted by X-ray equipment was often utilized for irradiation purposes. Nowadays particle accelerators, and first of all electron accelerators are the main *machine sources* of radiation.

During the last 100 years many different types of particle accelerators were developed. These accelerators may differ in

- the principle of acceleration (acceleration by static electric field, high-frequency microwave, etc.),
- the type of the accelerated particle (electron, proton, heavy ions),
- the shape of orbit used for acceleration (linear, cyclic, elliptical),
- the energy of accelerated particle (from a few 100 keV to a hundred MeV or so) and
- the power (from a few 100 W to 100 kW).

Most of the classical type electron accelerators work on the same principle. A hot filament is used to produce electrons. These are then accelerated through a vacuum towards a target. Usually 20–50 μm thick aluminum or titanium foil windows are applied to take the particle beam out of the accelerator for external irradiation.

Electron accelerators are often used to generate high-energy X-rays. The electrons are simply allowed to fall on a target of high atomic number. This method actually produces bremsstrahlung (German for “slowing-down radiation”), possessing all energies up to that of the bombarding electrons. Bremsstrahlung is often used when physicists need high-energy photon irradiation and also in radiation technological applications.

In the *Van de Graaff accelerators* a high voltage electric field (up to several million Volts) accelerates the electrons emitted by the cathode. The maximum energy of electrons is limited by the high voltage insulation: most accelerators work between 1 and 5 MeV electron energy. These machines normally produce a continuous beam, but some of the accelerators were modified to deliver pulses in the subnanosecond - microsecond ($1 \text{ ns} = 10^{-9} \text{ s}$, $1 \mu\text{s} = 10^{-6} \text{ s}$) range. Van de Graaff type accelerators are also used to accelerate particles other than electrons, e.g. protons, He^{2+} or C^{6+} ions: these accelerators are the principal radiation sources in heavy ion radiolysis studies. In the ion gun these heavy positive ions are produced in an electrical discharge.

In the *linear accelerators* (LINAC) a microwave produced by a magnetron or klystron power generator is used to accelerate the electrons entering the accelerator tube after a 50-100 keV preacceleration by static electric field. The accelerator tube is a few-meter long segmented wave-guide. The electrons injected into the wave-guide, after a bunching process, ride on the crest of waves traveling along the guide and are accelerated to high energies up to tens of MeV. Two microwave frequencies, $\sim 1400 \text{ MHz}$ (L-band) and $\sim 2800 \text{ MHz}$ (S-band) are most commonly used. The accelerated electrons are delivered in pulses of ns or μs duration with a repetition rate of 50-500 Hz. For scientific purpose usually low-power accelerators are applied, for industrial irradiation high-power machines, 10-100 kW, are also available. In the high-

power accelerators the beam can be magnetically deflected to sweep a larger irradiation area. There were about 1000 electron beam accelerators in industrial use in 1998.

Laser *plasma linac accelerators* are recently under development (Figure 1). In these systems a very intense (terawatt, TW) femtosecond laser pulse hits the gas flow in a supersonic gas jet. In the intense laser beam, beyond the critical value, ionization of the gas takes place. The wave energy is transferred to longitudinal electron momentum. These electrons are accelerated and bunched: they have broad energy distribution up to 30 MeV.

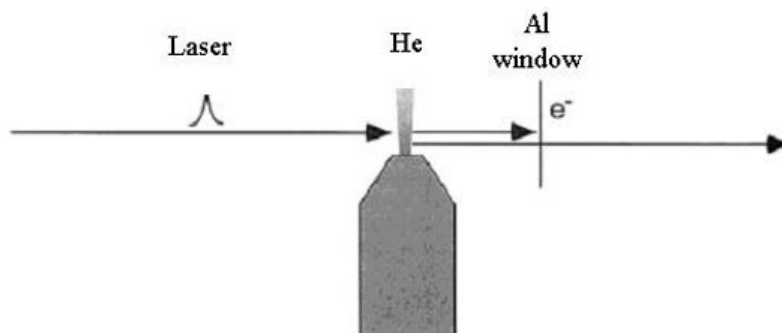


Figure 1: Scheme of a laser plasma linac accelerator.

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Biographical Sketch

László Wojnárovits was born in 1944 in Szeged (Hungary). He obtained his university degrees from the Technical University of Veszprém (chemical engineer 1967) and from the Technical University of Budapest (engineer specialized in radiochemistry 1970). In 1973 he obtained the title Candidate of Chemical Sciences from the Hungarian Academy of Sciences (HAS) for his dissertation entitled "Influence of Cyclic Structure on the Radiolysis of Hydrocarbons", in 1986 earned the title Doctor of Chemical Sciences from HAS for his thesis "Excited States in the Radiolysis and Photolysis of Liquid Alkanes". Not much later he was given a Honorary Professor title from University of Veszprém and

habilitation from Technical University of Budapest. At present he is the head of Department of Radiation Chemistry and director of Institute of Isotopes of HAS (Budapest).

His field of research is radiation chemistry. With the International Atomic Energy's scholarship in 1975-76 he visited the Tokyo Institute of Technology in Japan studying the oxidation of alkenes and the 7.6 eV photon induced processes in alkane systems. Coming back to Budapest he continued this work with aim of getting a deeper insight into energy transfer and final product forming reactions in photolysis and radiolysis. In 1982 for two months in the Laboratory of Photochemistry and Radiation Chemistry CNR (Bologna, Italy) he studied the fluorescence lifetime of alkanes. He carried out similar measurements in 1982-83 in cooperation with the Radiation Chemistry Group in Leipzig (Germany) and also in 1984 and 1985 in the Interuniversitair Reactor Institut in Delft (the Netherlands). Between 1993 and 1995 he spent 18 months in USA in the Radiation Laboratory (headed by Prof. Schuler) at University of Notre Dame. Together with Dr. LaVerne they studied the heavy ion radiolysis of alkanes. With Prof. Schuler they introduced gel permeation HPLC for the determination of alkyl iodides formed in iodine radical scavenging in alkanes. During the more than thirty years in science he has written about 250 scientific papers and given more than 100 lectures at conferences. He is the author of four books and two book chapters.