

## RADIOCHEMISTRY AND NUCLEAR CHEMISTRY

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### Summary

This chapter outlines the field of radiochemistry and nuclear chemistry (RC&NC). It gives a short summary of the early history of RC&NC as well as a detailed timeline of the development of subatomic concepts, nuclear science and technology. The basic nuclear concepts, units and relationships are also introduced here. The chapter explains in some detail radioactive decay, nuclear reactions, and radiation–matter interactions to serve with necessary background to related chapters on a selection of the applications of RC&NC written by specialists of their fields.

### 1. Introduction

The “official” definitions of *radiochemistry (RC)* and *nuclear chemistry (NC)* in the “IUPAC Gold Book” (<http://goldbook.iupac.org/index-alpha.html>) read as follows:

- Radiochemistry: “That part of chemistry which deals with radioactive materials. It includes the production of radionuclides and their compounds by processing irradiated materials or naturally occurring radioactive materials, the application of chemical techniques to nuclear studies, and the application of radioactivity to the investigation of chemical, biochemical or biomedical problems.”

- Nuclear chemistry: “The part of chemistry which deals with the study of nuclei and nuclear reactions using chemical methods.”

However there is a wide interdisciplinary area between chemistry and nuclear science/technology which is not always easy to unambiguously divide into the above categories. Keeping this in mind, we prefer to consider *radiochemistry and nuclear chemistry* as a single technical term (*RC&NC*) covering the whole area that can be characterized by either of the following criteria:

- nuclear science with relevance to chemistry
- chemistry with relevance to nuclear science.

There are workers in the field of what we call RC&NC who define themselves either as *radiochemists* or as *nuclear chemists*. However, a considerable part of the core of knowledge of the two areas is common, and they are interlaced to a degree where it is practically not worth the trouble trying to disentangle them here. The term *radionuclear chemistry* also occurs in the literature, which speaks for itself.

Historically, in the early years of RC&NC, the term RC was used for the whole field. Later on, in the 1930s, the term NC was introduced for the chemical aspects of the study, production, properties and reactions of atomic nuclei. Several decades later, either of these terms was also applied to all kinds of nuclear instrumental methods used as tools in chemical research, such as Mössbauer spectroscopy. By now a kind of “fusion” has taken place between the once distinct meanings and for many people active in these fields RC and NC have become practically synonyms.

Note that some of the nuclear methods of chemical structure investigation, e.g. *NMR*, are often not considered as being part of either RC or NC. With NMR this is partly so because the resonance energies involved are negligible, and partly because it has become a widely used independent method in its own right whose nuclear origins have been “forgotten” for various reasons. (It is probably no accident, e.g., that the adjective “nuclear” has been dropped out from the acronym of one of the best diagnostic methods, MRI.) On the other hand, some of the methods connected with X-rays, e.g. *XRFS*, are often included. So when outlining RC&NC, high-energy radiations (nuclear or other) can be useful practical markers. Accordingly, books on RC&NC always contain chapters on non-radioactive radiation sources such as *synchrotrons*. *Isotopic chemistry* (radioactive or not) is usually considered as part of the topics included. *Radiation chemistry* may or may not be included (but we give it a whole chapter, partly because it is not discussed elsewhere in the EOLSS material).

The ambiguity concerning the borderline between RC and NC can also be observed when browsing through books titled “Radiochemistry”, “Nuclear Chemistry”, “Nuclear and Radiochemistry” or the like. The contents are very similar or practically the same. As a matter of fact, the introductory chapters (dealing with subatomic particles, properties of nuclei, stability/instability, decay modes, interactions of radiations with matter, detection of radiations etc.) are in common with most fields of nuclear science although the depth of treatment may be different.

The selection of Topics elaborated by specialists as separate chapters may seem rather arbitrary to those familiar with the contents of books on RC&NC. Contrary to all appearances, a lot of time and thought has been devoted to find a sensible compromise between what is customary, what is very important and what is covered in the rest of the huge EOLSS material. The result of these considerations is reflected by the scope of the Topics selected.

Last but not least, it has also been considered that RC&NC, an interdisciplinary field in itself, has many “inter-interdisciplinary” applications in earth sciences, biology, life sciences etc. Some of the following Chapters are written on such multidisciplinary applications.

## 2. The Beginnings of RC&NC and the Timeline of Nuclear Science

The history of RC&NC cannot be separated from that of the rest of nuclear science (and related fields). This is so, because

- chemistry is much older than nuclear science (chemists held their first international meeting in Karlsruhe as early as 1860 on fundamental issues like the definition of atoms and molecules);
- chemists got involved in the research of radioactivity as soon as it was discovered in 1896.

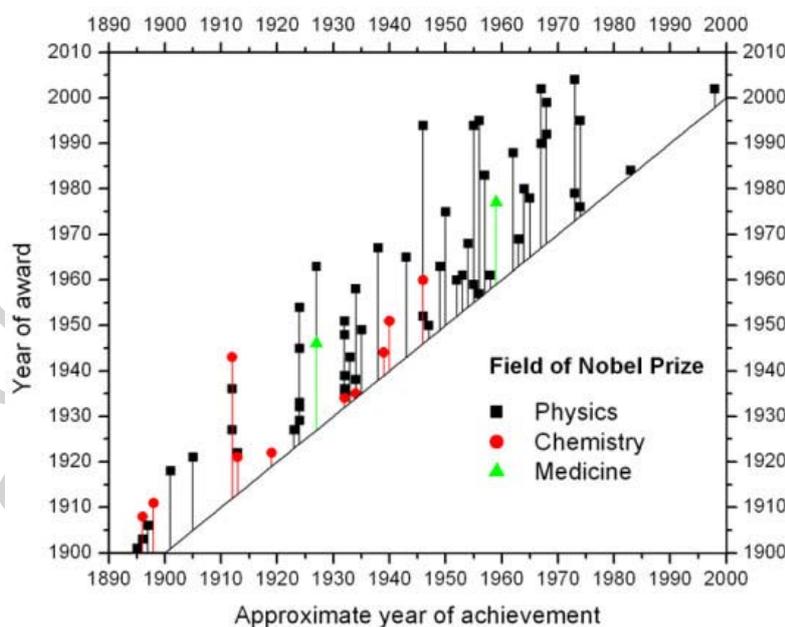


Figure 1: Nobel Prizes related to the development of nuclear science and subatomic concepts. The data are compiled from Appendix 1 giving a detailed chronology of key events. The length of each drop-line shows the delay between award and achievement. Note that some achievements had been awarded almost promptly, whereas the longest delay was almost half a century. (The drop-lines of the first few points are meaningless, as the Nobel Prize was first given in 1901.)

The role of chemistry in nuclear science (and related fields) can be seen in Figure 1 showing that there were 10 such years in the 105-year history of Nobel Prizes when the chemistry award was associated with this field. The achievements awarded cover the period from the very beginnings till about 1946. The last Nobel Prize in chemistry was awarded in this field in 1960.

The beginning of the nuclear epoch of humankind is marked by the 1896 discovery of the radioactivity of uranium by Antoine Henri Becquerel (1852-1908), representing the third generation of a family of French physicists, who won a 1903 shared Nobel Prize in physics “for the discovery of spontaneous radioactivity”.

Some put the year of birth of nuclear science one year earlier, to 1895, when the German physicist Wilhelm Conrad Röntgen (1845-1923) discovered his famous *X-rays*, *X-Strahlen* in German, a technical term coined by him. The expression X-ray, with X to remind of the traditional symbol *x* for unknown algebraic quantities, is an appropriate name indeed for a “mysterious” radiation that penetrates materials impenetrable to ordinary light. It is worth mentioning, however, that in some languages this same high-energy electromagnetic radiation bears its discoverer’s name and is called the equivalent of roentgen-rays, Röntgen-rays or similar. Röntgen also developed X-ray photography, revolutionizing medical diagnosis. He won the 1901 Nobel Prize in physics (which was also the first such prize given to anybody) for his discovery. His work also gave inspiration to Becquerel and Rutherford for their experiments on radioactivity.

Whichever date we accept as official year of birth of nuclear science in general (or nuclear and particle physics in particular), the year of birth of RC (and so that of RC&NC) is certainly at the latest 1898 when Marie Curie, née Maria Skłodowska (1867-1934), a Polish-born French chemist, together with her husband Pierre Curie (1859-1906), a French physicist, discovered *polonium* (and somewhat later *radium*) using the first *radiochemical methods* ever. The Curies were also the ones who shared the 1903 Nobel Prize with Becquerel. (In 1911, Madame Curie also won a second Nobel Prize, this time in chemistry, for “her services to the advancement of chemistry by the discovery of the elements radium and polonium, by the isolation of radium and the study of the nature and compounds of this remarkable element”.)

Considering the timeline of discoveries detailed in Appendix 1, one can easily understand why the year of birth of RC has been emphasized above. Namely, it would be rather anachronistic to talk about NC before the birth of the concept of the *atomic nucleus* itself, which dates from more than a decade later, 1911, when the New Zealand-born British physicist Ernest Rutherford (1871-1937) concluded that the vast majority of the mass of an atom must be concentrated in a tiny part of its volume, and this tiny but massive volume, referred to as the atomic nucleus, must be positively charged. His nuclear atom model replaced the Kelvin-Thomson “plum pudding” model according to which the atom should be pictured as a positively charged spherical mass in which *Z* electrons are embedded like raisins in a cake. The short-lived Kelvin-Thomson model was postulated in 1903 by Lord Kelvin, also known Sir William Thomson, (1824-1907), a Scottish-Irish mathematical physicist and engineer, and further developed by Joseph John Thomson (1856-1940), an English physicist, who studied the conduction of electricity by gases, discovered the electron (the first subatomic as well as truly

elementary/fundamental particle) and determined its charge to mass ratio, an important achievement that brought him a Nobel Prize in physics in 1906.

Although Rutherford's nuclear atom model changed the picture in the mind of chemists about the atom profoundly, he won his 1908 Nobel Prize in chemistry for an even more bizarre idea that contradicted the most sacred dogma of the chemists of his time, namely, that the atoms of the chemical elements are ultimate and unchangeable parts of matter. Around 1902-1903, together with the British chemist Frederick Soddy (1877-1956), he came to the heretical conclusion that *thorium and uranium became different elements through radioactive decay*. Rutherford was well aware how daring this conclusion was. Against all expectations, however, chemists turned out to be open-minded enough to acknowledge him, a physicist, as one of their kin by giving him the chemistry Nobel Prize.

By way of compensation maybe, physicists repaid this debt by giving 1/4 of the 2002 Nobel Prize in physics to a chemist, Raymond Davis Jr. (1914-2006), the discoverer of the *solar neutrino problem*, “for pioneering contributions to astrophysics, in particular for the detection of cosmic neutrinos”.

As these examples show, the roles of chemists and physicists got rather mixed up in nuclear and particle science in full accordance with the interdisciplinarity of the fields and the complexity of the problems to be solved. We can also see that although the last nuclear-related Nobel Prize in chemistry was awarded almost half a century ago, the last Nobel Prize to a radionuclear chemist was given much later, in 2002, although for an achievement that dates back to 1967.

Besides Nobel Prizes, the appreciation for the achievements of the forerunners of nuclear science has been expressed in several other ways:

- Elements 96, 104 and 111 bear the names of Curie (*curium*, Cm), Rutherford (*rutherfordium*, Rf), and Röntgen (*roentgenium*, Rg), respectively.
- The pre-SI unit of *radiation exposure to photons* also bears Röntgen's name (*roentgen*, R).
- Both “successful” units of *activity (disintegration rate)* had been named after the pioneers of radioactivity. The first activity unit, the *curie* (Ci), was introduced in 1930. Although not an SI unit, it is still widely used especially in medicinal applications (see Table 1). Since 1975, the official SI unit of activity is the *becquerel* (Bq) getting increasingly accepted by nuclear scientists. (A third and short-lived activity unit – the *rutherford*, meaning  $10^6$  decays per second – was also suggested in the 1930s in Rutherford's honor, but it has never become widely accepted.)

For further information on the development of subatomic concepts etc. see the introductory part of the next section as well as the detailed timeline in Appendix 1 which is meant to be self-contained as much as possible.

### 3. Nuclides and Nuclei – Isotopes, Isobars, Isotones, and Isomers

While studying naturally occurring decay series (see Figure 16), radiochemists were baffled because some of the members of the series could not be separated by chemical methods, although their radioactive properties (decay mode, half-life) were markedly different.

By 1907 “the doctrine of the complete chemical non-separability” of these members (an expression borrowed from Soddy’s Nobel lecture given in 1921) had become an accepted fact among the workers in this field. [Let us note it in parentheses that the Hungarian chemist George ‘György’ Hevesy (1885-1966) made good use of this fact by inventing the radioactive tracer technique in the meantime, thus earning the 1943 chemistry Nobel Prize.] To give an explanation for “non-separability”, as early as 1913 (i.e. six years before Rutherford discovered the *proton* as the constituent of the nucleus, thus taking the first large step towards identifying the atomic number of elements used by chemists for “ages” with something fundamental), Soddy introduced the technical term *isotope* from the Greek words “isos” (equal) and “topos” (place). His idea was that in spite of internal differences, different atomic species may occupy the same place on the periodic table, thus representing the same element. Note that isotopes are not really *inseparable* after all as it turns out from the chapter on *Isotope Effects, Isotope Separation, and Isotope Fractionation*.

Proof for the existence of isotopes as atomic species with different atomic masses was given within a year by J.J. Thomson via deflecting neon ions in electromagnetic fields. It turned out that natural Ne atoms could be divided in two classes which we now refer to as Ne-20 and Ne-22. (The third stable isotope, Ne-21 – making up only 0.27% of neon – escaped detection then.) This also showed that “isotopy” is not specific to radioactivity.

The deeper physical interpretation of isotopes followed about two decades later, after the discovery of the *neutron* in 1932 by the British physicist James Chadwick (1891-1974) who won the 1935 physics Nobel Prize for this achievement. According to this interpretation, the different isotopes of an element represent different classes of atoms/nuclei of that element, each containing the same number of protons ( $Z$ ), but different number of neutrons ( $N$ ).

Over the next decade or so, however, the term “isotope” had become overused/misused to the point where laypersons started to understand it as “*a dangerous type of atom that is radioactive*”. To straighten out things, the American physicist Truman P. Kohman introduced the concept of *nuclide* in 1947. However, a fat half century was not quite enough for this useful term to replace the term isotope everywhere it ought to. Therefore we devote this section to clarifying the difference between these terms and to introducing some closely related concepts.

#### 3.1. The Building Blocks of Atoms and Nuclei

Figure 2 shows the artistic representation of the structure of an “ordinary” helium atom based on the Standard Model of particles and interactions summarized in a separate

chapter (see: *Subatomic Particles, Nuclear Structure and Stability*). The *nucleus* of this particular atomic species is distinguished from other nuclei that it contains two *protons* (p) and two *neutrons* (n).

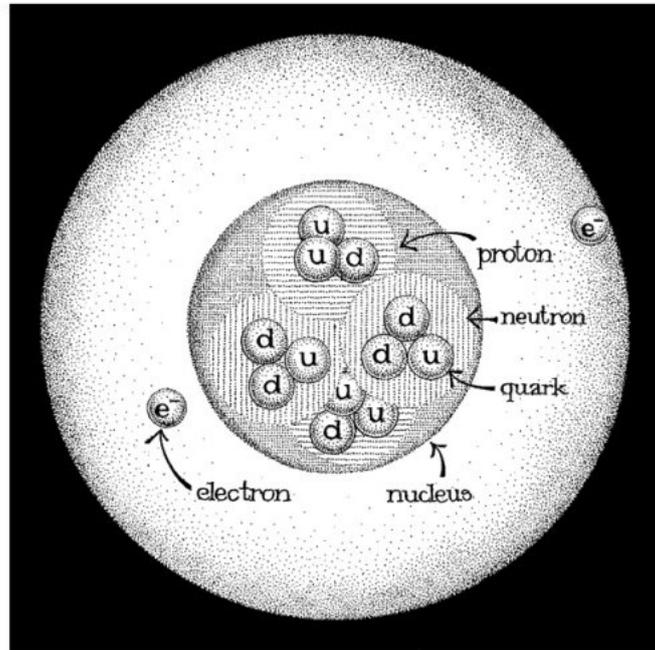


Figure 2: Naïve representation of the structure of a helium atom by “Iscsu” (pencil name of graphic artist István Molnár) based on the Standard Model (SM). Note that for better visibility, the individual parts of the atom are not all in proportion with each other. If a nucleon (neutron or proton) were 1 cm across, then the  ${}^4\text{He}$  nucleus would be 1.6 cm, but the atom itself would have a diameter of 1 km. It means that the atom is way undersized in the picture compared to its nucleus. The quarks and electrons, on the other hand, are fundamental particles according to the SM without any internal structure, i.e. point-like. Anyway, their size would be certainly not larger than 1–10  $\mu\text{m}$ , if the picture were proportional in every detail.

In general, a nucleus contains  $A$  nucleons ( $N$ : p and n together) out of which  $Z$  are protons and the rest are neutrons:

$$N = A - Z, \quad (1)$$

where the integer  $N$  is simply called the *neutron number* of the nucleus.

The integer  $A$  is often referred to as the *nucleon number* for obvious reasons, but it is most often called the *mass number*. The latter name is justified because protons and neutrons have very nearly the same mass (see Table 1) and therefore the nuclear mass  $m_N$  is approximately proportional to the number of nucleons:

$$m_N \propto A = Z + N. \quad (2)$$

Since electrons are much lighter than nucleons, the mass of the neutral atom “built” from the given nucleus and  $Z$  electrons will also be approximately proportional to  $A$ , and therefore the term mass number is also rightly applied to atoms.

The integer  $Z$  is known in chemistry as the *atomic number* (of the given element). It is also referred to as the *proton number* of the nucleus). Since the electric charge of the nucleus is  $eZ$ , where  $e$  is the charge of the proton called the *elementary charge* (Table 1),  $Z$  is also referred to as the *charge number* of the nucleus. As the electric charge of the electron ( $e$ ) is equal to  $-e$ , a neutral atom contains exactly  $Z$  extra-nuclear/orbital electrons. So  $Z$  also indicates the *electron number* of a neutral atom.

Protons and neutrons – and therefore the nucleus built from them – can exist in different energy states the lowest of which determines the *ground state* of the nucleus. *Excited-state* nuclei – same as excited atoms – seek higher stability and usually return to the ground state very quickly (say in  $10^{-7}$  s) by emitting electromagnetic radiation (called  $\gamma$ -rays in this case).

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### Biographical Sketch

**Sándor Nagy** was born in Budapest, Hungary, in 1949. He received his MSc in chemistry from Eötvös Loránd University (ELTE), Budapest, in 1972. He got his Dr. Univ. degree from ELTE in 1975, where he had also studied applied mathematics for five years. He received his CSc (PhD) in nuclear chemistry from the Hungarian Academy of Sciences in 1996.

He has been working for ELTE ever since graduating there. Presently he is Associate Professor in the Laboratory of Nuclear Chemistry, Institute of Chemistry, ELTE. In the meantime he was Visiting

Scientist at Lehigh University, Bethlehem, USA (1979-1980) and Postdoctoral Fellow/Adjunct Associate Professor at Drexel University, Philadelphia, USA (1987-1989). His research field has been chemical applications of Mössbauer spectroscopy. He teaches nuclear chemistry to chemistry majors. He co-edited with Prof. Vértes two books listed in the Bibliography. He also authored/co-authored some of the chapters of those books, and co-authored also the chapter on Mössbauer spectroscopy in Prof. Alfassi's book.

He is an IUPAC Fellow. He also used to work as National Representative for Hungary for IUPAC.

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