CHAPTER 1

Origin of Nuclear Science

Contents

1.1. Radioactive elements

In 1895 W. Roentgen discovered that when cathode rays (i.e. electrons) struck the wall of an evacuated glass tube, it caused the wall material to emit visible light (fluoresce), while at the same time a very penetrating radiation was produced. The name *X-ray* was given to this radiation. Learning about this, H. Becquerel, who had been interested in the fluorescent spectra of minerals, immediately decided to investigate the possibility that the fluorescence observed in some salts when exposed to sunlight also caused emission of X-rays. Crystals of potassium uranyl sulfate were placed on top of photographic plates, which had been wrapped in black paper, and the assembly was exposed to the sunlight. After development of some of the photographic plates, Becquerel concluded (erroneously) from the presence of black spots under the crystals that fluorescence in the crystals led to the emission of X-rays, which penetrated the wrapping paper. However, Becquerel soon found that the radiation causing the blackening was not "a transformation of solar energy" because it was found to occur even with assemblies that had not been exposed to light; the uranyl salt obviously produced radiation spontaneously. This radiation, which was first called uranium rays (or Becquerel rays) but later termed radioactive radiation (or simply *radioactivity*)[1], was similar to X-rays in that it ionized air, as observed through the discharge of electroscopes.

Marie Curie subsequently showed that all uranium and thorium compounds produced *ionizing radiation* independent of the chemical composition of the salts. This was convincing evidence that the radiation was a property of the element uranium or thorium. Moreover, she observed that some uranium minerals such as pitchblende produced more ionizing radiation than pure uranium compounds. She wrote: "this phenomenon leads to the assumption that these minerals contain elements which are more active than uranium". She and her husband, Pierre Curie, began a careful purification of pitchblende, measuring the amount of radiation in the solution

[1] The word radioactivity refers to the phenomenon *per se* as well as the intensity of the radiation observed.

and in the precipitate after each precipitation separation step. These first *radiochemical* investigations were highly successful: "while carrying out these operations, more active products are obtained. Finally, we obtained a substance whose activity was 400 times larger than that of uranium. We therefore believe that the substance that we have isolated from pitchblende is a hitherto unknown metal. If the existence of this metal can be affirmed, we suggest the name *polonium*." It was in the publication reporting the discovery of polonium in 1898 that the word radioactive was used for the first time. It may be noted that the same element was simultaneously and independently discovered by W. Marckwald who called it "radiotellurium".

In the same year the Curies, together with G. Bemont, isolated another radioactive substance for which they suggested the name *radium*. In order to prove that polonium and radium were in fact two new elements, large amounts of pitchblende were processed, and in 1902 M. Curie announced that she had been able to isolate about 0.1 g of pure radium chloride from more than one ton of pitchblende waste. The determination of the atomic weight of radium and the measurement of its emission spectrum provided the final proof that a new element had been isolated.

1.2. Radioactive decay

While investigating the radiochemical properties of uranium, W. Crookes and Becquerel made an important discovery. Precipitating a carbonate salt from a solution containing uranyl ions, they discovered that while the uranium remained in the supernatant liquid in the form of the soluble uranyl carbonate complex, the radioactivity originally associated with the uranium was

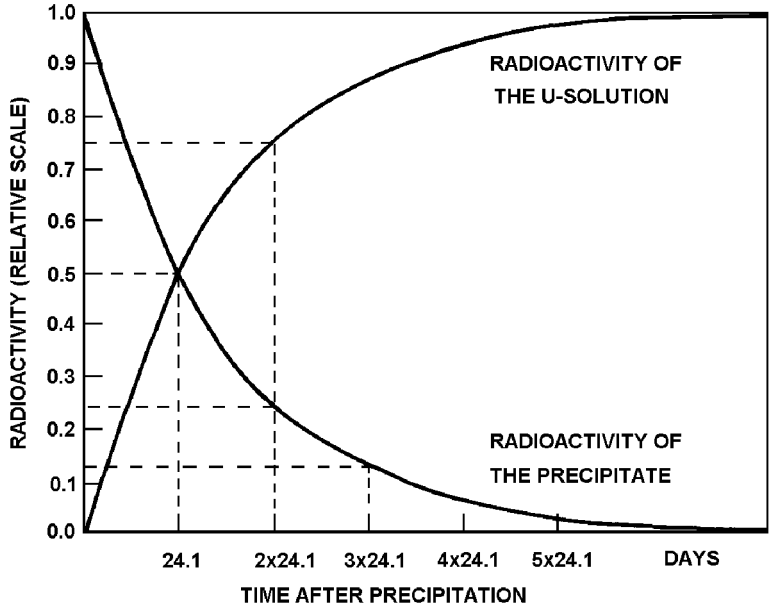

FIG. 1.1. Measured change in radioactivity from carbonate precipitate and supernatant uranium solution, i.e. the separation of daughter element UX (Th) from parent radioelement uranium.

now present in the precipitate, which contained no uranium. Moreover, the radioactivity of the precipitate slowly decreased with time, whereas the supernatant liquid showed a growth of radioactivity during the same period (Fig. 1.1). We know now that this measurement of radioactivity was concerned with only beta- and gamma-radiations, and not with the alpha-radiation which is emitted directly by uranium.

Similar results were obtained by E. Rutherford and F. Soddy when investigating the radioactivity of thorium. Later Rutherford and F. E. Dorn found that radioactive gases (*emanation*) could be separated from salts of uranium and thorium. After separation of the gas from the salt, the radioactivity of the gas decreased with time, while new radioactivity grew in the salt in a manner similar to that shown in Fig. 1.1. The rate of increase with time of the radioactivity in the salt was found to be completely independent of chemical processes, temperature, etc. Rutherford and Soddy concluded from these observations that radioactivity was due to changes within the atoms themselves. They proposed that, when radioactive decay occurred, the atoms of the original elements (e.g. of U or of Th) were transformed into atoms of new elements.

The radioactive elements were called *radioelements*. Lacking names for these radioelements, letters such as X, Y, Z, A, B, etc., were added to the symbol for the primary (i.e. parent) element. Thus, UX was produced from the radioactive decay of uranium, ThX from that of thorium, etc. These new radioelements (UX, ThX, etc.) had chemical properties that were different from the original elements, and could be separated from them through chemical processes such as precipitation, volatilization, electrolytic deposition, etc. The radioactive daughter elements decayed further to form still other elements, symbolized as UY, ThA, etc. A typical decay chain could be written: Ra → Rn → RaA → RaB → , etc.; Fig. 1.2.

A careful study of the radiation emitted from these radioactive elements demonstrated that it consisted of three components which were given the designation alpha (α), beta (β), and gamma(γ). *Alpha-radiation* was shown to be identical to helium ions, whereas *beta-radiation* was identical to electrons. *Gamma-radiation* had the same electromagnetic nature as X-rays but was of higher energy. The rate of radioactive decay per unit weight was found to be fixed for any specific radioelement, no matter what its chemical or physical state was, though this rate differed greatly for different radioelements. The decay rate could be expressed in terms of a *half-life*, which is the time it takes for the radioactivity of a radioelement to decay to one-half of its original value. Half-lives for the different radioelements were found to vary from fractions of a second to millions of years; e.g. that of ThA is 0.1 of a second, of UX it is 24.1 days (Fig. 1.1), and of uranium, millions of years.

1.3. Discovery of isotopes

By 1910 approximately 40 different chemical species had been identified through their chemical nature, the properties of their radiation, and their characteristic half-lives. The study of the generic relationships in the decay of the radioactive species showed that the radioelements could be divided into three distinct series. Two of these originated in uranium and the third in thorium. B. Boltwood found that all three of the series ended in the same

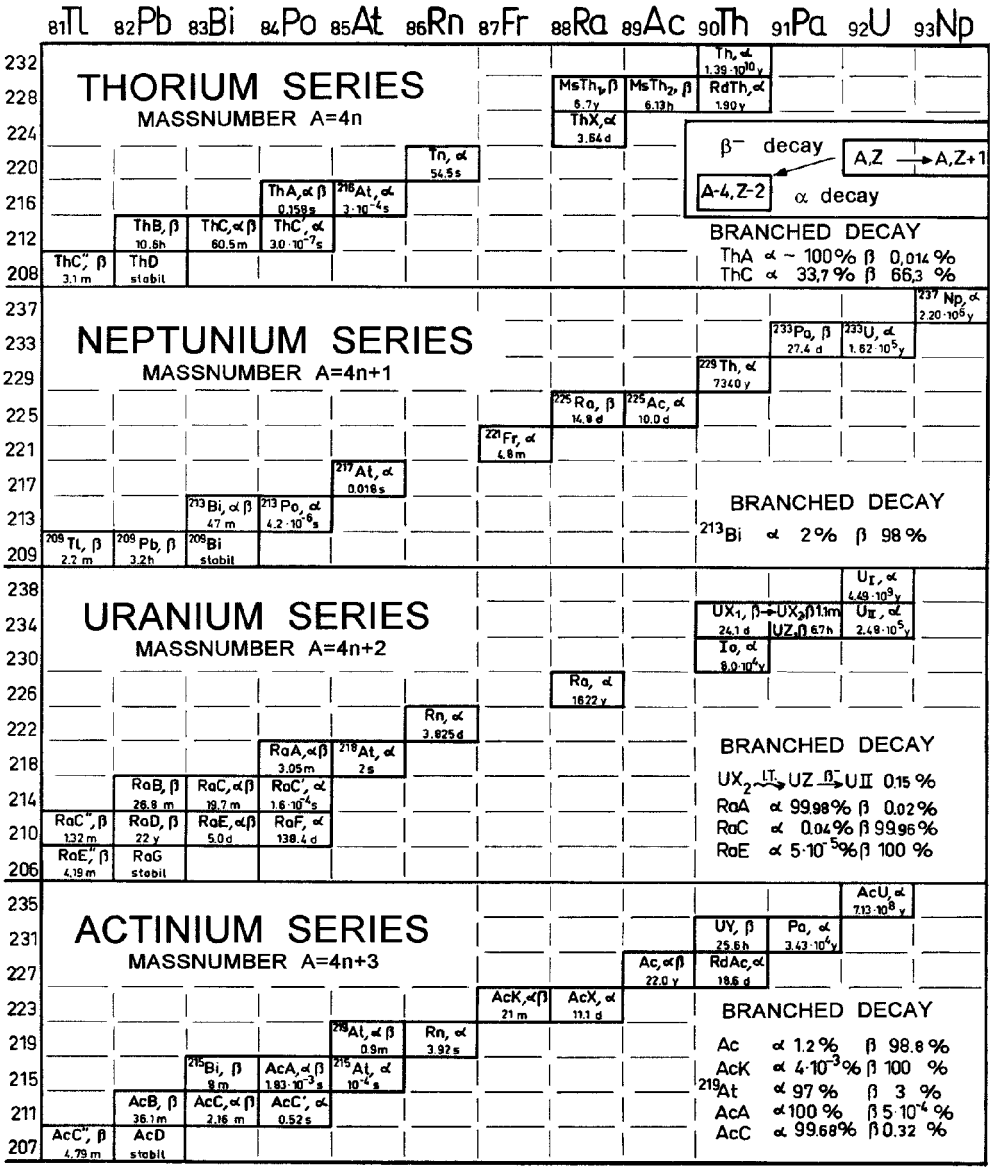

FIG. 1.2. The three naturally occurring radioactive decay series and the man-made neptunium series. Although ^{239}Pu (which is the parent to the actinium series) and ^{244}Pu (which is the parent to the thorium series) have been discovered in nature, the decay series shown here begin with the most abundant long-lived nuclides.

element – lead.

A major difficulty obvious to scientists at that time involved the fact that while it was known from the Periodic Table (Appendix I) that there was space for only 11 elements between lead and uranium, approximately 40 radioelements were known in the decay series from uranium to lead. To add to the confusion was the fact that it was found that in many cases it was not possible to separate some of the radioelements from each other by normal chemical means. For example, the radioelement RaD was found to be chemically identical to lead. In a similar manner, spectrographic investigations of the radioelement ionium showed exactly the same spectral lines that had been found previously to be due to the element thorium.

In 1913 K. Fajans and Soddy independently provided the explanation for these seemingly contradictory conditions. They stated that by the radioactive α-decay a new element is produced two places to the left of the mother element in the periodic system and in β-decay a new element is produced one place to the right of the mother element (Fig. 1.2). The radioelements that fall in the same place in the periodic system are chemically identical. Soddy proposed the name *isotopes* to account for different radioactive species which have the same chemical identity.

Research by J. J. Thomson soon provided conclusive support for the existence of isotopes. If a beam of positively charged gaseous ions is allowed to pass through electric or magnetic fields, the ions follow hyperbolic paths which are dependent on the masses and charges of the gaseous ions (see Fig. 2.1 and associated text). When these ion beams strike photographic plates, a darkening results which is proportional to the number of ions which hit the plate. By using this technique with neon gas, Thomson found that neon consists of two types of atoms with different atomic masses. The mass numbers for these two isotopes were 20 and 22. Moreover, from the degree of darkening of the photographic plate, Thomson calculated that neon consisted to about 90% of atoms with mass number 20, and 10% of atoms with mass number 22.

Thus a chemical element may consist of several kinds of atoms with different masses but with the same chemical properties. The 40 radioelements were, in truth, not 40 different elements but were isotopes of the 11 different chemical elements from lead to uranium.

To specify a particular isotope of an element, the *atomic number* (i.e. order, number, or place in the Periodic Table of elements) is written as a subscript to the left of the chemical symbol and the mass number (i.e. the integer value nearest to the mass of the neutral atom, measured in atomic weight units) as a superscript to the left. Thus the isotope of uranium with mass number 238 is written as $^{238}_{92}$U. Similarly, the isotope of protactinium with mass number 234 is designated $^{234}_{91}$Pa. For an alpha-particle we use either the Greek letter α or $^{4}_{2}$He. Similarly, the beta-particle is designated either by the Greek letter β or by the symbol $^{0}_{-1}$e.

In radioactive decay both mass number and atomic number are conserved. Thus in the decay chain of $^{238}_{92}$U the first two steps are written:

$$^{238}_{92}\mathrm{U} \rightarrow {}^{234}_{90}\mathrm{Th} + {}^{4}_{2}\mathrm{He} \qquad (1.1)$$

$$^{234}_{90}\mathrm{Th} \rightarrow {}^{234}_{91}\mathrm{Pa} + {}^{0}_{-1}\mathrm{e} \qquad (1.2)$$

Frequently, in such a chain, the half-life ($t_{1/2}$) for the radioactive decay is shown either above or below the arrow. A shorter notation is commonly used:

$$^{238}\mathrm{U} \xrightarrow[4.5\times10^{9}\,\mathrm{y}]{\alpha} {}^{234}\mathrm{Th} \xrightarrow[24\,\mathrm{d}]{\beta^-} {}^{234}\mathrm{Pa} \xrightarrow[1.1\,\mathrm{min}]{\beta^-} {}^{234}\mathrm{U} \xrightarrow[2.5\times10^{5}\,\mathrm{y}]{\alpha} {}^{230}\mathrm{Th}, \text{ etc.} \qquad (1.3)$$

where the half-lives are given in years (y) and days (d)[1]. The three naturally occurring radioactive decay series, which are known as the *thorium series*, the *uranium series*, and the *actinium series*, are shown in Fig. 1.2. A fourth series, which originates in the synthetic element neptunium, is also shown. This series is not found naturally on earth since all of the radioactive species have decayed away long ago. Both the present symbolism of the isotope as well as the historical (i.e. "radioelement") symbolism are given in Fig. 1.2. Note that the *rule of Fajans and Soddy* is followed in each series so that α-decay causes a decrease in atomic number by two units and mass number by four, whereas β-decay produces no change in mass number but an increase in atomic number by one unit. Moreover, we see a pattern occurring frequently in these series where an α-decay step is followed by two β-decay steps. All known isotopes of elements $_{92}$U to $_{81}$Tl are given in Figure 5.1.

1.4. Atomic models

Neither radioactive decay nor the discovery of isotopes provided information on the internal structure of atoms. Such information was obtained from scattering experiments in which a substance, such as a thin metal foil, was irradiated with a beam of α-particles and the intensity (measured by counting scintillations from the scattered particles hitting a fluorescent screen) of the particles scattered at different angles measured (see Fig. 12.4). It was assumed that the deflection of the particles was caused by collisions with the atoms of the irradiated material. About one in 8000 of the α-particles was strongly deflected through angles greater than 90^{o}. Consideration of these rare events led Rutherford in 1911 to the conclusion that the entire positive charge of an atom must be concentrated in a very small volume whose diameter is about 10^{-14} m. This small part of the atom he called the *nucleus*. The atomic electrons have much smaller mass and were assumed to surround the nucleus. The total atom with the external electrons had a radius of approximately 10^{-10} m in contrast to the much smaller radius calculated for the nucleus.

It was soon shown that the positive charge of the atomic nucleus was identical to the atomic number assigned to an element in the periodic system of Mendeleev. The conclusion, then, is that in a neutral atom the small, positively charged nucleus was surrounded by electrons whose number was equal to the total positive charge of the nucleus. In 1913 N. Bohr, using quantum mechanical concepts, proposed such a model of the atom which remains the basis of the modern atomic theory.

[1] IUPAC recommends a for *annum*, instead of y, however y will be used throughout this text as it remains the commonly used term.

Our understanding of the nucleus has grown rapidly since Rutherford's scattering experiments. Some of the important steps in the history of nuclear science are listed in Table 1.1. Many of these discoveries and their practical consequences are discussed in the subsequent text.

TABLE 1.1. *Historical survey of nuclear science*

	Essential steps in the development of modern science
~ 490–430 B.C.	**Empedocles** suggests that everything is made up of four elements: air, earth, water and fire. Every matter can be formed by transmutation between these. (This is principally correct if the four elements are interpreted as being the gaseous, solid and liquid states of matter, and fire interpreted as being energy.)
~ 460–370 B.C.	**Democritos** proposes that all matter consists of eternal, moving and indestructible atoms, qualitatively alike but differing in size, shape and mass.
1661	**Boyle** writes that the nature is made up of a limited number of substances (elements) which cannot be broken down into simpler ones.
1808	**Dalton**: All chemical compounds (molecules) are combinations of atoms in fixed proportions.
1896	**Becquerel** discovers radiation from uranium (radioactivity). The intensity of the radiation is measured either through its ionization of air or through the scintillations observed when the radiation hits a fluorescent screen.
1896–1905	**Crookes, Becquerel, Rutherford, Soddy, Dorn, Boltwood** *et al*. Radioactive decay is found to be transformation of atoms leading to different radioelements which are genetically connected in radioactive decay series.
1898	**P. and M. Curie** discover polonium and radium; the first radiochemical methods.
1898–1902	**P. Curie, Debierne, Becquerel, Danilos** *et al*. discover that radiation affects chemical substances and causes biological damage.
1900	**Villard** and **Becquerel** propose that γ-radiation is of electromagnetic nature; finally proven in 1914 by **Rutherford** and **Andrade**.
1900	**Becquerel**: β-rays are identified as electrons.
1902	First macroscopic amounts of a radioactive element (radium) isolated by **M.** and **P. Curie** and **Debierne**.
1903	**Rutherford**: α-radiation is shown to be ionized helium atoms.
1905	**Einstein** formulates the law of equivalence between mass and energy.
1907	**Stenbeck** makes the first therapeutic treatment with radium and heals skin cancer.
1911	**Rutherford, Geiger,** and **Marsden** conclude from measurement of the scattering of α-radiation against thin foils that atoms contain a very small positive nucleus.
1912	**Hevesy** and **Paneth**, in the first application of radioactive trace elements, determine the solubility of $PbCrO_4$ using RaD.
1912	**Wilson** develops the cloud chamber, which makes tracks from nuclear particles visible.
1913	**Hess** discovers cosmic radiation.
1913	**Fajans** and **Soddy** explain the radioactive decay series by assuming the existence of isotopes. This is proven by **J. J. Thomson** through deflection of neon ions in electromagnetic fields. **Aston** separates the isotopes of neon by gas diffusion.
1913	**N. Bohr** shows that the atomic nucleus is surrounded by electrons in fixed orbitals.
1919	**Rutherford**: first nuclear transformation in the laboratory, $^{4}He + {}^{14}N \rightarrow {}^{17}O + {}^{1}H$.
1919	**Aston** constructs the first practical mass spectrometer and discovers that isotopic weights are not exactly integers.
1921	**Hahn** discovers nuclear isomers: $^{234m}Pa(UX_2) \xrightarrow[1.2\ min]{\gamma} {}^{234}Pa(UZ)$.
1924	**de Broglie** advances the hypothesis that all moving particles have wave properties.
1924	**Lacassagne** and **Lattes** use radioactive trace elements (Po) in biological research.

1925–1927	Important improvements of the Bohr atomic model: Pauli exclusion principle, Schrödinger wave mechanics, Heisenberg uncertainty relationship.
1928	**Geiger** and **Müller** construct the first GM tube for single nuclear particle measurements.
1931	**van de Graaff** develops an electrostatic high voltage generator for accelerating atomic ions to high energies.
1931	**Pauli** postulates a new particle, the neutrino, to be formed in β-decay.
1932	**Cockcroft** and **Walton** develop the high voltage multiplier and use it for the first nuclear transformation in the laboratory with accelerated particles (0.4 MeV $^{1}H + {}^{7}Li \rightarrow 2\ {}^{4}He$).
1932	**Lawrence** and **Livingston** build the first cyclotron.
1932	**Urey** discovers deuterium and obtains isotopic enrichment through evaporation of liquid hydrogen.
1932	**Chadwick** discovers the neutron.
1932	**Andersson** discovers the positron, e^{+} or β^{+}, through investigation of cosmic rays in a cloud chamber.
1933	**Urey** and **Rittenberg** show isotopic effects in chemical reactions.
1934	**Joliot** and **I. Curie** discover artificial radioactivity: $^{4}He + {}^{27}Al \rightarrow {}^{30}P + n$; $^{30}P \xrightarrow[2.5\ min]{\beta^{+}} {}^{30}Si$.
1935	**DeHevesy** develops neutron activation analysis.
1935	**Yukawa** predicts the existence of mesons.
1935	**Weizsäcker** derives the semiempirical mass formulae.
1937	**Neddermeyer** and **Andersson** discover μ-mesons in cosmic radiation using photographic plates.
1938	**Bethe** and **Weizsäcker** propose the first theory for energy production in stars through nuclear fusion: $3\ {}^{4}He \rightarrow {}^{12}C$.
1938	**Hahn** and **Strassman** discover fission products after irradiation of uranium with neutrons.
1938–1939	**Meitner** and **Frisch** interprets the discovery by **Hahn** and **Strassman** as fission of the U-atom by neutrons; this is almost immediately confirmed by several laboratories in Europe and the USA.
1938–1939	**F. Joliot, von Halban, Kowarski** and **F. Perrin** in France apply for patents for nuclear chain reacting energy producing devices and starts building a nuclear reactor; the work is interrupted by the war.
1940	**McMillan, Abelson, Seaborg, Kennedy,** and **Wahl** produce and identify the first transuranium elements, neptunium (Np), and plutonium (Pu), and with **Segré** discover that ^{239}Pu is fissionable.
1940	Scientists in many countries show that ^{235}U is fissioned by slow neutrons, but ^{232}Th and ^{238}U only by fast neutrons, and that each fission produces two or three new neutrons while large amounts of energy are released. The possibility of producing nuclear weapons and building nuclear power stations is considered in several countries.
1942	**Fermi** and co-workers build the first nuclear reactor (critical on December 2).
1944	First gram amounts of a synthetic element (Pu) produced at Oak Ridge, USA. Kilogram quantities produced in Hanford, USA, in 1945.
1944	**McMillan** and **Veksler** discover the synchrotron principle which makes it possible to build accelerators for energies > 1000 MeV.
1940–1945	**Oppenheimer** and co-workers develop a device to produce fast uncontrolled chain reactions releasing very large amounts of energy. First test at Alamogordo, New Mexico, USA, on July 16, 1945 produces an energy corresponding to 20,000 tons of TNT; this is followed by the use of atomic bombs on Hiroshima (Aug. 6, 1945) and on Nagasaki (Aug. 9, 1945).
1944–1947	Photo-multiplier scintillation detectors are developed.
1946	**Libby** develops the ^{14}C-method for age determination.
1946	First Soviet nuclear reactor starts.
1949	Soviet tests a nuclear bomb.
1950	A nuclear shell model is suggested by **Mayer, Haxel, Jensen** and **Suess**.
1951	The first breeder reactor, which also produces the first electric power, is developed by Argonne National Laboratory, USA, and built in Idaho.
1952	The United States test the first device for uncontrolled large scale fusion power (the hydrogen bomb).

1953–1955	**A. Bohr, Mottelson,** and **Nilsson** develop the unified model of the nucleus (single particle effects on collective motions).
1955	**Chamberlain, Segré, Wiegand,** and **Ypsilantis** produce antiprotons.
1955	First nuclear powered ship (submarine *Nautilus*).
1954–1956	A 5 MWe nuclear power station starts at Obninsk, USSR, in 1954. First civilian nuclear power station (45 MWe) starts at Calder Hall, England, in 1956.
1956	**Reines** and **Cowan** prove the existence of neutrinos.
1957	Fire in carbon dioxide cooled graphite reactor at Windscale, U.K.
1957	Explosion in nuclear waste storage facility at Kyshtym (Chelyabinsk), USSR, with contamination of large areas.
1959	First civilian ship reactor used in the ice-breaker *Lenin*, launched in the USSR.
~1960	**Hofstadter** et al.; protons and neutrons contain unevenly distributed internal charge.
~1960	**Lederman, Schwarz** and **Steinberger** discover the muon neutrino.
1961	A radionuclide (^{238}Pu) is used as power source in a satellite (Transit-4 A).
1961	Semiconductor detectors are developed.
1963	End of atmospheric testing of nuclear weapons (see below).
1965	**A. Penzias** and **R. W. Wilson** discover the 3 K cosmic microwave radiation background.
~1970	Theory of quarks developed (**Gell-Mann**); quarks proven in nuclear scattering experiments (**Friedman, Kendall and Taylor**).
1972	French scientists discover ancient natural nuclear reactor in Oklo, Gabon.
1979	Core melt-down in PWR reactor at the Tree Mile Island plant near Harrisburg, USA; no environmental contamination.
1983	**Rubbia, van der Meer** & co-workers at CERN discover the W and Z weak interaction communicators.
1986	Explosion and fire in the Chernobyl-4 reactor unit at Pripyat, Ukraine, USSR, with contamination of large areas.

1955	Formation of United Nations Scientific Committee on the Effects of Atomic Radiation (UNSCEAR).
1957	Formation of the International Atomic Energy Agency (IAEA), with headquarters in Vienna.
1963	Partial Test Ban Treaty bans nuclear tests in the atmosphere, in outer space, and under water.
1968	Treaty on the Non-Proliferation of Nuclear Weapons (NPT) is signed by the "three depository governments" (USSR, UK, and USA), all nuclear weapons countries (NWC), and 40 other signatory, non-nuclear weapons countries (NNWC).
1971	The IAEA takes the responsibility for a safeguards system for control of fissile material in non-nuclear weapons countries.
1991	140 states have signed the NPT agreement.

1.5. Literature

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