The background of the cover is a high-magnification, colorized scanning electron micrograph (SEM) of a mineral surface. The image shows a complex, layered, and crystalline structure with various shades of blue and purple, suggesting a mineral like malachite or azurite. The texture is highly detailed, with visible crystal grains and surface irregularities.

LCC6 SPECIAL TOPIC REPORT

Radiochemistry in Nuclear Power Reactors
(Light Water Reactors)

Volume I

Radiochemistry in Nuclear Power Reactors (Light Water Reactors)

Volume I

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The Authors also want to thank Francis Nordmann and Wilfried Morell (as an ANT external reviewer) for reviewing this Special Topic Report on Radiochemistry in Nuclear Power Reactors (Light Water Reactors).

The technology described in the VGB documents has never been published, although it is in use for a considerable time. It would be desirable, if a group of young chemists who are specialists like the above mentioned colleagues would elaborate an updated version of this document.

The analytical procedures described as example in Appendix A.2 of this STR are internal procedures used in a German Nuclear Power Plant. They are based on several papers of open literature and on some thesis prepared by university students in collaboration with the before mentioned Power Plant. Unfortunately, an individual quotation is not possible. However, this should not be a problem because the user, who wants to apply these procedures, must adjust his own processes and calibration standards in advance.

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Preface

This Special Topic Report (STR) is written as an introductory text for Nuclear Chemists, i.e. Scientists with chemical background and chemical orientation whose primary interest is the study of nuclear properties and nuclear reactions. It is even more important for Radio chemists, i.e. Chemists concerned with the chemical manipulation of radioactive sources and with the application of radioactivity and other nuclear phenomena to chemical problems (whether in basic chemistry or in energy production, biology, medicine, earth and space scientists, etc.). However, in reviewing the available data on radiochemistry, it was decided to give the main emphasis to radiochemistry in Nuclear Power Plants (NPP) in order to avoid a splitting in too many directions. In arranging the subject matter, ANT International has been guided by the firm conviction that a good grounding in the fundamental of radioactivity and other nuclear phenomena is equally essential for the groups mentioned above. At the same time, this STR serves as a Handbook for Radio chemists working in NPP or other Utility Departments as well as many organisations related to Nuclear Energy such as Manufacturers, R&D Laboratories, technical support, Regulatory Body, etc.

Radiochemical surveillance of NPP is an important part of reactor operations. The plant operator is required to continuously monitor fuel performance, correctly account for release of radioactivity through gas and liquid effluents from the plant, and minimize the exposure of personnel to radiation. To accomplish these tasks, proper training must be given to the plant chemists (engineers and technicians).

A comprehensive textbook and/or manual is needed for their training, as well as for their use as a reference to develop good radiochemical procedures for routine analyses. Unlike a well-developed procedure in a research laboratory, a good procedure for routine analytical work in NPP should be simple and easy to follow, thus accuracy should not be compromised. Therefore, the first main objective of this STR is to provide the plant chemists not only with the fundamentals, but also with some major practical procedures collected from many years of reactor experience. The second main objective of this STR is to provide enough fundamental material to bridge the gap between industry and academia in the area of radiochemistry in NPP. It also may serve as an introduction to professionals in other related fields, such as health physics and nuclear engineering.

This STR deals with two major types of light water reactors: the Boiling Water Reactor (BWR) and the Pressurized Water Reactor (PWR/VVER).

In the first volume of this STR there will be a description of the following areas:

- Introductory remarks to “Radiochemistry”.
- Radioactivity production in NPPs.
- Sampling and measurement of radionuclides.

The second volume will describe radiochemical technologies in several major areas, like:

- Fuel performance surveillance and fission product chemistry.
- The chemistry and transport behaviour of activation products.
- Radiation chemistry in the coolant
- Assay of radioactive waste.
- Special radiochemical studies and tests in the reactor system.

Some selected procedures for sampling, radiochemical separation, and activity measurements are also included in the Appendices.

1 Description of nuclear power plants and primary coolant chemistry (Rolf Riess)

In an operating NPP about one hundred radionuclides can be identified. The reasons for monitoring these nuclides can be summarized as follows:

1. Track the occurrence, progression and characteristics of fuel defects.
2. Check for inadvertent ingress into the reactor coolant system of impurities that could affect dose rates, surface contamination, fuel performance, material integrity or airborne contamination after shutdown. Examples include chemical additive contaminants, ion exchange resin contaminants, recycled liquids, and resin ingress into the Reactor Coolant System (RCS).
3. Monitor filter and demineralizer performance.
4. Provide insights into the quantity and quality of the make-up water entering the RCS.
5. Monitor reactor coolant system events (e.g. crud burst, Control Rod Drive Mechanism (CRDM) surveillance, forced or refuelling shutdown, etc.) or equipment problems that result in release of materials (e.g. wear products) into the RCS.
6. Monitor the production of long lived radionuclides that will show-up in radioactive waste.

The primary emphasis in this report is to describe the technologies for measuring the radionuclides in the plant. However, in the introductory section there will be a description of the LWR systems and how their design, material selection, and water chemistry is connected to the production of radionuclides. Furthermore “Radiochemistry” is considered as an integral part of Chemistry Control in NPP.

1.1 PWRs

1.1.1 Introduction

A typical PWR reactor primary system is shown in Figure 1-1 and it consists of the reactor vessel with its internals that support the fuel core, piping for the coolant recirculation system, a pressurizer, steam generators (SG), pumps and valves. Auxiliary systems control the boron (B) and lithium (Li) and hydrogen additions and removals as well as a demineralizer clean-up system for removal of impurities and a system to provide make-up water.

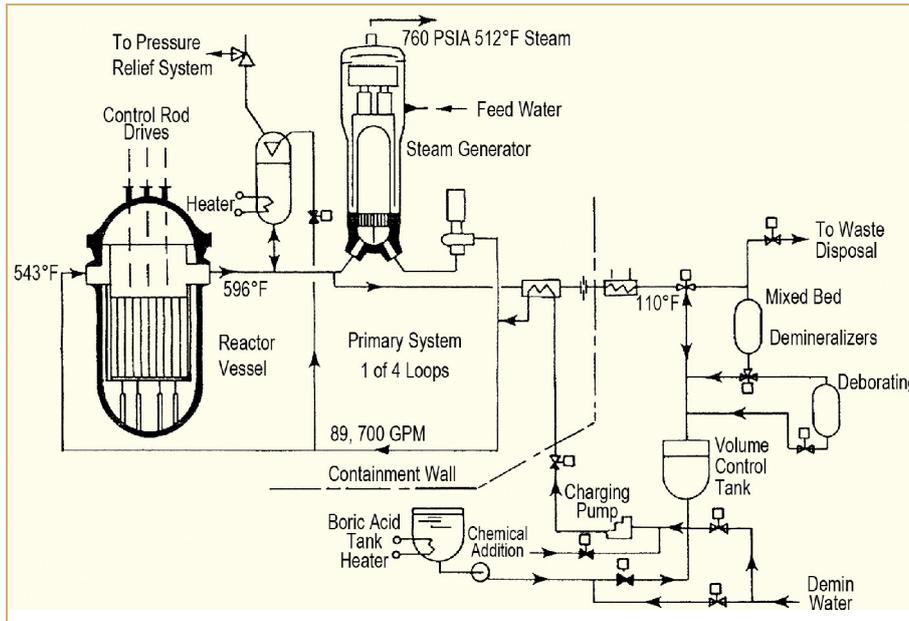


Figure 1-1: Primary coolant systems for a large PWR [Cohen, 1985].

The chemical and volume control system is of specific importance for Chemistry and Radiochemistry. It is designed to control the volume of primary coolant as well as its chemical composition.

1.1.2 Main components

The primary system operates at a pressure of 2,250 psia (15.5MPa) and the *pressure boundary* consists of the reactor pressure vessel, the recirculation piping, pressurizer, SG tubing and pumps as shown in Figure 1-2. There are *heat transfer surfaces* within the system that consist of the fuel cladding transferring heat to the primary coolant (approximately 25% of the total RCS surface) and the SG tubing transferring heat to the secondary coolant (about 65% of the total RCS surface). The gamma heating of all the components within the reactor vessel is relatively minor, but is also removed by the coolant.

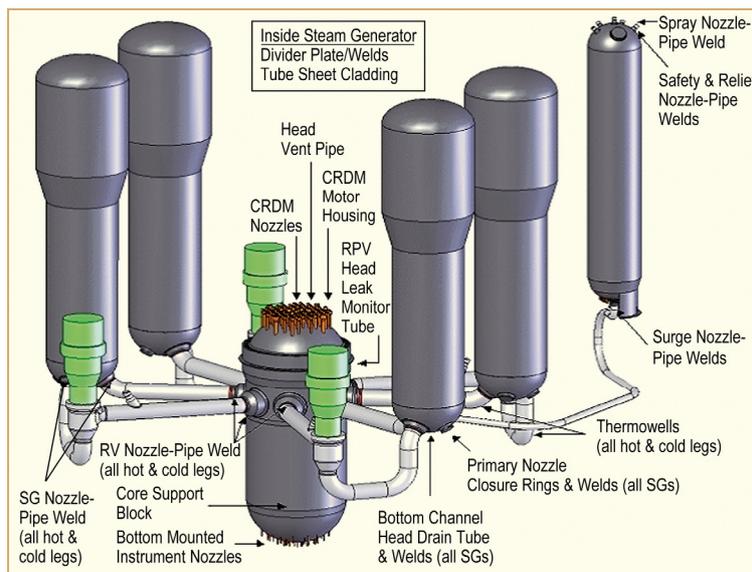


Figure 1-2: PWR primary system – pressure boundary, [Garbett, 2006].

2 The structure of the atoms (Rolf Riess)

2.1 Chronology of the model development

The discovery of radioactivity and the discovery of the electron were the starting points for theories of the atomic structure. One of the problems associated with the discovery of the electron as a fundamental constituent of matter was how to explain the electrical neutrality of the atom. Authors [Magill et al, 2006] have summarized the chronology of the various steps, which led finally to the Bohr/Sommerfeld model of the atoms which is the basis or background for all the discussions coming up in this STR. According to [Magill et al, 2006], a first step was taken by Kelvin and Thompson who proposed more or less at the same time the first models of the structure of atoms. Kelvin proposed in 1902 that the negative charges of the electrons and the positive charges are cancelling out each other. He considered the atom to consist of a sphere where the mass and the charge are distributed uniformly with electrons embedded like “plums in a pudding”. The model also described that electromagnetic radiation would be emitted if external forces would generate vibrations in the atom. Thompson proposed a modification of this model in which the electrons are moving at high speed in concentric circles in a sphere of continuous positive charge.

In the next step, Rutherford, 1911, postulated that in contrast to the “plum pudding model” the positive charge of the atom was concentrated in a central nucleus much smaller than the atom which contained most of the atoms mass, with the negative electrons orbiting around the nucleus similar to the way planets move around the sun. Each atom with its specific number of electrons must contain an equal and opposite number of positive charges in order to ensure electrical neutrality. Since the electron is the fundamental unit of negative charge, Rutherford stated that the unit of positive charge in the nucleus is the proton.

In order to confirm these ideas, experiments were performed to support the theory existing at that time. Although the ideas were revolutionary at the time, there were two unsatisfactory aspects of the proposed atomic structure: the first concern was how the electrons are held in place outside the nucleus and the second how the protons can be held together in view of the strong repulsive forces of the positively charged particle.

The idea of a small solar system with the electrons orbiting the positively charged nucleus seemed attractive, but it was unacceptable to classical theory. According to this theory such orbiting electrons experienced a radial acceleration and should emit radiation. As a consequence they would lose their kinetic energy and spiral into the nucleus. Another limitation of the model was the fact that it could not account for the emission spectra of atoms which were identified at that time and which could be used as a fingerprint to identify atoms. The solution for all these questions of Rutherford’s model was developed by Bohr, 1913. He developed a model based on the new quantum theory proposed by Planck. Planck had shown that bodies emit radiation only in discrete amounts which are some multiple of $h\nu$, where ν is the frequency of the radiation and h known as Planck’s constant. These ideas were further substantiated with the discovery of the photoelectric effect by Einstein. In his investigation of how electrons are emitted from metal plates under the action of ultraviolet light, Einstein showed that the energy of the emitted electrons depends only on the frequency of the incident light and not on its intensity. He showed further that the light must be composed of discrete “photons” each with the energy $h\nu$. On the basis of Planck and Einstein’s work, Bohr developed a new model for the atom. He proposed that atoms are in stationary states and that any emission of energy is associated with a transition of one state to the other. On this basis, emitted radiation must satisfy the condition $h\nu = E_1 - E_2$. Based on this model, Bohr could explain that electrons are orbiting around the nucleus in discrete energy states without emitting radiation and secondly when electrons jump from a higher energy E_2 state to a lower energy state E_1 , radiation of the frequency $h\nu$ is emitted.

Sommerfeld, 1916, extended Bohr's circular orbit, with the main or principle quantum number n to an elliptical orbit with an azimuthal quantum number l . Shortly later the magnetic quantum number was introduced to account for the effect of magnetic fields. These three quantum numbers n , l , and m can be regarded as giving the size, shape, and spacial orientation of the orbit.

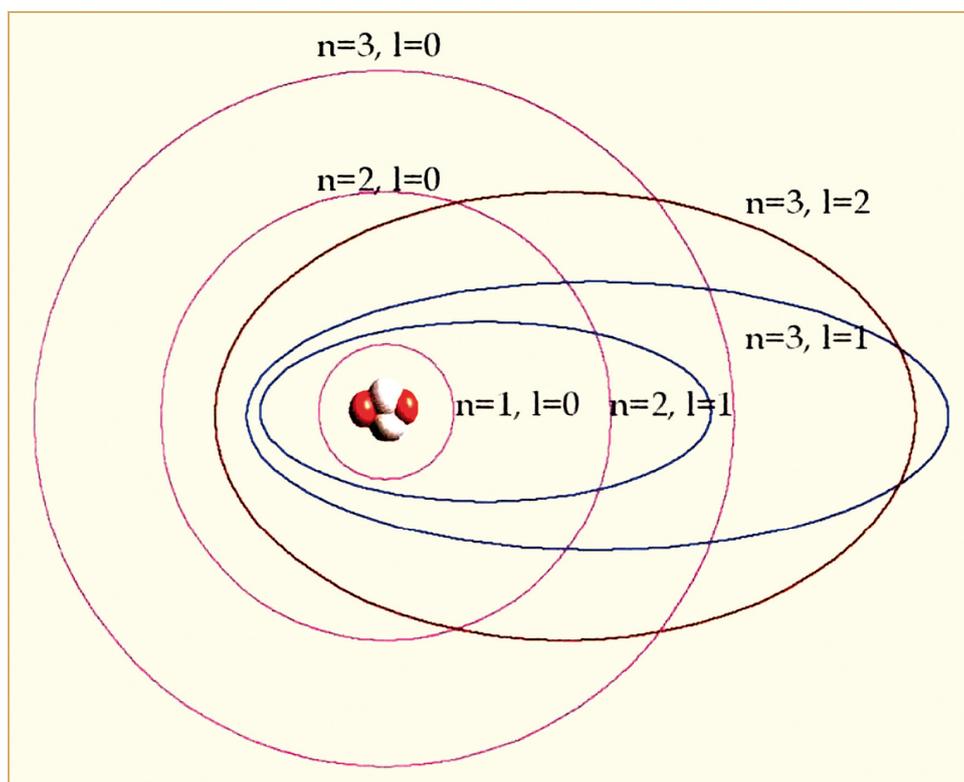


Figure 2-1: Bohr-Sommerfeld model of the atom [Magill et al, 2006].

The second difficulty of Rutherford's model of the atom was to explain how the protons can be held together in view of the strong repulsive forces of the positively charged particles. Hydrogen, the simplest atom, consists of a single proton in the nucleus. If heavier nuclei contain multiple protons, then the mass number and the atomic numbers should be the same. This was not the case. The mass numbers were found to be approximately twice the atomic numbers. These difficulties were resolved by the discovery of the neutron by Chadwick, 1932. Its presence in the nucleus explains the difference between the atomic and the mass number. More important the neutron is responsible for the cohesive force that holds the nucleus together. This nuclear force is attractive and extremely short range of about $2-3 \times 10^{-15}$ m. In addition, because of the very short range of the nuclear force, neutrons can only interact with their nearest neighbour nucleons in contrast to the longer range repulsive electrical forces of the protons. For this reason, in a stable nucleus, the number of neutrons must increase more rapidly than the number of protons. The discovery of the neutrons also explains the existence of isotopes discovered for radioactive elements.

In addition to the proton, neutron, and electron, there are considerably more than 100 other fundamental particles which have been discovered or hypothesized. The majority of these fall into one of two classes, leptons or hadrons. However, these findings are not relevant for the discussion in this STR. It should only be mentioned that nowadays about 3000 nuclides are known, of which 10% are stable.

3 Radioactivity in nuclear reactors (Rolf Riess)

3.1 Introduction

Operation of nuclear power plants is inevitably connected to radionuclides not only as components of the fuel in the fuel rods, but also in the reactor and the connected systems. According to their chemical features, the nuclides can be dissolved or suspended in water and steam, or can be bound to materials or corrosion products on the surfaces of the construction materials. During the operation of a NPP, the behaviour of the radionuclides may also change due to the existing electrochemical potential.

In PWRs the chemical environment is “reducing”, which means, the radionuclides will be in the lowest stable oxidation state. For example, the Iodine does exist as I⁻ (Iodide) but during shutdown conditions I₂ may be formed due to the switch-over to an oxidizing environment. Other examples for chemical elements changing their oxidation state due to changes in the ECP are: Fe and Cr.

In BWRs the situation is even more complex: under NWC-conditions all elements are in the highest stable oxidation state. However, during HWC operation the ECP values are changing permanently leading to an increased metal release rate of the system surfaces. This instable situation led to the introduction of zinc injection because zinc is stabilizing mixed oxide layers.

There are two different types of radionuclides differing by the mechanism and location of its generation. One type is fission products from the fuel, which can leave the fuel rods and enter the coolant by leakages or in a small extent by diffusion. The other type is activation products, which are generated in the neutron field of the reactor. These nuclides can be differed between those being generated out of corrosion products and those coming from ingredients of the coolant.

Differentiation and identification of the radionuclides is possible by radiochemical and physical measurements, what means chemical separation procedures and differentiating physical measurements. Analytical procedures can be found in Appendix A.

The purpose of the radiochemical measurements are:

- Determination of the concentrations for fission- and activation products during constant operation and during load changes in order to monitor the tightness and integrity of the fuel.
- Monitoring the behaviour of released fuel.
- Control the function and efficiency of purification systems.
- Monitoring the auxiliary systems because of contamination by radionuclides and control the “secondary barrier” for its tightness.
- Monitoring the released water or gas because of possible activity release according to the local and international regulations.

In order to comply with these demands, adequate laboratories, equipment, measuring devices, and skilled personal have to be available.

The radionuclides and their emitted radiation are described below. Furthermore the behaviour of the different radiation when passing through matter is described as well as the measurement methods for the different radiators.

3.2 Terms and definitions

Activity

Activity is the number of decaying atoms of a radionuclide per time unit. The unit of activity is Becquerel (Bq).

$$1 \text{ Bq} = 1 \text{ decay per second}$$

Previously also Curie was used which is defined as follows:

$$1 \text{ Curie (Ci)} = 3.7 \times 10^{10} \text{ decays per second}$$

$$1 \text{ Bq} = 2.7 \times 10^{-11} \text{ Ci}$$

The units Ci and Bq do not indicate whether alpha or beta particles or gamma quanta are emitted.

Activity concentration

For more details see Section 3.3.1.6.

The activity concentration is the amount of activity of a radionuclide which is contained in a defined volume of liquid or gas for a certain pressure and temperature.

Alpha transformation (alpha decay)

The alpha transformation is the radioactive transformation under emission of an alpha particle.

Beta transformation (beta decay)

The beta transformation is the radioactive transformation under emission of a beta particle. This includes also the capture of an electron of the shell.

Capture

Capture is a process by which an atom or a nuclear system swallows another particle. During this process only photons or neutrinos are emitted.

Carrier

A carrier is a substance which does exist in an amount which can be weighed and which during a chemical or physical process carries along certain ingredients, which exist only in trace amounts.

Compton effect

The Compton effect is the interaction of x-rays or gamma rays with matter. The Compton effect is the elastic scattering of a photon caused by an electron in case it can be seen as free and stationary.

A part of the energy and the impulse of the incoming photon will be transferred to the electron and the residual energy remains with the scattered photons.

Contamination (radioactive)

The radioactive contamination is a pollution by radioactive substances.

Daughter nuclide

A daughter nuclide is the immediate decay product of a radionuclide.

Decay constant

The decay constant (λ) is the fraction of transferred atoms per time unit.

Decay series (radioactive)

Radioactive decay series consists of the total number of nuclides which can be generated by a radionuclide by radioactive transformation, except the spontaneous fission.

Energy absorption

The energy absorption is a process by which incoming radiation transfers its energy partial or in total to the matter which is to be crossed.

Equilibrium (radioactive)

A radioactive equilibrium is the status in which a radioactive decay series is established and fulfils certain conditions. Such a condition is given when 1. The half life of the mother nuclide is bigger than the half life of the succeeding products and 2. A certain time has passed which is longer than the half life of the daughter nuclide with the longest half life.

Gamma radiation

Gamma radiation is the penetrating electromagnetic radiation coming from the atomic nucleus. The energies of gamma radiation are between 0.01 and 10 MeV. Also x-rays can exist in this energy range but their origin is not the atomic nucleus.

Geometry

Under the aspect of radiation measurements, geometry means the local arrangement of the sample to be measured and the radiation detector.

Half life (radioactive)

Half life is the time in which half of the original atoms of a radionuclide are decayed or in case of isomers have been transferred to the ground state.

Isobars

Isobars are nuclides with the same mass number M , *which is the sum of proton and neutron numbers*. Isobars are nuclides of different chemical elements because of their different proton number Z . Their neutron number N is also different because the mass number remains the same.

Isomers

A nuclide isomer is a type of those atoms of the nuclide whose nuclei are in a certain metastable and excited condition. Transition to an energetically lower state, mainly the basic state, occurs under emission of gamma radiation.

Isomer status

An isomer status is a status of a nucleus with a lifetime which is considerably greater than the average life time of a normal excitation.

Isotones

Isotones are nuclides with the same number of neutrons but differing number of protons. Isotones neighbour each other vertically.

Example: Carbon-14, Nitrogen-15, Oxygen-16 in the table of Radionuclides (Table A-1).

Isotopes

Isotopes are nuclides with the same number of protons but differing numbers of neutrons; that is, they have the same atomic number and are therefore the same chemical element. Isotopes neighbour each other horizon ally e.g. Carbon-12, Carbon-13, and Carbon-14 (See Figure 3-1 below).

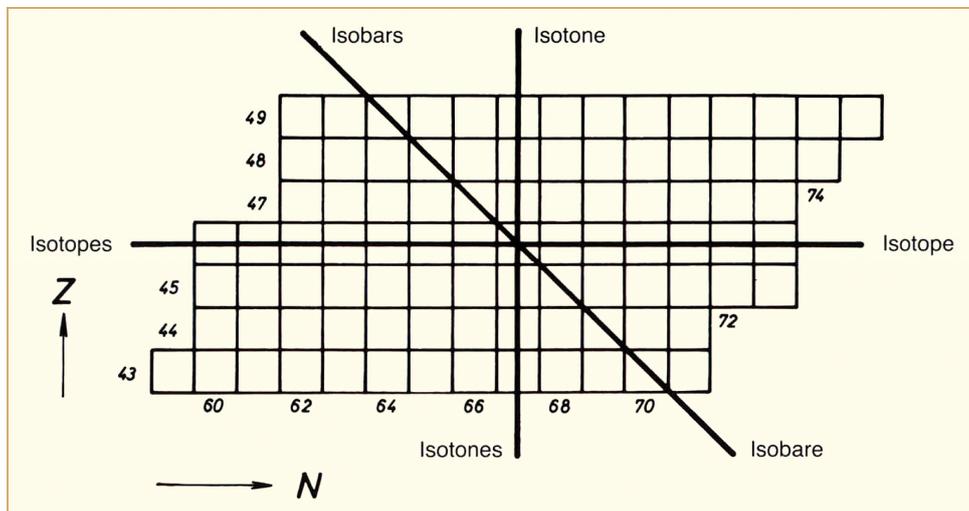


Figure 3-1: Arrangement of the defined nuclides in chart of nuclides (see also Section 3.2).

Mother nuclide

A mother nuclide is a radionuclide which precedes the specific radionuclide in the decay chain.

Nuclear charge number Z

The nuclear charge number Z is also referred as atomic number and it defines also the number of electrons in the electron shell. Thereby the type of chemical element is established.

The role of the neutrons is to stabilize the atomic nucleus. At small atomic numbers their number N is about equal to the number of protons. However, with increasing nuclear charge numbers an increasing neutron surplus is to be observed.

3.5 Chart of nuclides

The chart of nuclides diagram can be shown by using the Karlsruhe nuclide chart (Figure 3-10).

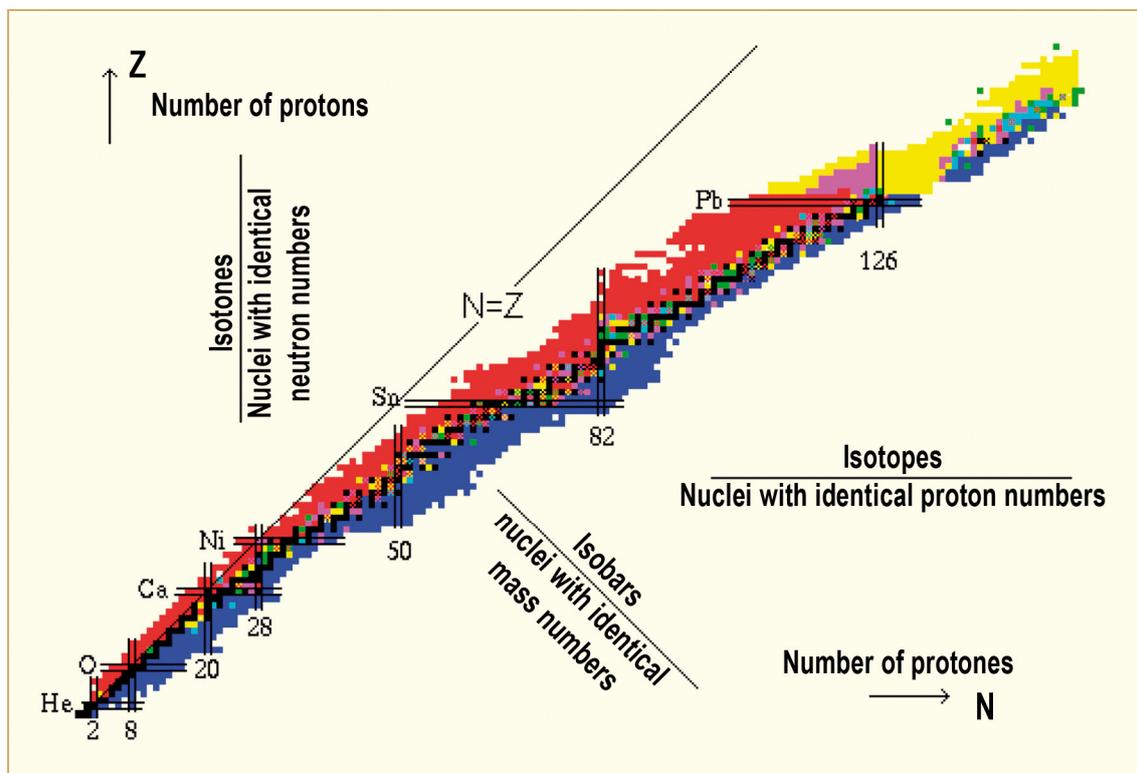


Figure 3-10: Karlsruhe nuclide chart.

A chart or table of nuclides is a simple map to the nuclear or radioactive behaviour of nuclides, as it distinguishes the various isotopes of an element. It contrasts with a periodic table, which only maps their chemical behaviour, since isotopes of the same element do not chemically differ. Nuclide charts organize isotopes along the X axis by their numbers of neutrons and along the Y axis by their numbers of protons, out to the limits of the neutron and proton drip lines. This representation was first published by Giorgio Fea in 1935, and expanded by Emilio Segrè in 1945. In 1958, Walter Seelmann-Eggebert and Gerda Pfennig published the first edition of the Karlsruhe Nuclide Chart. Its 7th edition was made available in 2006. The authors of this edition are [Magill et al, 2006], and the document can be purchased at Marktdienste Haberbeck, Industriestraße 7, D-32791 Lage.

Today, one finds several nuclide charts, four of them have a wide distribution: the Karlsruhe Nuclide Chart, the Strasbourg Universal Nuclide Chart, the Chart of the Nuclides from the IAEA and the Nuclide Chart from Knolls Atomic Power Laboratory (KAPL). In this STR, the Karlsruhe Nuclide Chart is the selected document. Regarding the KAPL product, it shall be mentioned that the 17th Edition is targeted for a mid 2010 release. All names in this section are taken again from the description of the Karlsruhe Nuclide Chart of 2006.

Nuclide charts are based upon the proton-neutron model of the nucleus and are essentially a plot of the number of protons vs. the number of neutrons in stable and unstable nuclei.

The charts contain information on the basic nuclear properties of known nuclides. Each nuclide is represented by a box containing basic nuclear data. This data consists of the half-life, neutron cross-sections, main gamma lines etc. of that nuclide. An important characteristic of the charts is the use of colour to denote the mode of decay, half-life or cross section. If the nuclide has one or more meta stable states, the box is subdivided into smaller boxes for each state.

It can be seen that stable isotopes lie within a relatively narrow range, indicating, that the neutron to proton ratio must have a certain value or range of values to be stable.

Radioactive nuclei mostly lie outside this range. The plot also shows that for low atomic numbers the neutron to proton ratio is 1. At higher atomic number, this value increases, indicating a higher ratio of neutrons to protons in heavy atoms.

The extremities of the white regions above and below the region of stabilities are known as the proton and neutron “driplines” beyond which nuclei are extremely unstable (i.e. if a nucleon is added it will “drip” out again). As nucleons are successively added to a nucleus on the stability line, the binding energy of the last nucleon decreases scatterly until it is no longer bound and the nucleus decays by either neutron emission or proton emission.

Nuclei with even numbers of protons and neutrons are more stable than nuclei with other combinations of neutrons and protons. For odd numbers of protons and neutrons, there are very few stable nuclides. The stability of nuclei is extremely significant for special numbers of protons and neutrons. These (magic) numbers are 2 – 8 – 20 – 28 – 50 – 62 – 126 and correspond to full shells in the shell model of the nucleus. The element tin (Sn) with the proton number $Z = 50$ for example has 10 stable isotopes, more than any other elements.

When the proton and neutron numbers both have magic values, the nucleus is said to be “doubly magic”. Doubly magic, stable nuclides are for example He-4, the alpha particle, as well as the nuclide Pb 208, which is reached in several decay processes, for example in the decay chain.

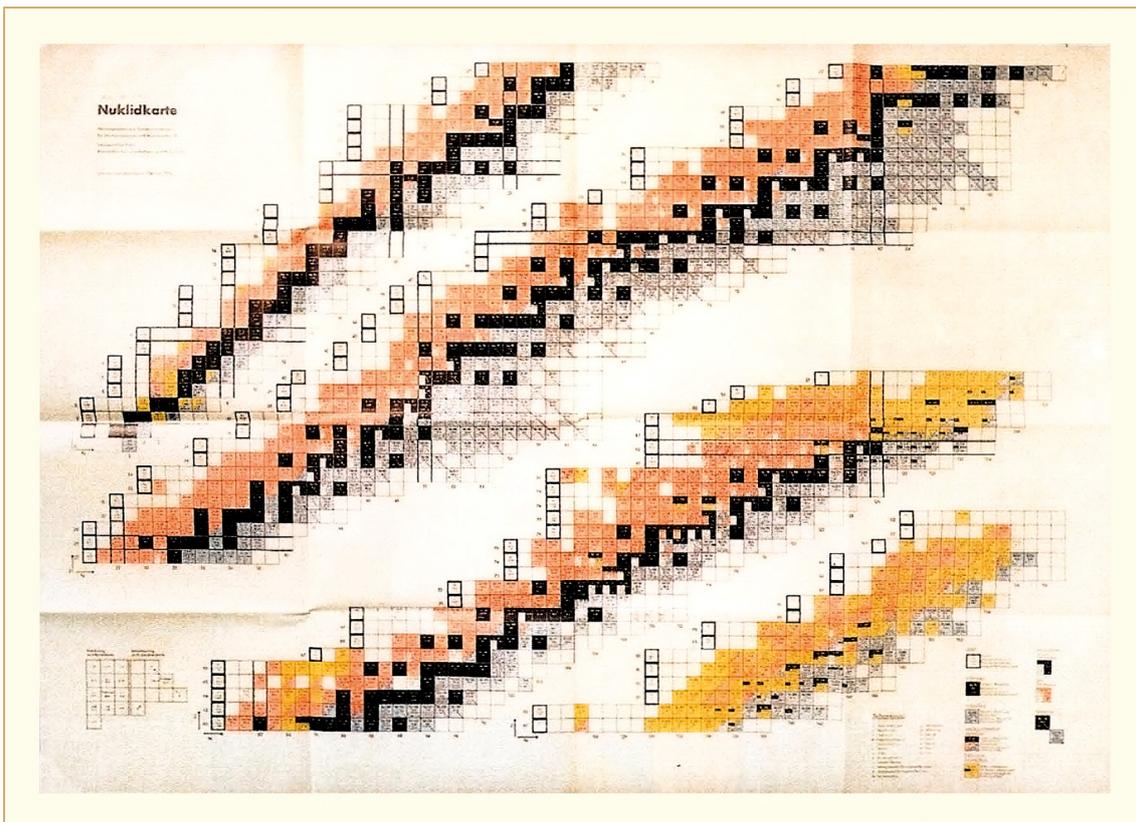


Figure 3-11: First edition (1958) of the Karlsruhe chart [Seelmann-Eggebert & Pfennig, 1958].

Explanation of the chart of the nuclides

In the latest version of the chart of nuclides, each experimentally observed nuclide is represented by a square containing the symbol of the element and the number of nucleons A. In the charts, the nuclides are arranged such that the proton number Z is given on the ordinate and the neutron number $N = A - Z$ on the abscissa.

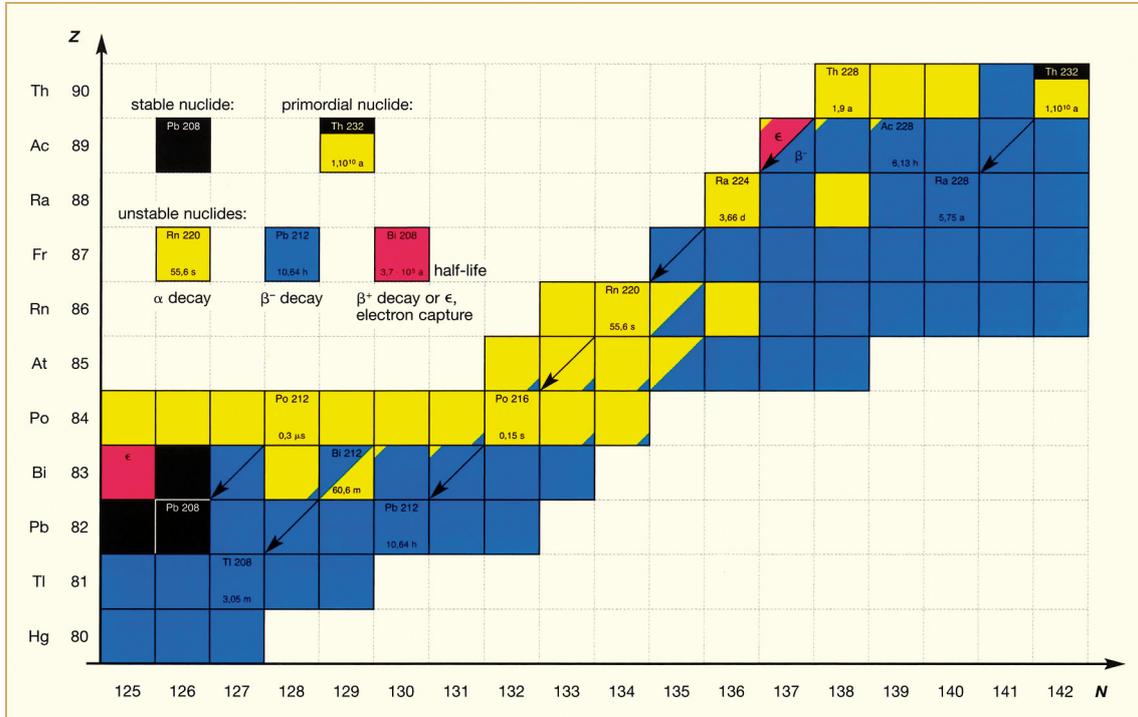


Figure 3-12: Section of the Karlsruhe Nuclide Chart showing the radioactive decay of Th-232.

In the present nuclide chart update, the 1998 edition of the Karlsruhe chart was compared to the NUBASE 2003 evaluation [Audi et al, 2003] to establish a list of nuclides which were not present in the 1998 version. From the list, nuclides were selected which have been measured experimentally. In particular nuclides were selected where the half-life of the mass has been determined or the nuclide has been clearly identified. Where a nuclide has been identified but the half-life has not been measured a detection limit for half-life is given (greater or lower than a value).

Metastable states, which do not undergo α -, β -decay or spontaneous fission, i.e. decay only by gamma-emission are included only if their half-life is larger than 1 s. Where emission of a particle results from a resonance state in unstable nuclides, both the resonance width and corresponding half-life are given.

For mass numbers in the range $A = 266-294$, the latest nuclear data sheet, rev. 2005, are used. For the period not covered by NUBASE, i.e. 2003 until summer 2006, nuclide information has been taken from nuclear data sheet 100-107.

4 Sampling (Suat Odar)

4.1 Introduction

In a NPP, one of the most frequent mistakes and largest errors introduced in radiochemical analysis is sampling and sample preparation. Sampling the reactor coolant, off-gas, liquid waste, and airborne samples, etc. may present different problems in each case. Some problems can be avoided but some can only be minimized.

Sampling and sample preparation is a very important and challenging issue in NPP. During sampling the reactor coolant, off-gas, liquid waste, and airborne activities, different critical issues may arise. Some of these issues may be avoided but some can only be minimized.

The procedures described in this section are based on many years of experience. Some techniques are standard some are modified to suit the needs which are requested by the plant manufacturer.

All primary water chemistry guidelines are requiring to monitor the chemistry and radiochemistry to control the applied chemistry regime. The medium to be sampled and their sampling locations, depending on reactor type, might be different, but for the same reactor types these sampling locations are worldwide almost the same. The essential sampling locations mandatory for plant start-up, shutdown, and power operations are described in the following sections.

4.2 Sampling locations

4.2.1 PWR plants

All PWR plants have sampling points at three locations to sample and monitor the reactor coolant (see Figure 4-1). These locations are the RCS loop piping; the Pressurizer and the Chemical and Volume Control System (CVCS).

The RCS loop sampling is usually used to sample the reactor coolant during the power operation, plant start-up and shut-down operating modes to monitor the chemistry and radiochemistry parameters. The RCS loop sampling is usually located at the hot leg. In some Siemens designed PWRs, besides of hot leg sampling, cold leg loop sampling is also considered. The hot leg sampling together with cold leg sampling can provide valuable information regarding the corrosion product transportation in the reactor coolant system. The sampling of the pressurizer is used mainly during plant start-up to monitor the coolant oxygen content or during plant shut-down to monitor the dissolved hydrogen content, if necessary. There are three sampling locations in the CVCS at its let-down line. Two of them, at up- and down-stream the ion exchange resin filters for coolant purification, are used to monitor the purification efficiency of the resin columns during power operation. The third one at the downstream HP Charging pump is used in several PWRs (in all Siemens designed PWRs) for oxygen control.

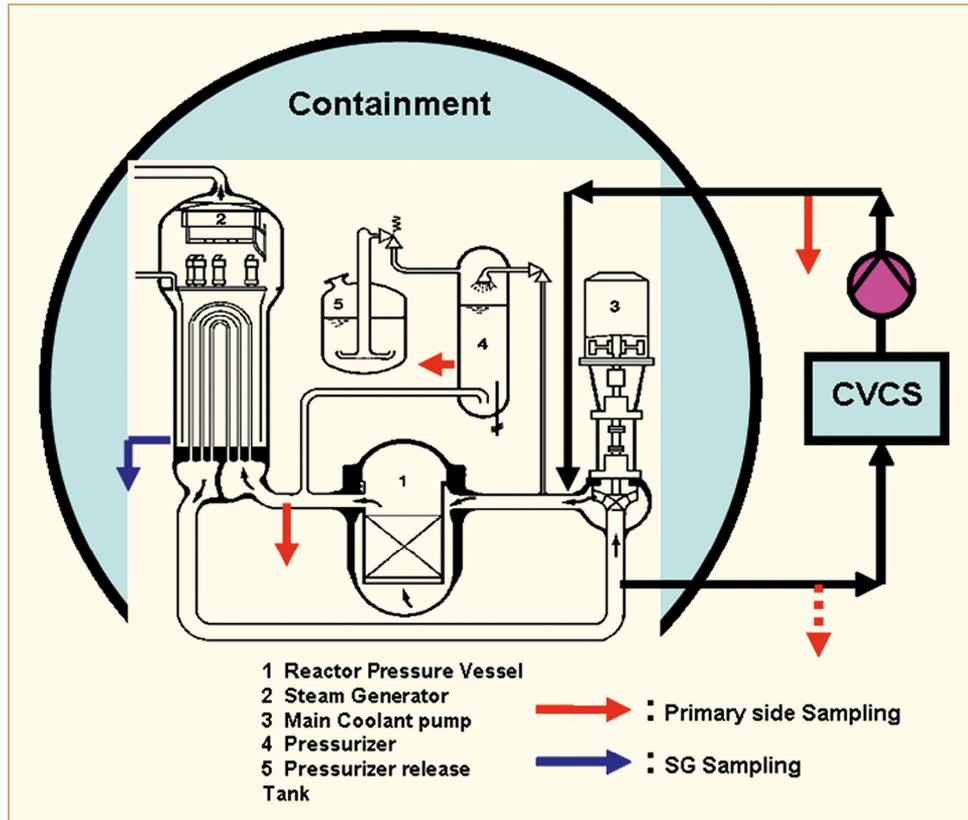


Figure 4-1: PWR primary side sampling locations (schematic).

In US PWR plants designed by Westinghouse and CE, the RCS sampling location is normally at the hot leg loop piping, although a RCS cold leg sample may also be available in several plants. B&W PWRs use one of the RCS cold leg sample lines for the RCS coolant sampling. The RCS hot leg (or B&W cold leg) line usually has a delay coil to provide 60 sec or longer delay to reduce nitrogen-16 activity. Usually the sampling line is made from 10 mm (3/8-inch) stainless steel tubing and is routed through the containment wall to a central sampling room located in the nuclear auxiliary building. Typical sampling line lengths can be up to 200 m long. Normally, two containment isolation valves are fitted to the RCS sampling line. At most US PWRs these double containment valves are not allowed to remain open due to safety considerations. They must be opened each time when RCS coolant sample needs to be collected. This is a big handicap to get representative samples, as it will be discussed later. At most of the European PWRs, these containment isolation valves remain normally open and a continuous sample line flow exists to a boron monitoring equipment and/or to volume control tank.

Beside these sampling locations, almost in all PWRs, the SG blow-down sampling is also considered in the design of primary side sampling system, although they sample the SG secondary side water. This is because of their location in the reactor building / containment and due to the risk of radioactive contamination in case of SG tube leakage. This SG sampling is usually also used for monitoring the SG leak tightness. In addition, there are numerous sampling systems for nuclear auxiliary systems, like in CVCS, coolant purification system, coolant degasification system (if available), Residual Heat Removal (RHR) system, boron recycling and reactor make-up systems, etc. Depending on the plant design, these nuclear auxiliary system samplings might differ from plant to plant.

Usually, all RCS sampling lines end in a box in the hot chemistry laboratory. In this sampling rack box, provisions exist to separate the suspended corrosion product particles or gases (hydrogen and/or radioactive noble gases) from the coolant without contamination. The lengths of all sampling lines within the reactor containment are, depending on vendors reactor design, in the range of 70 to 140 meters. For example, for the Westinghouse designed PWR Ringhals-3, the length of two hot leg sampling lines is reported to be 103 and 127 m respectively, [Polley & Anderson, 1989]. The lengths of the hot and cold leg loop sampling lines in Siemens designed PWR Philippsburg-2 are 70 and 110 meters respectively [Rühle, 2005]. It should be noted that the RCS sampling lines within the reactor containment are all only isolated pipes having only double isolation valves just at the containment walls. The sampling coolers and pressure reducers are usually all outside of the containment, and close to the sampling rack. Accordingly, the RCS sampling lines are high temperature lines, containing oxide scales and deposits on their pipes inner surfaces. As it will be discussed later, these oxide scales and high temperatures in the long sampling lines may change the chemical composition of the sampling medium, due to chemical reactions or interaction with the oxide scales.

The sampling line cooler in later Siemens designed PWRs is located in the containment (reactor building), providing the cooling for the most of the sampling line length, which gives probably more reliable results. In contrary to RCS sampling line, CVCS sampling lines up-stream coolant purification system are relatively cold and short. Therefore they can be considered as a good alternative for representative coolant sampling, as confirmed by field experience in several PWR plants.

Usually RCS sampling systems are not designed to obtain representative samples of trace corrosion product species, and radio-cobalt isotopes. Instead, the design criterion was to deliver a representative liquid sample for soluble boron and soluble fission products samples without exposing the operator to excessive radiation. The sample lines installed are long, have inadequate sampling nozzles, and are small diameter tubes, which have a high surface to volume ratio, so that significant interaction can occur between the sample medium and oxide layers on the sample line surface. This interaction and also uneven temperature distribution over the sampling line length can significantly affect the sampling composition, as it will be discussed later.

As discussed in LCC4 Annual Report Section 3, for particulates in liquid sampling medium, it is not necessary to use isokinetic sampling nozzles; the RCS samplings are also not isokinetic. In some PWRs, so-called capillary sampling lines are used, which are actually also not isokinetic.

4.2.2 BWR plants

Worldwide water chemistry guidelines for BWR plants require the control of water chemistry in the reactor water and in the FW. Radiochemistry parameters are usually monitored in the reactor water. Accordingly, in all BWR plants the essential sampling locations are up-stream and down-stream Reactor Water Clean-Up (RWCU) system and in final FW system before entering into the reactor vessel. Beside these sampling locations, there are more sampling systems which belong to the nuclear sampling system of BWR plants, such as:

- Main steam and re-heated steam,
- Moisture Separator Reheaters (MSR) drains,
- Condensate at up- and down-stream Condensate Polishing System (CPS),
- Condenser hot wells for leak monitoring.

Worldwide in all BWR plants, the on-line conductivity measurements are used for impurity control. In addition to conductivity measurements, depending on the plant specific chemistry control programs, like HWC, NMCA and/or Zinc Chemistry, additional adequate analytical monitoring equipment are used.

Due to SCC experienced in reactor vessels and at their internals, the ECP measurements in/ or close to reactor vessel for control of oxidizing conditions are getting mandatory for many BWR plants.

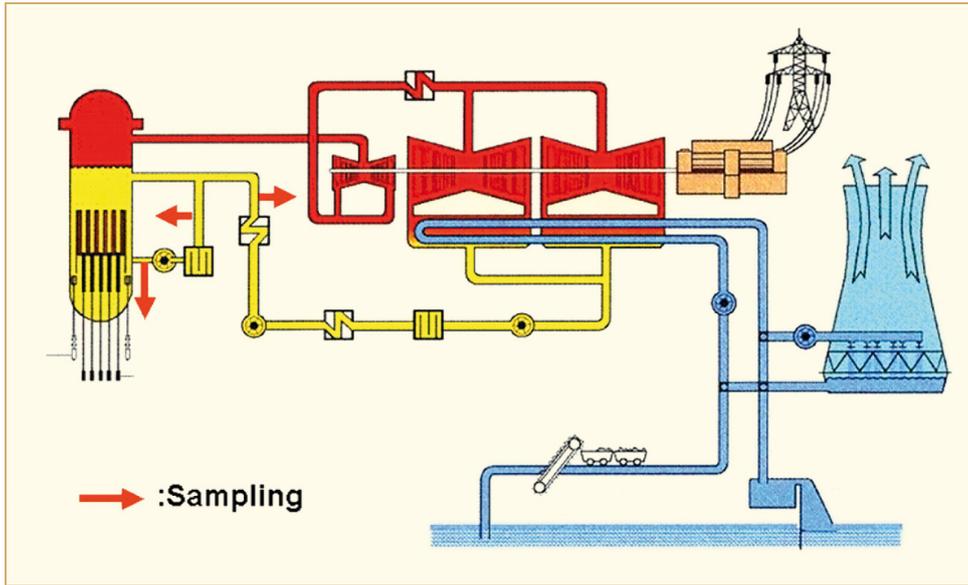


Figure 4-2: Essential sampling locations in BWR plants.

4.2.3 VVER units

In VVER Voda Voda Energo Reactor (a Russian designed PWR) plants, the reactor coolant is sampled usually directly from the RPV (2 lines: TV30 (40) S07) and at primary coolant cleanup systems (see Figure 4-3), [Yurmanov, 2008]. The later includes primary coolant blow down system TC (4 lines after high temperature filters at standardized V-320 NSSS modification or 2 parallel Mixed Bed at 1st VVER-1000 generation) and primary FW cleanup system Total Elongation (TE) (2 chains of 3 exchangers). There are also 2 lines from upper (steam phase) and bottom part (water phase) at the pressurizer. In these sampling media impurity concentrations, oxygen, and treatment chemicals (ammonia, hydrogen, potassium, lithium, sodium, and boron) are monitored. Whereas pressurizer sampling is usually used only during plant start-up and shutdown operations, the RPV and TC & TE coolant sampling is used during all operational modes. During power operation, dissolved oxygen is monitored in the coolant and deaerator let-down line downstream HP charging pump for oxygen control by means of high sensitive analysers. Simultaneously measured corrosion product concentration (Fe, Ni, Cr, Co, Zr) and activity (A) in RPV, TC and TE sampling can provide valuable information regarding the corrosion product transportation within the RCS, including corrosion product removal from RCS by TC & TE systems.

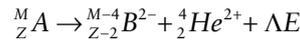
5 Alpha radiation measurement (Rolf Riess)

5.1 Physical properties of α -emitting nuclides

Alpha-radiation is an ionizing radiation emitted as a result of α -decay of unstable nuclides. It is a corpuscular radiation consisting of doubly charged helium ions:



Alpha-particle emission is therefore always accompanied by a decrease of two in atomic number and a decrease of four in mass number:



In case that the α -emission takes place between a single energy level of the α -emitting nucleus and the ground state of the product nucleus, alpha-particles emitted from a given nuclide all have the same energy so that the resulting energy spectrum is a line spectrum.

The emission of α -particles of several different energies by one nuclear species may be due to the existence of several energy levels either preceding or following the α -emission.

As α -decay, in any cases, gives line-spectra, the measurement can be used for determination of the emitting nuclide.

In most cases the emission of several α -particle groups of different energies from a given nuclide is due to the fact that the product nucleus can be left in different excitation states which later transform to the ground state by γ -emission. Each γ -energy observed is equal to the energy difference between the disintegration energies with two of the α -particle groups. If for one nuclide all the α - and γ -energies are known, an energy level diagram of the product nucleus can be drawn (Figure 5-1).

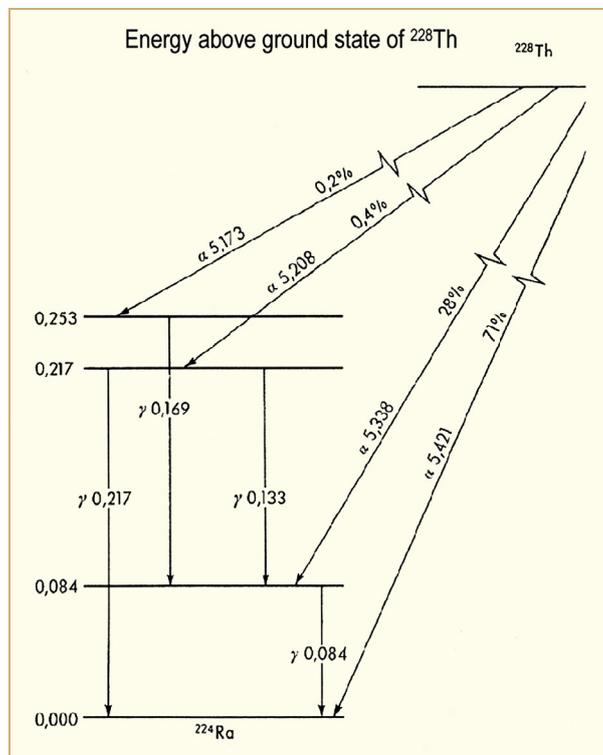


Figure 5-1: Energy-level diagram for the α -particle decay of ${}^{228}\text{Th}$ to ${}^{224}\text{Ra}$. The α -particle energies (MeV) given are the kinetic energies, not total disintegration energies [VGB documents].

During an α - decay process, the α -particle or the ^4He -nucleus leaves the mother nucleus with high kinetic energy, which is nearly equivalent to the mass defect transformed by the disintegration. The total disintegration energy associated with an α -particle emission is larger than the α -particle energy by the recoil energy of the nucleus, which is in the range of 0.1 MeV.

The α -particle in the nucleus is attracted by the powerful but short-range interaction known as the strong nuclear force. In addition, a long-range electrostatic force acts between the positively charged α -particle and the remainder of the positively charged nucleus, and has the effect of repelling the α -particle from the nucleus. The resulting electrostatic force is called the “Coulomb barrier” (Figure 5-2).

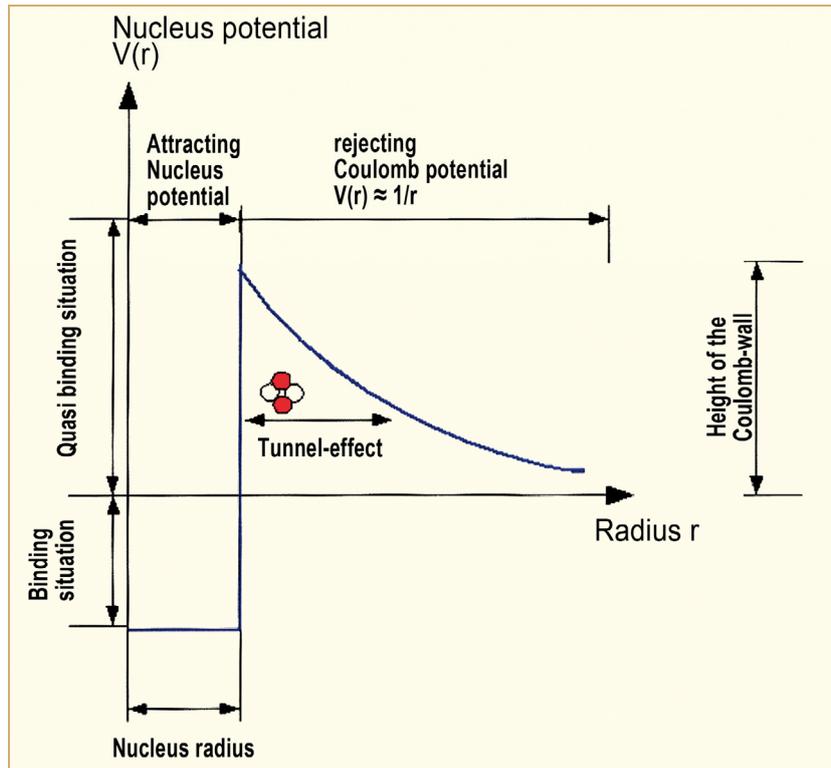


Figure 5-2: The potential energy function for an alpha particle in the vicinity of an atomic nucleus. The total potential is the sum out of the short-range strong nuclear force and the long-range electrostatic force between the positively charged α -particle and the remainder of the positively charged nucleus [VGB documents].

Normally the kinetic energy of an α -particle in a nucleus is too small to overcome this barrier, but in quantum physics there is a possibility to tunnel the barrier. The tunnelling effect can be quantitatively calculated with the Schrödinger equation (Figure 5-3).

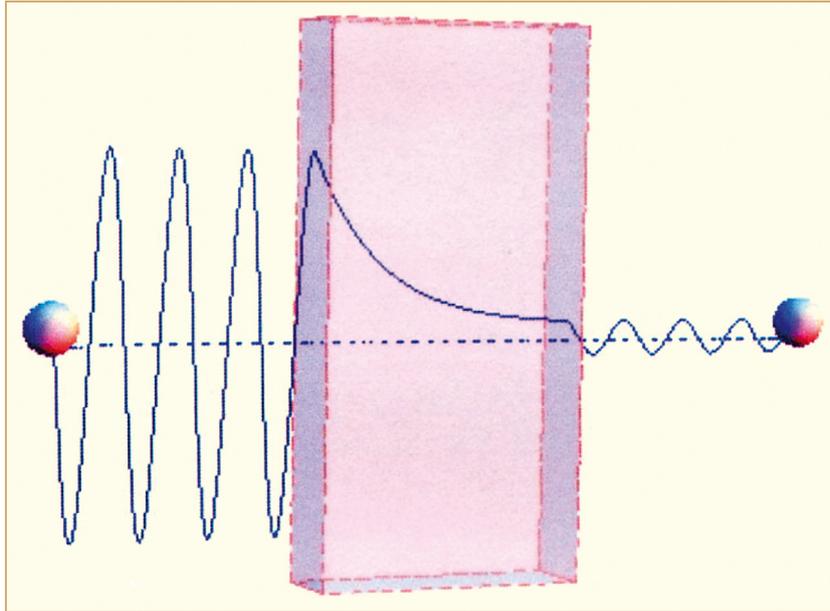


Figure 5-3: Schematic representation of quantum tunnelling through a barrier. The energy of the tunnelled particle is the same, only the quantum amplitude and hence the probability of the process is decreased [VGB documents].

The alpha emission energies of α -decaying nuclei only cover a small range, from about 2 MeV to about 8 MeV. But the half-lives of the corresponding nuclei cover an enormous range, from 10^{-12} s to 10^{17} s. Experiments have shown that, within certain families of alpha-emitting nuclei, the half lives and alpha emission energies are related to one another. This is called the Geiger-Nuttall relation (Figure 5-4).

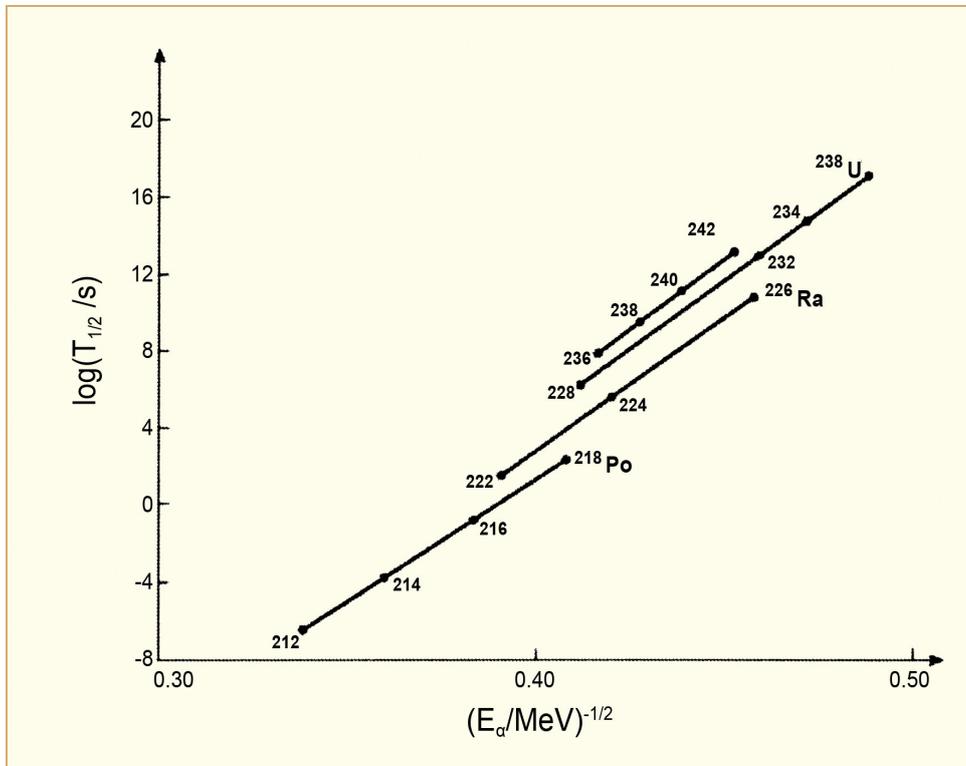


Figure 5-4: A comparison of the Geiger-Nuttall relation with experimental data for different families of nuclei. In this plot, the straight lines confirm the exponential dependence of $T_{1/2}$ on $E_{\alpha}^{-1/2}$ [VGB documents].

6 Beta radiation measurement (Wilfried Rühle)

Beta decay is a radioactive transformation under emission of a beta particle i.e. of a negatively charged electron (β^- -decay) or of a positively charged electron, also called positron (β^+ -decay).

During emission of a negatively charged electron, a neutron in the nucleus is transformed to a proton that causes the proton number Z to increase by 1. During emission of a positively charged electron (positron), a proton in the nucleus is transformed to a neutron that causes the proton number Z to decrease by 1. The mass number M does not change during beta decay.

Antineutrinos or neutrinos that are also emitted during beta decay are of importance as they may carry with them part of the decay energy which causes continuous energy distribution of emitted beta particles. As already said in Section 3.3.1.2, during β^- -decay the atomic number is changed by one unit, and the mass number remains constant. Each β^- -decay, β^+ as well as β^+ , is connected to the emission of neutrinos or anti neutrinos, to ensure that energy and impulse remain constant.

Electron capture is a radioactive transformation by capture and transfer of an electron from one of the inner shells into the nucleus. It is actually β^+ -decay and always occurs when the positron has insufficient energy to leave the nucleus.

Electrons from the K-shell are mainly captured. These radionuclides are also known as K-emitters because a characteristic X-radiation (mainly K-radiation) of the following nuclide is emitted during refill of the atomic shell. The energy range of the emitted X-radiation is in the range of 3–80 keV.

A proton is transformed into a neutron during electron capture in the same way as during β^+ -capture whereby the proton number Z is reduced by 1 while the mass number M remains the same.

Emission of protons or neutrons may occur and is already commented in Section 3.3.1.2.

Emitted radiation by transition from an excited state to the ground state

Some statements regarding this subject can already be found in Section 3.3.1.2.

In case that the nucleus conveys its energy to an electron from the electron shell, predominantly from the K-shell, instead of γ -particles, conversion electrons are emitted. This is accompanied by emission of characteristic x-ray radiation.

If the exciting energy is ≥ 1.02 MeV, there is a possibility, that, to go to the ground state, a positron and an electron are generated at the same time.

6.1 Transformation by β^- -emission

With the β^- -decay, electrons together with antineutrinos are emitted from the nucleus. The atomic number of the daughter nuclide is by one unit higher than that of the mother nuclide, whereas the mass number remains constant:



The decay energy splits into repulsion energy of the emitted nucleus, excitation energy of the emitting nucleus, energy of the β^- -particle and energy of the anti neutrino.

The repulsion energy is very small and can be disregarded. As the total decay energy and the internal excitation energy for a certain nuclide is definite, the sum of the energy of the β^- -particle and the energy of the anti neutrino must also have a definite value. This energy is statistically partitioned between the β^- -particles and the anti neutrinos. As a consequence, each of the two particles can have all energy values between a minimal and a maximal energy. The maximum energy of a β^- -particle or of an anti neutron is equal to the difference of total decay energy and inner excitation energy. For characterization of a β^- -decay, the maximum energy is always used.

The frequency distribution of the β^- -particles in correlation to its energy is called beta-spectrum (Figure 6-1).

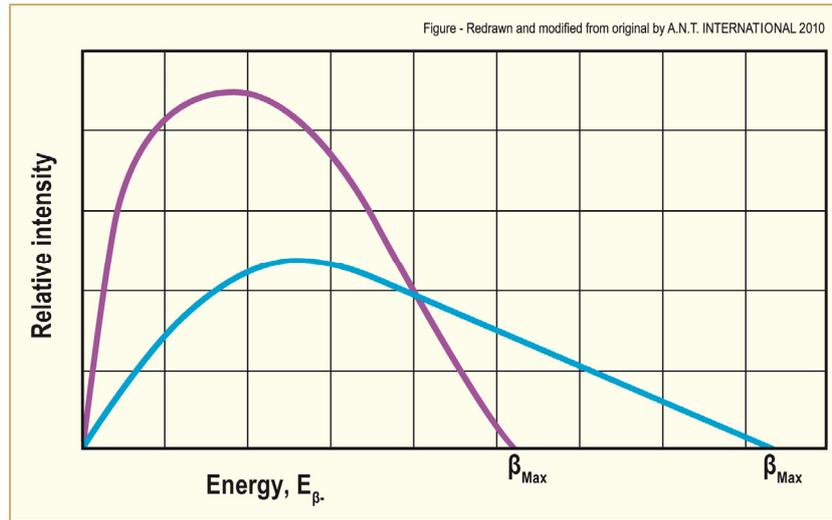
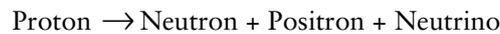


Figure 6-1: Examples for β^- -spectra.

With a few nuclides only, β^- -decay ends at the ground state of the daughter nuclide (pure β^- -emitters). In most cases, the daughter nuclide is generated in an excited status, whereas the β^- -decay is connected to the emission of a γ -particle. Furthermore the daughter nuclide can be generated in different excitation states, whereas several groups of β^- -particles, each with its own energy distribution and maximum energy, are produced. These cases are called “complex β^- -decay” and “complex β^- -spectrum”.

6.2 Transformation by β^+ -emission and EC

At the β^+ -decay, positrons and neutrinos are emitted. The atomic number of the daughter nuclide is smaller by one unit compared to the mother nuclide. The mass number remains unchanged.



The conditions for the kinetic energies are the same as in case of β^- -particles. The shapes of the spectra are similar too, but the positron spectra are a little bit moved to higher energies, as positrons are, in the contrary to negatrons, repelled by the positive loaded nucleus.

The positrons, after having transferred their kinetic energy by interaction with matter, fuse with an electron from the atoms shell, whereby the rest mass of both particles is reflected in two opposed γ -particles with energy of 511 keV each (annihilation radiation). In case of β^+ -decay, γ -radiation with this particle energy always occurs. A small amount of the annihilation radiation consists of particles with higher energy. These are generated in case, that the positrons at the moment of the annihilation have residual kinetic energy.

β^- -decay is only possible, if the total decay energy is bigger than the energy of the annihilation particles, that means, if the energy is bigger than two electron masses or 1.02 MeV. If the energy is smaller, the nuclide can only be changed to its daughter nuclide by capturing a shell electron, combined with the emission of neutrinos.

The daughter nuclides are identical; regardless they have been originated by electron capturing or by positron decay. That means that in case of electron capturing and in case of β^+ -decay, a proton is changed into a neutron:



Electron capturing is also a competitively decay process, if decay by positron emission is possible. After having captured an electron, the generated gap in the shell is refilled by an electron out of a higher shell. Thereby the characteristic x-ray spectrum for the daughter nuclide is generated. According to the shell, where the captured electron comes from, the process is called K-, L-, or M-capturing. The probability for electron capturing decreases with the distance of the electron orbit from the nucleus. That is why predominantly electrons out of the K-shell are captured. The probability for capturing an electron from the L-shell is lower by a factor of seven.

The lines in an x-ray spectrum, especially the K-line, are an indicator for the electron capturing process. If at the electron capturing process, an excited daughter nucleus is generated, additionally γ -radiation is emitted.

6.3 Absorption of β -radiation

The absorption of β -radiation occurs by energy transfer of the β -particles during interaction with matter. The following interaction processes are relevant:

- Excitation or ionisation of the absorber atoms.
- Inelastic scattering at the nucleus with emission of Bremsstrahlung.

In case of absorbers with low mass numbers, electrons lose their energy predominantly by excitation or ionisation. For higher mass numbers and higher energy of the β -radiation, the emission of bremsstrahlung increases.

The measurement of absorption curves is a standard method for the characterisation of β -radiation. The thickness of the absorber, which can be penetrated by a β -particle or its maximum range in the specific material, depends on the energy of the β -particle and on the concentration of electrons within the absorber material.

Instead of the range in cm, normally the mass per area unit in g/cm^2 is used, which can be calculated by multiplication of the range (cm) with the density of the absorber (g/cm^3). This value for the maximum range in g/cm^2 is nearly independent from the material used. This rule is absolutely valid in case where the electron transfers its total energy by excitation or ionisation.

It is useful to plot absorption curves in a half logarithmic scale. Figure 6-2 shows an absorption curve of a β -continuum. The first part of the curve follows an exponential law:

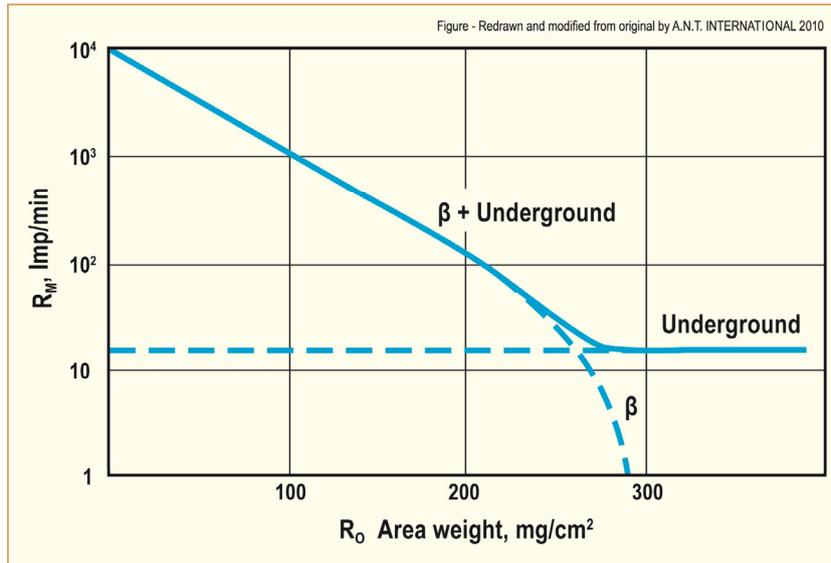


Figure 6-2: Absorption curve of a β -spectrum [VGB documents].

$$R_{N_D} = R_{N_0} * e^{-\mu_m * d}$$

R_{N_D} = net counting rate after passing the absorber thickness d

R_{N_0} = net counting rate with absorber thickness “zero”

μ_m = mass absorption coefficient

D = absorber thickness

In the direction of higher densities, the curve deviates from a straight line, as the mass absorption coefficient μ_m is not constant, but increases with growing area density.

For absorption measurements between the sample, which has to be measured, and the detector, sheets with different thickness (for example Al-sheets) are positioned.

Doing this, care has to be given to the fact, that the support plate and the protective plate for the sample should be as thin as possible to avoid back scattering or to keep absorption and scattering as low as possible. The absorber sheet has to be positioned as close as possible to the detector to avoid inaccurate measurements because of scattering caused effects.

By subtraction of the back ground from the measured absorption curve (caused by back ground rate and Bremsstrahlung), the absolute absorption curve is got. The value on the x-axis, where the y-value has dropped down to the 10^{-4} -fold of the start value, complies with the maximum range. Out of the maximum range got by this method, β -energy can be determined using energy-range-diagrams (Figure 6-3), which are available in literature.

7 Gamma radiation measurement (Rolf Riess)

7.1 Background information

The electromagnetic radiation of short and shortest wavelength located adjacent to the UV radiation is called Gamma Radiation. Its energy E is given by the following equation:

$$E = h \cdot \nu \quad h = \text{Planks Electromagnetic Quantum}; \quad \nu = \text{Frequency}$$

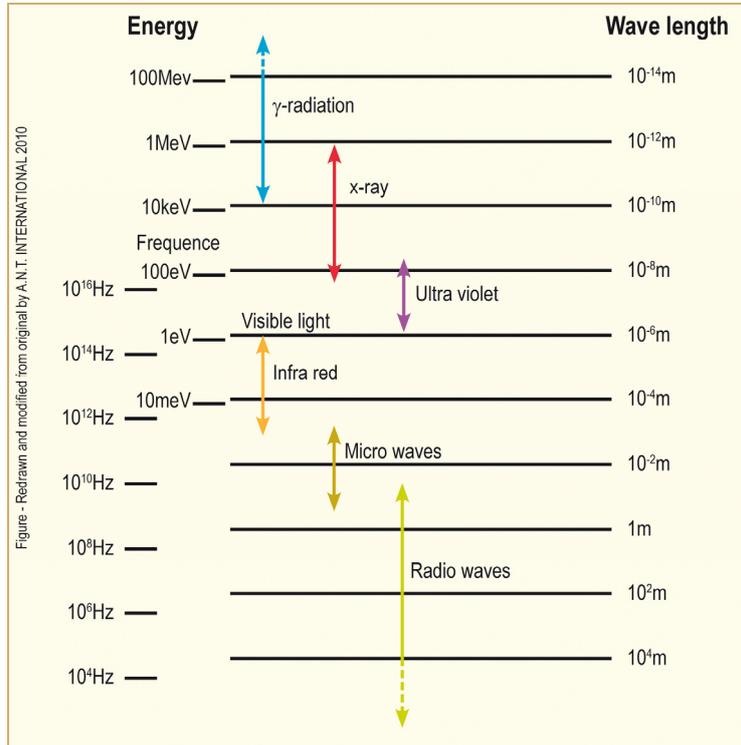
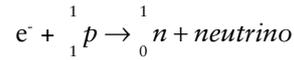


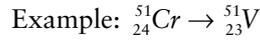
Figure 7-1: The electromagnetic spectrum.

Gamma radiation can be generated in the following processes:

- Decay of a nucleus.
- Fission of a nucleus.
- Neutron capture.
- Unification of a positron and a negatron (annihilation radiation).
- Non-elastic impact between electron and nucleus (x-ray Bremsstrahlung).
- Transition between electron-shells (characteristic x-ray radiation)
If an atom in an inner electron shell is ionized, its place can be taken by an electron of an outer shell. During this transition a characteristic x-ray transition occurs.
- Electron capture (characteristic x-ray radiation)
Nuclei with an excess of protons are able to capture electrons of the innermost shell (K-). This process is called electron capture or K-capture. An outer electron takes this free space by emitting a characteristic x-ray which is used to prove this effect. The captured electron combines with the proton whereby a neutron and a neutrino are generated.



During the electron capture the atomic number is reduced by one unit but the mass number is constant.



The gamma rays generated in the processes a) to g) have different names due to their origin. However, when passing the matter they are reacting identically. Especially they are absorbed by the Photo effect, the Compton effect, and the Pair Production. The dominating processes among the 3 onesis depends on the atomic number of the absorbers and the energy of the gamma radiation (see Figure 7-2). Gamma radiation with sufficiently high energy may also produce nuclear reactions.

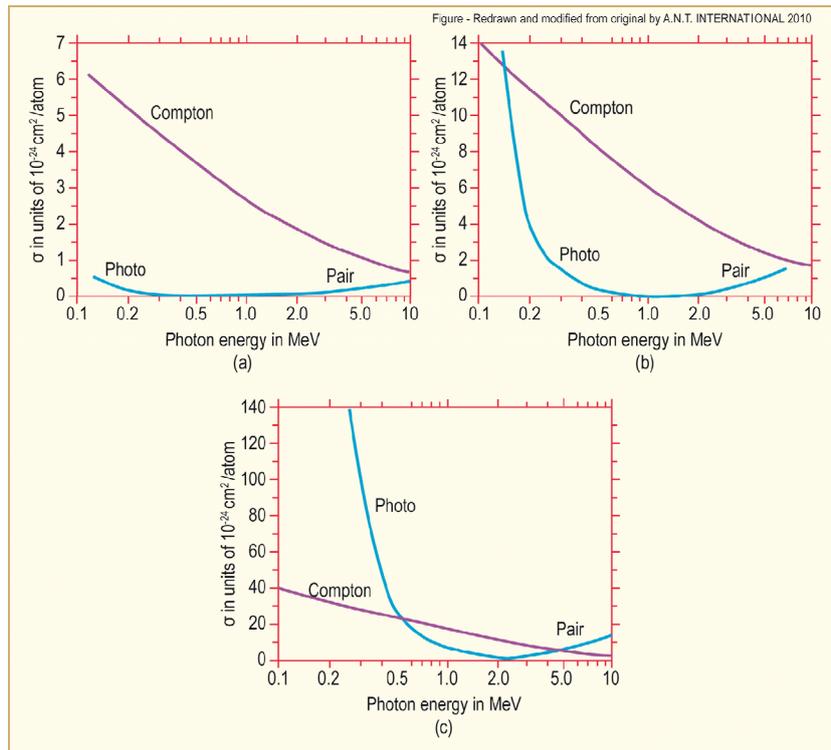


Figure 7-2: Energy dependence of the atomic cross sections for photo electric effect, Compton scattering, and pair production in a) Aluminum; b) Copper and c) Lead.

A gamma quantum can deposit its energy through an ionization of an atom. The secondary electron which is produced during this process is used for the identification of the gamma quantum. This gamma quantum generates energy of about 1 MeV and about 30000 ion-pairs along its track. Compared to this high number, the gamma quantum generated primarily can be neglected.

Remark: The further evaluation deals only with gamma radiation that occurs during decay of a nucleus and the effects which are competing (conversion electron, pairing) and the interaction of gamma radiation with matter and the measuring of gamma radiation.

After a radioactive decay, the new nucleus may be in an energetically enhanced state. Such state of higher energy is changed by the emission of energy. The nucleus will then be in a lower state or in the ground state. The ground state is the energetically lowest level of a nuclide. Should the nuclide be stable, there can be no radioactive energy release. If it is, however, a radionuclide, it will be changed by an alpha-decay, beta decay or spontaneous fission.

In most cases, the excess energy of the excited nucleus will be released spontaneously ($<10^{-14}$ sec). In these cases the emitted gamma quanta (respectively the conversion electrons) are associated with the decaying radionuclide and not the excited consecutive nuclide. If, however, the half life of an excited state will be long, it is called a metastable state and the radiation is attributed to the consecutive nuclide. In order to separate between the ground state and the long-term excited state, the metastable state is indicated by a small m. It can also be the case that a metastable state will not change into the ground status but it will be changed by beta decay. For this reason, as an example, the gamma radiation of ^{110m}Ag is coming from the excited status of ^{110}Cd .

As already mentioned, the gamma radiation occurs during the transition of an excited state with high energy in a state of low energy as for example in Figure 7-3. The energy of the emitted photons is consequently equal to the energy difference of the participating nuclides. Therefore, the gamma emitting nuclides own a spectrum of discrete energy.

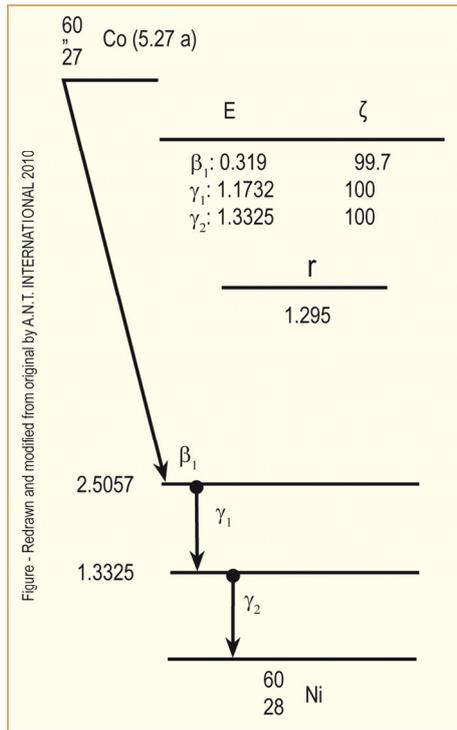


Figure 7-3: Decay scheme for the decay of cobalt-60 (energies in MeV).

In some cases, the gamma quanta are replaced by electrons called “conversion electrons”. The nucleus transfers in this case its exciting energy not as gamma quanta but it transfers its energy by a direct exchange to an electron in a shell, which is preferentially a K shell (inner conversion). As a symbol for the conversion electron the abbreviation e^- is used. After the release of a conversion electron, a free space in the electron shell remains – as with the electron capture – which is compensated by electrons with a higher energy status. Thereby, the characteristic x-ray (K-radiation) can be observed in Figure 7-4.

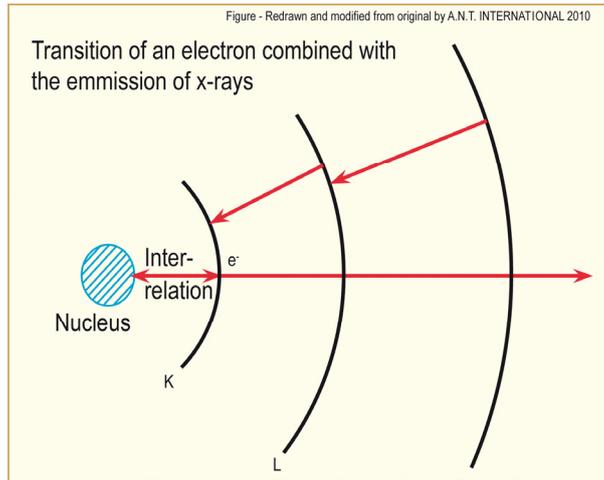


Figure 7-4: Inner conversion.

The conversion electron obtains a line spectrum as well. The energy of a line cannot be the energy difference of the status of the excited nucleus, because the electron has to invest bonding energy in order to leave the atom. The lines of the conversion electrons are therefore reduced by their bonding energy which is below the gamma line.

In many cases, the conversion is in competition with the gamma emission. Whereas one excited nucleus transfers a certain energy either by conversion or by the gamma emission, there is a great number of transition where a reproducible relation of conversion electrons to gamma quanta is given. This is called α/γ relation.

Pairing

If the decay energy is bigger than the necessary energy for pairing (1.02 MeV) this energy can also be released as a negatron/positron pair.

7.2 Interaction of gamma radiation with matter

The transition of gamma rays through matter generates the following processes as shown in Figure 7-5.

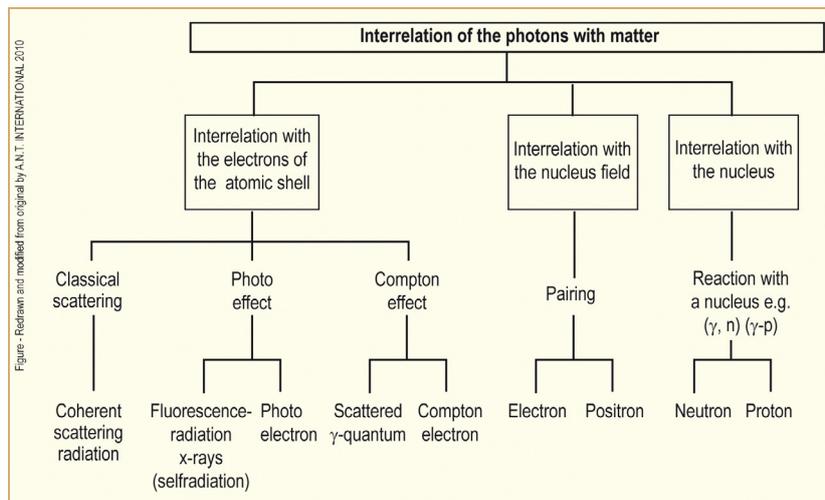


Figure 7-5: Interaction gamma radiation/matter [VGB documents].

7.4.5 Determination of detection and recognition limit

According to the KTA Guidelines 1503, 1503.1, and 1504 a difference has to be made between detection and recognition limit.

The recognition limit relates only to measuring results. It helps in the decision if there is a contribution of the examined media or if only the background was measured.

The detection limit helps to find out whether the measuring equipment can meet the measuring task.

In the interim the regulators try to fix the definitions of both terms in extended guidelines.

7.5 Measurement and corrections of gamma emitters

7.5.1 Measurement of a sample

The procedure for measuring the gamma ray emission rate can be described as follows:

- 1) Place the sample to be measured at the source-to-detector distance for efficiency calibration.
- 2) Accumulate the gamma ray spectrum, recording the count duration.
- 3) Determine the energy of the gamma rays present by use of the energy calibration curve.
- 4) Obtain the net count rate in each full energy gamma ray peak of interest as described in efficiency calibration.
- 5) Determine the full energy peak efficiency, for each energy of interest, from the curve or function obtained in efficiency calibration.
- 6) Apply any sample geometric correction factors, if the sample is not in the same geometry as the calibrating standard.
- 7) Calculate the number of gamma rays emitted per unit time for each full energy peak as follows:

$$N_{\gamma} = C_{\gamma} / E_{\gamma}$$

When calculating a nuclear disintegration from a gamma ray emission rate, determined for a specific radionuclide, a knowledge of the gamma ray intensity per decay is required. That is:

$$A = N_{\gamma} / P_{\gamma}$$

7.5.2 Corrections of counting data

Decay Corrections

1) Simple Decay Correction

The measured activities are corrected for the decay between the times of sampling and counting by using:

$$A_0 = Ae^{\lambda t}$$

Where:

A = measured activity

A₀ = activity at t = 0

T = time between counting and t = 0

λ = decay constant

Generally, the midpoint of the sampling and counting is used for decay correction if the sampling or counting duration is less than 1/3 of the decay half-life. When the sampling or counting duration is longer than 1/3 of the decay half-life, correction for the decay during counting or sampling should be applied (see below), and the end of sampling and the beginning of counting are used in decay correction.

2) Correction for Decay during Counting and Sampling

If the value of a net count rate is determined by a measurement that stands a significant fraction of a half life and the value is assigned to the beginning of the counting period, the multiplicative correction factor, F, must be applied.

$$F = \lambda t / (1 - e^{-\lambda t})$$

$$A_c = AF = A \cdot \lambda t / (1 - e^{-\lambda t})$$

Where:

F = correction factor

A = activity or average counting rate during the counting period

A_c = activity at beginning of count (T = 0)

T = duration of counting

λ = decay constant

The same correction factor is also used in correction for decay during sampling. In this case

$$A_T = A_m = \lambda T / (1 - e^{-\lambda T})$$

Where:

A_T = total activity collected in the sample

A_m = activity measured at the end of sampling

T = duration of sampling

λ = decay constant

Coincidence photon summing correction

When another gamma ray or x-ray is emitted in cascade with the gamma ray being measured, in many cases a multiplicative coincidence summing correction C must be applied to the net full energy peak count rate if the sample-to-detector distance is ≤ 10 cm. Coincidence summing correction factors for the primary gamma rays of Co-60 and Y-88 are approximately 1.09, 1.04, and 1.01 for a 65 cm³ detector at 1 cm, 4 cm, and 10 cm sample-to-detector distances, respectively. The data for cascade-summing corrections for some major nuclides can be found in the literature [Shima & Hoppes, 1983].

Similarly, when a weak gamma ray occurs in a decay scheme as an alternate decay mode to two strong cascade gamma rays, with energy that total to that of the weak gamma ray, a negative correction would be applied to the weak gamma ray. However, the correction factors must be negligible for most of the radionuclides observed in NPP.

Sample geometry calibration

The gamma ray full energy peak efficiency is sensitive to the following counting geometry factors:

- 1) Source to detector distance,
- 2) physical form of the source (gas, liquid, solid); and
- 3) size and shape of the source or source container (point source, filter paper of various sizes, charcoal cartridge, liquid bottles of various sizes, gas vials of various sizes, Marinelli beakers of various sizes).

For most accurate results the source to be measured must be duplicate, as closely as possible, the calibration standard in all aspects. If this is not practical, appropriate correction must be determined and applied. The methods of determining geometric correction factors and the preparation of standard source for various geometric calibrations are described in a separate chapter.

7.5.3 Resolution of a composite decay curve

For a mixture of several independent activities, the result of plotting $\log A$ vs. ϵ is always a curve concave upwards (convex towards the original). This curvature results of the shorter life component become relatively less significant as time passes. In fact, after sufficient time, the longest life activity will entirely predominate, and its half life may be read from this late portion of the decay curve. Now, if this last portion, which is a straight line, is extrapolated back to $t = 0$ and the extrapolated line subtracted from the original curve, the residual curve represent the decay of all components, except the longest lived. This curve may be treated again in the same way, and in principle any complex decay curve may be analysed into its components. In actual practice, experimental uncertainties in the observed data may be expected to make it difficult to handle systems of more than 3 components, and even two component curves may not be satisfactorily resolved if the two half lives differ by less than about a factor of 2.

An example of reactor water sample containing N-13, F-18, and Cu-64 is described in Section 7.5.7.

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