Detecting and measuring ionizing radiation – a short history

by F.N. Flakus*

Ionizing radiation causes neutral atoms or molecules to acquire either a positive or negative electrical charge. The most commonly known types of ionizing radiation are alpha, beta, gamma, X, and neutron rays. Charged-particle radiation, such as alpha or beta rays, has a direct ionizing effect; whereas neutral radiation, such as X, gamma, or neutron rays, have an indirect ionizing effect, i.e. these radiations first generate charged particles which then have the ionizing effect.

Radiation is a form of energy. This energy can be partly or wholly deposited in a suitable medium and thus produce an effect. The detection and measurement of radiation is based upon the detection and measurement of its effects in a medium, and the history of the emergence of radiation detectors is closely related to the discovery of radiation and radiation effects.

Radioactive material emits ionizing radiation without having been subject to any external influence. The type of radiation emitted and its associated energy is characteristic for the kind of radioactive substance. In many applications of radiation detectors, the object is to measure the energy distribution of the radiation (spectrometry).

Although ionizing radiation has been present in nature (cosmic rays, naturally occurring radioactive materials) throughout man's history, it remained unnoticed until less than 100 years ago. Man has no specific sense which could have responded to this kind of radiation and it had not been possible for scientific initiative to develop any instruments that would have amplified a human response to that radiation, as was the case for example in the area of visible light waves (optics)

As we know today, tools for detecting ionizing radiation were, in principle, in existence rather early: the phenomenon of thermoluminescence was first described in the 17th century; the gold leaf electroscope was invented in the 18th century; and photography was developed during the early 19th century. However many years passed until the doors to these completely new fields of science were opened by W.C. Rontgen in 1895, and by H. Becquerel in 1896.

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Emergence of radiation detectors from the early discoveries

On the evening of 8 November 1895, while working in a carefully darkened room, Rontgen noticed that a piece of a cardboard coated with barium platinocyanide showed a faint, flickering, greenish light (fluorescence) when electrical discharges took place in a Hittorf-Crookes tube near the screen. The tube itself was carefully covered with a black shield impervious to any visible light. Rontgen verified that the tube was the source of a new kind of radiation, which was invisible but which revealed its existence when hitting the luminescent screen. Rontgen subsequently performed many careful experiments on the radiation which he named X-rays. His first important step was to replace the fluorescent screen by a photographic plate· this was susceptible to X-rays and thus provided a tool for recording X-ray pictures.

On 22 December 1895, Rontgen made the first medical X-ray photograph. It shows the hand of his wife and marks the birth of radiography. Subsequently, mainly photographic plates were used in X-ray studies. The fluorescent screen lost importance, although fluoroscopes or sciascopes were improved by W.F. Magie, E.P. Thomson, and T.A. Edison. During his many experiments on X-rays, Röntgen also observed that air would conduct electricity when traversed by the rays. This effect was later-used as the operating principle for several kinds of radiation detector. For his discovery, Rontgen was awarded the first Nobel Prize in Physics in 1901.

It is interesting that the valuable possibilities of the use of X-rays for many purposes, medical and non-medical, became immediately apparent and that during the following year, 1896, over 1000 articles and more than 50 books were published on the subject. Knowledge of the new findings spread along with ignorance and scattered opposition· for instance, it was proposed "to prohibit the use of X-rays in opera glasses at theatres" or "to burn all work on the X-rays and to execute all the discoverers". One company "made a prey of ignorant women by advertising the sale of X-ray-proof underclothing".

In early work with X-rays it had been quickly recognized that the spot in the discharge tube where the cathode rays caused the emission of X-rays, was characterized by a strong fluorescence and a correlation

between this fluorescence and the emission of X-rays was suspected. Following suggestions made by H. Poincaré, Becquerel in 1896 undertook a systematic study of the connection between the emission of visible light and of X-rays. He wrapped phosphorescent substances, after exposure to sunlight, in black paper and placed them on photographic plates to detect any emission of X-rays. It happened that Becquerel started his experiments with highly fluorescent uranium compounds. He demonstrated that the position of the uranium minerals was revealed by black spots occurring on the developed photographic plate and that penetrating rays like X-rays were emitted. When he later developed a photographic plate that had lain for several days in a drawer under a tray with uranium salts which had *not* been exposed to sunlight, he found the same dark spots on the plate. He repeated the experiment and concluded that neither sunlight, phosphorescence nor fluorescence was necessary to produce the effect, and that invisible rays were emitted from all uranium compounds and from metallic uranium. This marks the discovery of radioactivity in 1896.

Becquerel continued carefully to investigate the properties of the rays. Among other important facts, he found that the rays discharged a charged electroscope (invented in 1787 by A. Bennett). After Becquerel's discovery, scientists started the search for further elements with radiation-emitting properties.

As already mentioned, one property of X-rays discovered very early was their ability to make dry air electrically conductive, i.e to liberate electrical charges in air. These charges could be collected by producing an electric field across that volume of air. However, it was not until the work of J.J. Thomson in 1899 that the operating characteristics of these ionization chambers were well understood. Madame Sklodowska Curie realized that the ionization method of measuring the intensity of X-rays would be well suited to detecting the radiation found by Becquerel. She equipped an electrometer used by her husband Pierre Curie with a suitable ionization chamber and repeated Becquerel's experiments. The method proved very satisfactory. She found that the intensity of the radiation was proportional to the amount of uranium. In July 1898, together with her husband, she discovered the radioactive element polonium. In December 1898 they announced the discovery of radium. Together with Becquerel, they were awarded the Nobel Prize for Physics in 1903. It was also Madame Curie who proposed calling the new phenomenon *radioactivity.*

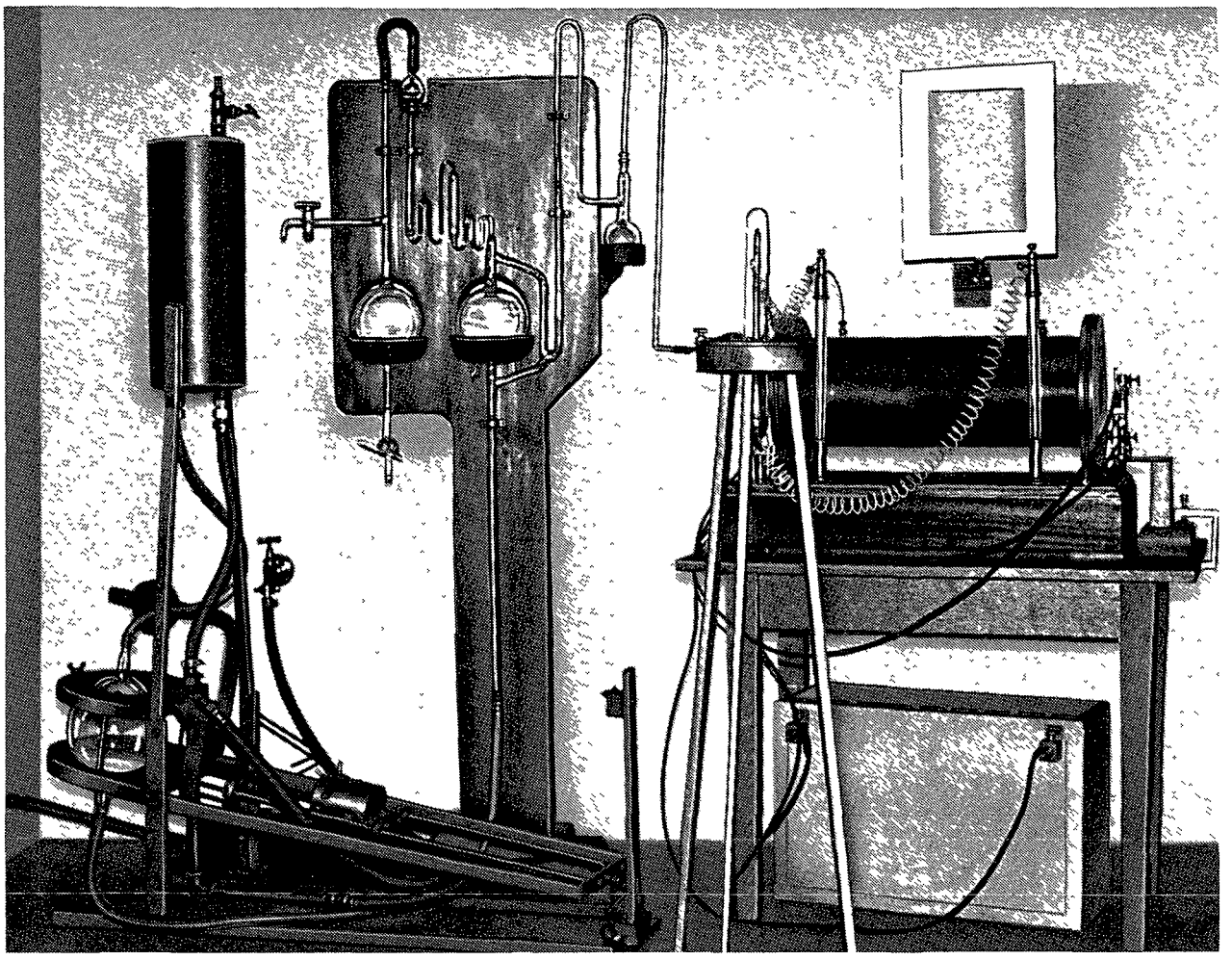
It is interesting to note that in 1899, J. Elster and H. Gertel, and independently C.T.R. Wilson, had found that an electroscope steadily lost charge without apparently being exposed to radiation, and suggested that this resulted from the presence of radioactivity in the earth. The deflection of part of the "Becquerel rays" in a magnetic field was also noticed in 1899. A few years later Rutherford succeeded in giving a complete explanation of the nature of alpha, beta, and gamma rays.

In the beginning of radium research work, the photographic plate was used to study the new phenomena. However, ease of handling, relatively low cost and high sensitivity soon made the gold-leaf electroscope the preferred measuring instrument for work with radioactive material. Various types of electroscopes were developed for this purpose: the alpha electroscope, where the ionization chamber and the measuring chamber were closely connected; the beta electroscope, where ionization chamber and measuring chamber were combined, and the emanation electroscope for measuring radioactive gases. The use of ionization chambers in conjuction with galvanometers remained restricted to the measurement of intense radiation. The very early detection methods, namely the fluorescent screen, the photographic plate, the gold-leaf electroscope, and the ionization chamber in combination with an electroscope or galvanometer, had one common characteristic: they could only be used to respond to "bulk" radiation but they were insensitive to single rays.

The first instrument that was able to detect individual rays was the spinthariscope, invented by Crookes in 1903. It had been discovered that the luminosity of a zinc sulphide screen, when exposed to alpha particles, consisted of a number of individual scintillating points of light that could be observed with a strong magnifying glass. Rutherford and Geiger found that, on a uniform screen, each scintillation corresponded to the impact of one alpha particle only. Thus a very simple method for counting individual alpha particles was available which eventually gained great practical importance. The method was refined and used most profitably by Rutherford and his students and was extensively used for many years until the 1930s. However, it imposed considerable strain and fatigue on the observers: the usual procedure was for an observer to count for one minute, then be relieved by another observer. Each observer might have to count for 20 periods of one minute each during one experiment.

Another instrument that made single radiation events visible was the cloud chamber, first built in 1911 by C.T.R. Wilson. A cloud chamber usually consists of a flat glass front-plate for observation, glass side-walls for illumination, backed by a movable piston in order to expand the volume of the chamber in a small fraction of a second. Droplets form along the path of electrically charged particles and these visible tracks can be photographed.

Between 1923 and 1930 the cloud chamber was developed into a most useful research tool which has a very large number of important discoveries to its credit. In 1939, A. Langsdorf built the first diffusion cloud chamber, which is continuously sensitive. Since the 1950s bubble chambers, which make particle trails visible in liquid medium, have been in wide use in high-energy physics experiments. In the 1960s streamer chambers were developed.



Reconstruction of the experimental set-up by which Röntgen discovered X-rays on 8 November 1895. The discharge tube is fixed to the tripod, the fluorescent screen is located in the right upper corner. (Photo: Deutsches Rontgen-Museum.)

Besides photographic, ionization, and scintillation effects, other effects of radiation were noticed very early. The fact that ionizing radiation has biological effects was known from the earliest days of radiation work. In April 1896 Daniel described a serious skin reaction which occurred after prolonged exposure to X-rays. Similar observations were also made by several other X-ray investigators. One X-ray burn is known to have occurred even before X-rays were discovered! In summer 1895 while working with cathode discharge tubes, E. Grubbé developed a curious dermatitis of the hands which neither he nor his physician understood. He died of cancer in 1960. Becquerel, in 1901, found a reddening of the skin under his waistcoat, where he had pocketed a radiation source. P. Curie intentionally exposed part of his arm to a radiation source for 10 hours which resulted in a sunburn-like skin reaction and wound and which took four months to heal. Danlos, of the Hospital Saint-Louis, investigated whether the effects of radium on the skin could be utilized for the treatment of certain skin diseases. However many of the pioneers in radiation research became martyrs to their work. A memorial to these martyrs was erected in Hamburg in 1936, listing 178 names. In 1928, at the Second International Congress of Radiology, the International Commission on Radiological Protection (ICRP) was set up, originally under the name of the International X-ray and Radium Protection Committee This body has since taken the lead in establishing radiation safety principles.

Another physiological effect that was observed soon after the discovery of radium was the rapid depression of leucocytes in the blood after injection of radioactive liquids. Also it became known that blind people had a sensation of light when their eyes were exposed to a radium source, the reason for this being a fluorescence of the eye's lens, body, and retina under gamma-ray exposure.

Further effects of radiation were the colouring of glasses or minerals as well as the bleaching of colours and a slightly elevated temperature of strong radium samples. These effects played only a minor role in detection and measuring in the early days of radiation work. Later however, chemical effects were widely used for quick dosimetry in strong radiation fields, and the thermal effect gained particular importance for absolute radiation measurements of alpha emitters with microcalorimeters.

Evolution of radiation detectors

The effects discovered in the early research work with X-rays and radioactive materials remain the underlying principles of radiation detection into modern times. Detection instruments have been improved many times over the years and numerous investigators have contributed to a tremendous development, of which only the main lines can be sketched here.

Photographic emulsions

As already outlined, using photographic plates to record ionizing radiation dates back to the discovery of X-rays and radioactivity. In later years special photographic emulsions were developed, particularly for use in X-ray radiography and autoradiography. The early applications made use of the darkening of the emulsion under irradiation. But in the 1930s, other special emulsions, nuclear emulsions, were developed which allowed individual nuclear tracks to be observed. These were extensively used in research work on cosmic rays.

Photographic dosimetry was made quantitative only in 1942, when radiation film badges for routine personnel monitoring were designed. Before then, some use had been made of film to estimate the exposure of personnel to radiation, but the results were vague at best: dental-film packages, carried for some time in the pocket, were developed and a density that made newspaper print illegible was taken as an indication that protective measures should be investigated. Photographic emulsions were also used as an aid in work with cloud chambers and particularly in magnetic spectrographic systems for particle spectrometry. Today, photographic emulsions are widely used in medicine, industry, and research.

Scintillation

Detecting ionizing radiation by the scintillating light induced in zinc sulphide was one of the first radiation detection and counting techniques. It was almost completely superseded by electrical counting methods in the 1930s. But the method came to life again in the 1940s. electronic photomultiplier tubes were introduced and matched with scintillators to convert the weak light flashes into usable electric pulses that could be counted electronically. Counting by eye became obsolete.

To be a good scintillator, a material should convert as large a fraction as possible of the incident radiation energy into prompt fluorescence, the light-yield should be proportional to the deposited energy, the luminescence should be of short duration, the material should be homogeneous and of good optical quality. No material simultaneously meets all these criteria, and the choice of a particular scintillator is always a compromise among these and other factors.

Several good scintillating media were found in the years 1947 and 1948. Organic scintillators were introduced in 1947. In 1948 the thallium-activated sodium iodide crystal was discovered which became commercially available in 1950, and was widely used thereafter. This was virtually the first pratical solid detection medium for gamma-ray spectrometry, and remains the most popular scintillation detector for this application today. Liquid scintillation detectors were reported in 1948, but interest in these detectors did not develop until the 1960s. Although the NaI(Tl) crystal is technically a solid detector, it has become the practice to characterize only devices that are based on semiconductor media as solid-state detectors.

Ionization

Various materials can be ionized, but to be of use in a detection instrument, charges generated by radiation must move under the influence of an electric field. Gases fulfill this condition easily, and the first generation of ionization detectors were gas-filled. The simplest type is the ionization chamber which was already an essential tool in the early studies of X-rays. By using an ionization chamber during a balloon flight, Hess in 1910 provided the first evidence for cosmic rays.

In 1908 Rutherford and Geiger described the first cylindrical electrical counter for alpha particles, which they improved in 1912 by introducing a spherical counter. Further progress was achieved in 1913 when a detector for counting beta particles was developed. In 1928 Geiger and Muller introduced a new type of gas-filled counter that responded to individual radiation-induced events by giving a high-level output signal. This was the Geiger-Muller counter or GM counter. The counter was further developed in the 1930s and, because of its simplicity, ease of operation, and cheapness, rapidly found wide use. The GM counter however could not directly measure the energy of radiation and was limited to relatively low counting rates. However it is still often the best choice when a simple and economic counting system is needed.

The development of gas-filled detectors reached further intensity in the 1940s. In 1940, Frisch invented the gridded ionization chamber, which still finds limited application in alpha spectrometry. In the late 1940s a third type of gas-filled detector, the proportional counter, was introduced which amplified the charge originating in the gas. An important application of proportional counters was the spectrometry of low-energy X-rays. Today proportional counters are still widely used in many laboratories for alpha or beta radiation measurements. Although gas-filled detectors are still in routine use in nuclear laboratories, the classical gas-ionization detectors have disappeared from almost all high-precision work.

It was soon realized that the use of a solid detection medium instead of a gas would be of great advantage in radiation detection applications, as solids are about 1000 times denser than gases and the dimensions of solid detectors could be kept much smaller than those of equivalent gas-filled detectors. Some work on crystal

counters was reported as early as 1932 by Jaffe, and in 1945 by Van Heerden. However, radiation detectors which measure the ionization produced in solid dielectrics, and which are the solid-state analogue of gaseous chambers, emerged only in the late 1950s and early 1960s. These semiconductor detectors were then rapidly developed. Diffused-junction and surface-barrier detectors found widespread application in the detection of alpha particles. The process of ion-drifting, first demonstrated by Pell in 1960, provided the practical method through which semiconductor detectors could be fabricated with larger active volumes. These "drifted detectors" quickly spread throughout nuclear science.

Today, silicon as semiconductor material predominates in diode-detectors used for charged-particle spectrometry, while germanium is more widely used in ion-drifted detectors for gamma-ray measurements. Silicon detectors have become the most common choice for low-energy X-ray spectroscopy systems and also for the spectroscopy of beta particles. The main characteristic of germanium detectors is their excellent energy resolution in gamma-ray spectroscopy, and here these detectors have now almost supplanted all other systems. The production of Ge(Li)-detectors requires great skill. However, when these detectors became commercially available, they spread around the world like wildfire.

A practical limitation of Ge(Li) detectors is that they have to be stored and operated at very low temperatures. During the period 1970–1972 ultrapure germanium became available which enabled the production of germanium detectors without lithium compensation, the so-called intrinsic germanium detectors. These can be stored at room temperature. It is estimated that about 1000 intrinsic germanium detectors are in use today all over the world.

Chadwick discovered the neutron in 1932. Modest neutron sources soon became available, but neutron detection and measurement posed much greater difficulties than for the other types of radiation. Early work focused almost entirely on secondary ionization detectors, where the charged products of neutron-induced reactions, or the charged recoils, were detected in appropriate ionization or proportional counters. Only several years later, after 1942, when very strong neutron sources in the form of fission reactors became available, were more sophisticated neutron detectors developed.

Virtually every type of neutron detector involves the combination of a target material, designed to convert the neutrons into energetic charged particles, together with a conventional radiation detector. As neutron physics covers a very broad energy span of about 14 decades (10^{-6} to 10^{8} eV), rather different techniques have been developed for neutron detection in different energy regions, generally distinguished as slow, intermediate, and fast neutrons.

The most popular reactions for the conversion of slow neutrons into detectable charged particles are the (n, α)

X-ray photograph taken by Röntgen on 22 December 1895, showing the hand of his wife.
(Photo: Deutsches Rontgen-Museum.)

reactions with boron-10 and lithium-6, and the (n, p) reaction with helium-3. A widely used detector for slow neutrons is the proportional tube filled with boron trifluoride gas.

Several fast-neutron detectors were based on the principle of placing a slow-neutron detector in the centre of a moderating medium. A popular detector, a long counter which consists of a BF_3 tube surrounded by a cylindrical moderator and which delivers a "flat response" curve, was first suggested by Hanson and McKibben in 1947. The discovery of organic scintillators for neutron detection around 1950 expanded the field of fast neutron physics. The development of inorganic scintillators (Li-6 glass scintillators) was important for the advances in low-energy neutron physics since the early 1960s. The greatest precision in slow-neutron detection, however, was attained with so-called "time of flight" systems.

In 1960, Bramblett, Ewing, and Bonner first developed a "spherical dosimeter" whose neutron detection probability well matched the dose delivered by neutrons in a biological medium, and which today carries the name "Bonner sphere". Development of small detectors for neutron exposure control, particularly for the intermediate neutron-energy region, still requires particular attention.

Many other detectors which have not been mentioned, play an important role in radiation detection today: in

certain materials, notably plastic foils, the trail of damaged molecules along a particle track can be made visible by enlarging the damage through etching with chemical agents Materials used to detect charged particles in this way are termed *track-etch detectors.* Special read-out methods for counting such tracks are available and the track detection method has found useful application for the detection of heavy ions and fission products

The radioactivity induced in a great number of materials by neutron interaction can serve for neutron measurements. Measurement of radiation emitted from these materials can be used to deduce information about the incident neutrons. Such detectors are termed *activation detectors.* Complex physical limitations are involved, nevertheless activation detectors have found limited application for neutron radiation measurements.

Inorganic scintillation materials emit light in the form of prompt fluorescence when exposed to radiation. A different class of inorganic crystals however can "trap and store" the excitations caused by irradiation and this excitation can be freed as light emission, under elevated temperature. Materials with such properties function as integrating detectors. Detectors based on this principle are termed *thermoluminescent dosimeters* The phenomenon of thermoluminescence was first described by Boyle in 1663 and later by Wick in 1927–1928 The studies which led to the development of the thermoluminescent dosimeter (TLD) however started only in 1948 by L.F. Heckelsberg. The relationship found between radiation-dose and the amount of thermoluminescence led Daniels to suggest the use of alkali halide crystals for radiation dosimetry. Small pieces of LiF first came into large-scale use as personnel dosimeters in 1966. These little TLD chips are gradually replacing photographic film badges in personnel dosimetry particularly in tropical countries.

A further detector that is based on a different principle and that deserves particular mention is the *Cherenkov detector.* Light emitted by fast charged particles passing through a transparent medium was first observed by Cherenkov in 1937 and named after him. Detectors based on sensing Cherenkov light are widely used in high-energy physics experiments.

Detector instrumentation

The development of radiation detectors could not have taken place without an important development that occurred in parallel: the development of equipment to receive signals from radiation detectors, process them and produce a convenient output. Some landmarks in this evolution may be recalled Williams, in 1931, extending the work of Greinacher in 1926, developed the first linear amplifier capable of amplifying the pulse produced in a shallow ionization chamber by the passage of a single alpha particle. In 1940 the photomultiplier was developed. In the early 1950s, a broad range of amplifiers based on vacuum-tube circuits was on hand. Their performance was steadily improved, for instance in 1956 by the development of feedback circuits.

During the late 1950s and early 1960s, the vacuum-tube circuits were replaced by transistor circuits. the advantages being low energy consumption, small size (integrated circuits), and reliability. This quickly led to the development of many very capable portable radiation detectors. Instruments were extended to cope with higher counting rates. During the 1960s computer technology was applied to radiation detectors. The many advantages that microprocessors (central processing units on integrated circuit chips) will bring to radiation measuring instruments can only be imagined today.

Outlook

Over a period of less than a century, nuclear radiation detectors have steadily extended human senses into entirely new realms, they have virtually provided man with a sixth sense. A great many reliable instruments for detecting and measuring ionizing radiation are now available, ranging from instruments that are widely used as routine tools in nuclear laboratories, to highly sophisticated complex instrument systems designed for very special applications. Detectors have not only investigated the nature of nuclear radiation and radiation emitters, but they have served the beneficial application of radiation in medicine, industry, and research, and also the control of any hazards that might arise from ionizing radiation.

The rapid development of measuring instruments and their wide availability, however, has brought with it new problems: one can see highly sophisticated instruments being used in many places where a simpler instrument would be sufficient, for example multichannel analysers might be used where a single-channel analyser would serve.

Radiation measuring instruments have given man the ability to characterize his environment at levels of precision that even a few years ago would have been considered impossible. For many elements, sensitivity has reached the level of parts per billion – very much smaller concentrations than have ever before been accessible to any measurements.

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