EMANATIONS AND "INDUCED" RADIOACTIVITY: FROM MYSTERY TO (MIS)USE*)

Z. I. KOLAR

Department of Radiochemistry, Interfaculty Reactor Institute,
Delft University of Technology,
Mekelweg 15, 2629 JB DELFT, The Netherlands
E-mail: kolar@iri.tudelft.nl

Radon, Rn; atomic number Z = 85; is a (gaseous) chemical element of which no stable but only radioactive isotopes exist. Three of them, namely actinon (^{219}Rn), thoron (^{220}Rn) and radon (^{222}Rn) are the decay products of naturally occurring radioisotopes of radium: ^{223}Ra, ^{224}Ra and ^{226}Ra, respectively. The natural Rn isotopes were discovered within the period 1899- 1902 and at that time referred to as emanations because they came out (emanated) of sources/materials containing actinium, thorium and radium, respectively. The (somewhat mysterious) emanations appeared to disintegrate into radioactive decay products which by depositing at solid surfaces gave rise to "induced" radioactivity i.e. radioactive substances with various half-lives. Following the discovery of the emanations the volume of the research involving them and their disintegration products grew steeply. The identity of a number of these radioactive products was soon established. Radium- emanation was soon used as a source of RaD (^{210}Pb) to be applied as an "indicator" (radiotracer) for lead in a study on the solubility of lead sulphide and lead chromate. Moreover, radium and its emanation were introduced into the medical practice. Inhaling radon and drinking radon-containing water became an accepted medicinal use (or misuse?) of that gas. Shortly after the turn of the century, the healing (?) action of natural springs (spas) was attributed to their radium emanation i.e. radon. Bathing in radioactive spring water and drinking it became very popular. Even today, bathing in radon- containing water is still a common medical treatment in Jáchymov, Czech Republic.

1 Introduction

In 1896 i.e. three months after the publication by Wilhelm Conrad Röntgen (1845-1923) dealing with his discovery of X-rays [1], Antoine-Henri Becquerel (1852-1908) found that uranium salts emit invisible radiation capable of darkening photographic plates wrapped in black paper [2]. Later he noticed that this radiation could discharge an electroscope directly [3] or even indirectly by ionising a current of air as it passed over the uranium salts. "Moreover, the rate of discharge by the "modified air" was proportional to the amount of uranium over which it had passed" [4].

In 1898 Gerhard Carl Schmidt (1865-1949) [5] and Marie (born Marya Sklodowska) Curie (1867-1934) [6] reported that thorium and its compounds could bring about the same effects, namely blackening a wrapped photographic plate and discharge an electroscope.

*) Based on the authors lecture delivered at Radium Palace in Jáchymov on 23rd April, 1998 during the "Jáchymov" historical session of the 13th Radiochemical Conference held in Mariánské Lázně & Jáchymov, Czech Republic, 19th – 24th April

In the very same year 1898 discoveries were published of two radioactive substances, namely polonium by Pierre Curie (1859-1906) and his wife Marie Curie [7] and subsequently radium by the Curies together with Gustave Bémont (1867-1932) [8]. Both substances i.e. radioelements were isolated from an uranium mineral, the pitchblende originating from St. Joachimstahl or Jáchymov, a town in Bohemia then a part of Austria-Hungary and now of the Czech Republic. "The town, founded by Count Štěpán Šlik in 1516 has repeatedly produced a most striking effect on the global history" [9]. Radium and radioactivity certainly evoked such an effect. More information on Jáchymov mines and their importance for the early history of radioactivity can be found in [10, 11] and [11], respectively.

Henri Becquerel and the Curies were awarded the Nobel Prize in Physics for 1903. Eight years later Marie Sklodowska Curie was awarded a second Nobel Prize, namely the prize in chemistry for 1911. Fig. 1 shows Pierre and Marie Curie in 1904 [12].

Fig. 1. Pierre and Marie Curie in 1904 (nine years after their marriage and two years before Pierre's accidental death in 1906) [12].

2 Discovery of emanations & induced radioactivity

Working at McGill University, in Montreal, Canada on the investigation of the radiation emitted by thorium and its compounds, Ernest Rutherford (1871-1937) and Robert Bowie Owens (1870-1940) "found that all the compounds of thorium examined, viz. thorium oxide, thorium sulphate, and thorium nitrate gave out the same kind of radiation as measured by the transparency of aluminium foil for the rays. The intensity of the radiation differed largely; for equal weights of active substance being greatest for thorium oxide. The nature of the radiation is thus independent of the particular state of chemical combination of the compound, but depends only on the presence of thorium in the material" [13]. However, already very early, they "observed that the radiation from thorium oxide was not constant, but varied in a most capricious

Fig. 2. Ernest Rutherford as a young man [17].

manner. This was the more peculiar, as the sulphate and nitrate were fairly constant. All the compounds of uranium also give out radiation which remains remarkably constant..." [13]. The sensitiveness of thorium oxide to slight currents of air was remarkable. Even a slight draught of air caused by opening or shutting the door of the room affected the radiation intensity measurements i.e. the rate of the discharge of an electrometer [13]. In January 1900 E. Rutherford reports that "thorium compounds continuously emit radio-active particles of some kind, which retain their radio- active powers for several minutes. This "emanation", as it will be termed for shortness, has power of ionising gas in its neighbourhood..." [14]. "The emanation passes through a plug of cotton-wool without any loss of its radio-active powers. It is also unaffected by bubbling through hot or cold water, weak or strong sulphuric acid. In this respect it acts like an ordinary gas". Moreover, E. Rutherford found that "the emanation gradually loses its radio-active power" and "the intensity of the radiation has fallen to one-half its value after an interval of about one minute" [14]. After about 10 minutes the radio-active power of the emanation became too small for measurements with an electrometer. He also derived an equation delineating the decrease of radio-active power viz. decay of the number of ions produced by the emanation:

$$dn/dt = -\lambda n$$

where n = number of ions produced per second by the radioactive particles between the plates of the electrometer, t = time in seconds and λ = constant in 1/second.

If $n = N$ when $t = 0$, it is easily seen that:

$$n/N = exp(-\lambda t)$$

It has been shown by E. Rutherford that $\exp(-\lambda t) = 1/2$ when $t = 60$ seconds [14]. The present value for t, being the half-life of thorium emanation ≡ thoron ≡ ^{220}Rn is 55.6 seconds!

In the last paragraph of the latter publication E. Rutherford reports that "Experiments, which are still in progress, show that the emanation possesses a very remarkable property. I have found that the positive ion produced in a gas by the emanation possesses the power of producing radio- activity in all substances on which it falls. This power of giving forth a radiation lasts for several days. The radiation is of more penetrating character than that given out by thorium or uranium. The emanation from thorium compounds thus has properties which the thorium itself does not possess" [14]. In his next publication E. Rutherford concluded that the presence of the emanation is necessary for the production of radioactivity in substances: a phenomenon for which he proposed the term "excited radioactivity", and that the amount of radioactivity depends upon the amount of the "emanation". "A radio-active substance like uranium, which gives out no emanation, produces no trace of exited radioactivity" [15]. The intensity of radiation emitted by the "excited radioactivity" falls of in a geometrical (exponential) progression with time, decreasing to half its value in about 11 hours. Moreover, it appeared that hydrochloric and sulphuric acids (but neither water nor nitric acid) rapidly remove the radioactivity collected on the surface of a platinum wire previously being negatively charged and immersed into emanation-containing gas [15]. "The solution, when evaporated, leaves the active portion behind".

E. Rutherford went on with his studies on thorium emanation and "excited" or "induced" radioactivity. For two years he did it in collaboration with Frederic Soddy (1877-1956). "In this work the conclusion was drawn that thorium emanation is a specific type of radio-active matter of gaseous character in infinitesimal quantity" [16]. "Various experiments were performed on the action of temperature and chemical reagents upon the emanation. The general result showed that it was completely unaffected by any means employed including acids or different reagents. It was not altered by passing red-hot lead chromate, magnesium powder, zinc dust etc. and in this respect it showed the same chemical inertness as the argon, discovered in 1894, and other members of the argon family of gasses" [16], namely helium, krypton, neon and xenon, all discovered in the period 1895-1898.

E. Rutherford was awarded the Nobel Prize in chemistry for 1908 "As a reward of your research on the disintegration of the elements and the chemistry of radioactive matters". Fig. 2 shows him as a young man [17].

During his early work on emanation and "excited" or "induced" radioactivity E. Rutherford was unable to find any substance but thorium compounds, especially thorium oxide giving rise to these phenomena. The Curies had more luck! Already in 1899 they reported that objects placed near to radium or polonium preparations become radioactive. The effect was by them referred to as: "radioactivité provokuée" or induced radioactivity. However, no emanation was mentioned by Curies. Moreover, they saw the phenomenon of induced radioactivity as some kind of secondary radiation excited by Becquerel-rays emitted by radium [18].

Fig. 3 shows a closed vessel used in 1904 and earlier by Curies and their collaborators for activation of bodies using radium [19]. The "induced" radioactivity was the same for all the exposed materials. The activated bodies maintained their activity for certain time, then the activity slowly decreased and eventually disappeared [19].

Around 1900 Ernest Dorn (1848-1916), a chemist working at the Physical Institute in Halle, Germany set out to investigate the emanation properties and the power to

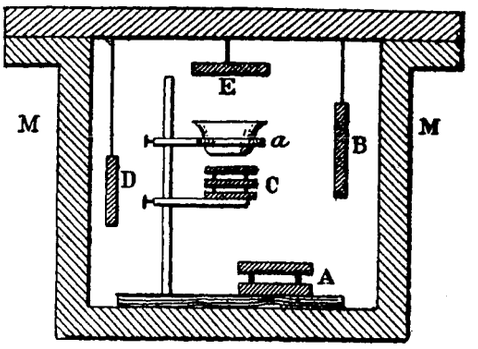

Fig. 3. A closed vessel used by the Curies and their collaborators for activation of bodies. M: closed vessel/space; a: radium salt; A, B, C, D, E; materials to be activated (lead, copper, glass, ebonite, cardboard, paraffin, celluloid) [19].

induce radioactivity ("sekundäre Aktivität") of a number of radioactive substances available in his institute. The E. Rutherford's publications and his finding that no other radioactive substances but thorium-containing ones seem to possess emanating power and are able to induce radioactivity triggered E. Dorn to start to work on these phenomena [20]. His experimental approach was similar to that of E. Rutherford. He tested metallic uranium powder, thorium oxide, radioactive i.e. radium-containing barium bromide obtained from "Chemische Fabriek E. de Haän" (Chemical Factory E. de Haän) in List near Hannover in Germany [21], radioactive i.e. radium-containing barium chloride and bismuth sub-nitrate (Polonium) of French origin (the last two). Although, his publication on this matter goes no further than just summing up the experiments and the obtained instrument readings, one may infer that in tests with radioactive barium salts he actually recognized the existence of radium emanation (radon) and demonstrated the "induced" radioactivity (half-life about 45 minutes) due to it. Anyhow, his article lacks a proper elaboration of the results of measurements and clear conclusions.

The existence of a third emanation was reported by Fridrich Oskar Giesel (1852-1927), a chemist working at Buchler & Co., a quinine factory in Braunschweig (Brunswick) in Germany. In 1902 [22], when discussing the utility of his zinc sulphide screens for detecting alpha radiation he mentions his observation of the "Rutherford's Emanation" given of by some radium impurities. The emanating body occurred after separation of rare earths from pitchblende with oxalic acid in the next ammonium precipitation residue. Eventually, F. O. Giesel was able to isolate an emanating substance allied with lanthanum but free from thorium [23]. He named it "emanium" [24]. However, Debierne claimed that "emanium" was actually "actinium" which he discovered a few years ago and he insisted on discarding the name "emanium". Later analysis of Debirene's publications revealed that his preparations contained very little actinium [25]. However, not the name "emanium" but "actinium" remained in use for the emanating body [25] and the obtained emanation was referred to as actinium emanation or actinon.

In 1903 H. Becquerel published a book [24] comprising an overview of the early research on the "new property of matter" i.e. radioactivity [26]. An important part of this book is a comprehensive bibliography of all relevant publications pertaining to radioactivity and radioelements and covering the period of February 1896 to 1 September 1903 (214 references).

3 Emanations & induced radioactivity: the follow-up

Following the discovery of emanations the research involving them and their disintegration products became very popular. The (chemical) identity of a number of these radioactive products was soon established. In Fig. 4 the status of the knowledge in 1911 regarding three disintegration series is given [27]. Each of the three emanations decays via a number of successive decay steps i.e. radioelements into – at that time still to be identified – final products. Today it is known that thorium, radium and actinium emanations (i.e. thoron, radon and actinon) belong to one of the three natural decay series starting with the long-lived radionuclides: ^{232}Th(orium), ^{238}U(ranium) and ^{235}U(ranium) and ending with stable lead isotopes ^{208}Pb, ^{206}Pb and ^{207}Pb, respectively. For complete decay schemes and characteristics of the involved radionuclides (including those which have been referred to as "induced" radioactivity) see the 1995 edition of the Karlsruhe Chart of Radionuclides [28]. According to that chart thorium emanation or ^{220}Rn (Tn) is given off by a radionuclide of radium, namely ^{224}Ra (ThX) and not of thorium; radium emanation or ^{222}Rn (Rn) is given off by radium, namely ^{226}Ra (Ra) and finally the actinium emanation or ^{219}Rn (An) is also given off by a radionuclide of radium, namely ^{223}Ra (AcX) and not of actinium. The half-lives of ^{220}Rn, ^{222}Rn and ^{219}Rn are 55.6 seconds, 3.8 days and 3.9 seconds, respectively.

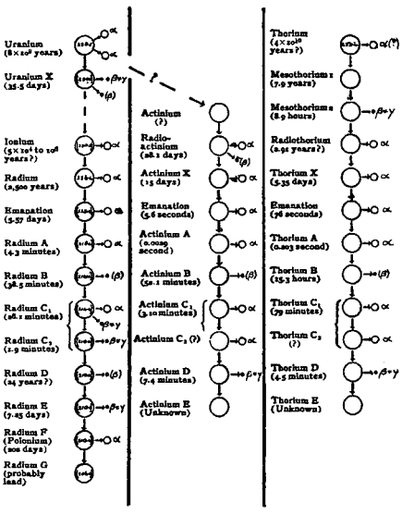

Fig. 4. The disintegration series as they were known in 1911. Each of the three emanations decays in a number of products giving rise to "induced" radioactivity [27].

Being radionuclides of the chemically inert element radon all the three emanations are indeed gaseous and belong to the 18th group (noble gases) of the Periodic Table of the Elements.

Soon after the discovery of emanations it became clear that a highly emanating substance is one from which a large fraction of an emanation i.e. a radioactive inert gas may escape before it decays [29]. "Emanating power was defined as the fraction of radioactive inert-gas atoms formed in solid that escape from the solid" [30]. The emanating power of a solid depends on the composition, crystal structure, specific surface, temperature of the solid and also on the half-life and recoil energy of the radioactive inert gas [29]. The remarkable difference between the emanating power of thorium oxide on one side and thorium sulphate and thorium nitrate on the other side [13] was most probably due to the unlikeness of their physical properties relevant to the gas (thorium emanation) transport through and out of the solid.

The emanating power – nowadays usually referred to as exhalation – of radon and thoron from soil and building materials governs the concentration of these emanations in open air as well as in the buildings. In the room air of some buildings high radon concentrations occur giving rise to considerable radiation exposure mainly through deposition of decay products in the lungs. Therefore, one has to assume that the general population has significant cancer risk, especially for lung cancer, from such exposure [31]. See also [32].

4 Emanations in science and medicine

Since its discovery, radium emanation (radon) has been used for many purposes; scientific and others. One of the salient applications of radon is its use as a source of RaD (^{210}Pb) to be applied as an "indicator" (radiotracer) for lead in a study on the solubility of lead sulphide and lead chromate in water.

To this end about 1/5 Curie of the emanation was allowed to decay above distilled water in a closed vessel. After complete decay of the emanation the obtained solution contained RaD. Ten mg $PbCl_2$ was added to the solution, lead was quantitatively precipitated with K_2CrO_4 as $PbCrO_4$ and the precipitate separated from the solution. Subsequently the precipitate was suspended in 100 cm^3 water and the suspension was shaken for 24 hours. After removal of the precipitate by filtration the filtrate was evaporated to dryness and the β^-- activity of RaD measured after the establishment of the decay equilibrium between RaD and RaE (^{210}Bi, half-life = 5.0 days). Comparison of thus obtained activity measurements with those of the equilibrated RaD solution used to prepare the radioactive lead chloride solution yielded the solubility of lead chromate of $1.2 \cdot 10^{-5}$ g per litre water. This study was carried out at the "Institut für Radiumforschung" (Institute for Radium Research) in Vienna, Austria, by Fritz A. Paneth (1887-1958) and Georg von Hevesy (1885-1966) and published in 1913 [33]. It is actually the very first publication based on the G. von Hevesy's concept of radioactive indicators (radiotracers)!

In 1944 G. von Hevesy was awarded the Nobel Prize in Chemistry for 1943 for "For his work on the use of isotopes as tracer elements in researches on chemical processes". Fig. 5. shows F. A. Paneth and G. von Hevesy in their twenties [34].

Fig. 5. Fritz A. Paneth (left) in 1912 and G. von Hevesy somewhere between 1911 and 1913 [34].

Not long after the advent of radium it was observed that it must not be handled without precaution. Strong radium preparations could cause skin burns to develop. After it was established that the rays from radium have a physiological action, it was suggested that they might have an application in medicine. Betterment in some skin diseases as a result of irradiation with radium emitted rays was reported. Carcinoma appeared to respond to such treatment as well. Much success has also been attained in cases of gout and rheumatism. Moreover, the science of that time has been inclined to attribute the healing action (?) of many natural springs (spas) to their content of radium emanation [35, 36].

With regard to medical applications the most widely used radioelement was radium followed by its emanation. Some other radioelements, for example thorium emanation (^{220}Rn) in equilibrium with its parent, thorium X (^{224}Ra) have also found their way into medical practice [37].

The most common routes to administer emanations to the patients were inhalation, drinking of emanation-containing water or bathing in it. All these routes eventually lead to entry of emanation into the blood. However, the inhalation is probably the best way for achieving high emanation concentrations in blood [36].

An early unit of measurement for emanation concentrations is called a "Mache-Einheit" ("M.-E.") or a Mache Unit (M. U.) [38]. One M. U. is equal to $3.64 \cdot 10^{-10}$ curie per litre. One curie (Ci) is the activity of emanation in equilibrium with 1 gram of radium and corresponds to $3.7 \cdot 10^{10}$ disintegrations per second or becquerels (Bq). Thus 1 M. U. = 13.5 Bq.

Fig. 6. shows an advertisement page in a book printed in 1904 [39]. As can be seen a London firm was offering – among many other materials and instruments related to radioactivity and radiation – their thorium inhalers by saying: "Our newly invented thorium inhalers may be had on hire". It seems that the therapeutic use of thorium emanation even preceded that of radium emanation [40].

Radium and Radio-Active Substances.

RADIUM

ON SALE AND LET ON HIRE.—Write for Terms.

The following can be had by return of post.

PITCHBLENDE, direct from Joachimsthal, very radio-active, from 1/- to 30.- per piece.

ITACOLOMITE, or flexible sandstone. 10/- to 25.- per piece (very rare).

KUNZITE, 1/- per gramme. Selected, clear, 2/-.

SPARTEITE (*see* "NATURE" 31/3/04, fol. 523), 2/- and upwards.

CHLOROPHANE, 2/- and upwards. BARIUM PLATINO CYANIDE IN LARGE CRYSTALS in course of manufacture. (Oxford Bouklis.)

ZINC SULPHIDE. Phosphorescing a beautiful Green, 2/6 per tube.
" " " " Yellow, 2/6 "
CALCIUM SULPHIDE " " Violet, 1/- "

RADIO-ACTIVE RESIDUE from which Radium is made: very scarce, 2/- per tube.

POLONIUM SULPHIDE, in tubes of one gramme, 21/-
" on Bismuth Rod or Disc 35/-
" on Copper " " 25/-

RADIO-ACTIVE SCREENS (Willemite), 6d. per sq. inch. Plat. Bar. Cyan Screen, 9d. per sq. inch.
" " (Zinc. Sulph.), 10 × 10 cm. 7/6.

SAMARSKITE,	THORITE,	BLACK and YELLOW
CARNOTITE,	ARRHENITE,	WUCKITT,
EUXENITE,	ORANGITE,	URANITE,
URACONITE,	AUTUNITE,	FERGUSSONIT,
BROGGERITE,	ALVITE,	FORBERITE,
SIPILITE,	URAN GLIMMER,	CHALCOLITE, &c.

RADIOMETERS, ELECTROSCOPES, SPINTHARISCOPES, VACUUM TUBES.

All may be had at reasonable prices.

Professional Men, Universities, Schools, &c., allowed special terms.

OUR NEWLY INVENTED THORIUM INHALERS MAY BE HAD ON HIRE.

ARMBRECHT, NELSON & CO.,
71 & 73, DUKE ST., GROSVENOR SQ., LONDON, W.
Telephone: OXBRIDGE 1052.

N.B.—We have received a consignment of the New Zealand Vegetable Caterpillar; it is from 2 to 3 inches long, with a stem showing fructification growing out of the head. Scientific name of the insect is *Hepialus virescens*: the name of the fungus is *Sphæria Hobertsiana*. Specimens may be had from 15/6 to 21/-, according to quality and size.

TERMS, CASH WITH ORDER.

Goods may be returned if not approved of, when money will be refunded.

TWO SPACIOUS DARK ROOMS TO SHOW RADIUM ACTIVITY.

Fig. 6. Advertisement page in a book printed in 1904. Note the offer reading: "Our newly invented thorium inhalers may be had on hire" [39].

Fig. 7 – 10 show pages of a four-page price-current (in Dutch) of radium and radium emanation related preparations, measuring instruments and services, emanation inhalers and apparatuses for drinking emanation containing water as offered in 1912 by the Amsterdam branch of the "Algemene Radium Maatschapij" (General Radium Society) "ALLRADIUM". The range of products and services is quite impressive. Both the inhalers and drinking apparatuses were available in two sizes, namely for domestic use and for hospitals.

Fig. 11 shows a large scale inhaler capable of providing radium emanation for a whole room [41], large enough to accommodate a number of patients. An emanation treatment session per day in a room of 60 m^3 required 17000 M. U. = 2.3 . 10^5 Bq radium as source of emanation (one of the four small cylinders contains radium solution) [42]. Typical values for emanation concentrations in treatment rooms were 2 – 4 M. U. [43] or 27 – 54 Bq per litre!.

Already for thousands of years people have been seeking health and youth in natural springs (spas). Although, all these people took for granted that mineral water coming out of such springs has curative properties it remained unestablished which of the (mineral) constituents of the spring water or even some other, unidentified property of water is actually the essential one. The balneologists claimed that only the natural mineral waters have medicinal value and that artificial waters although containing all the constituents found in natural waters still lack curative properties! Moreover, according to them an improvement in the state of illness can only be expected in the vicinity of the source of the mineral water to be consumed.

In 1903 Allen (?) [44] found radium emanation in water of a mineral water spring in Bath in England. This led to an avalanche of radioactivity i.e. radium emanation measurements in other baths and springs. However, no correlation has been found between the (alleged) effectiveness of a mineral water and its emanation content. Notwithstanding the fact that the mysterious active agent of mineral waters was still unrevealed [45] their application for medical purposes assumed large proportions.

According to [46] the radioactivity of spring waters is composed of:

(1) Emanation dissolved in water and also emanation transferred from water to air;

(2) Emanation in spring gases originating from radium but also from thorium and actinium;

(3) Radioactive salts dissolved in water;

(4) Radioactivity substances in the sediments.

In most mineral waters the radium emanation is the major fraction of the total radioactivity.

Fig. 7-10. Price-current for radium preparations, accessories, services and apparatuses for delivering radon or radon-water to the patients, issued by "Algemeene Radium Maatschappij" (General Radium Society) "ALLRADIUM", Amsterdam, 1912, 4 pp. Items pertaining to radium emanation: Radium-drinkapparaat "Allra" = Radium drinking-apparatus "Allra" (Capacity 1000, 2000 or 3000 Mache Units per day of radium emanation); Aqua Radium. Sol. bestendig radioactief. = Radium solution of lasting radioactivity; Radium-water, genomen uit den emanator: Radium-water taken out of the emanator; Inhalatorium voor Ziekenhuizen = Inhaler for hospitals; Inhalatieapparaat voor privégebruik = Inhaler for domestic use; Drinkapparaat voor privégebruik = Drinking-apparatus for domestic use and Drinking-apparatus for hospitals. All prices in *f* = Dutch Florins or Guilders (10 *f* was a gold coin).
The original price-current belonged to Godefridus Andreas Alphonsus Maria Kerssemakers (1875-1936) pharmacist in Tilburg, The Netherlands. It is now in possession of his granddaughter, Ms. Catharina Hermina Kolar-Janus, the author's wife.

PRIJS-COURANT.

Radium Pur.

Radium Sulfate „Alradium".

Radium-Compressen „Alradium".

Radium-Ampullen „Alradium".

Radium-Injectie in Phiolen à 2 cM. „Alradina".

Radium-Inhalapparaat „Alra".

Fig. 8

RADIUM-OPLOSSINGEN

voor Drink- en Badkuren.

RADIUM

Meet-Instrumenten

Onderzoekingen.

Fig. 9

Fig. 10

Fig. 11. Radon inhalation-room as used for patient treatment in Homburg von der Höhe, Germany, around 1912 [41].

With regard to their radioactivity (emanation) the mineral springs can be divided in strong with 100 and more M. U./litre water, medium with 50-100 M. U./litre and week ones with 20-50 M. U./litre. Some of the strongest springs were located in Jáchymov and Bad Gastein (Austria) [47].

Drinking radium emanation i.e. radon containing water as well as bathing in such water were the two most common ways of patient treatment. In the early days of the use of spring water 600 M. U./litre = 8100 Bq/litre water was a common therapeutic concentration of radon in Jáchymov [11]. Today radon baths of the activity of 5500-7500 Bq/litre are used as the basic treatment in Jáchymov [9].

At the end one may put the question whether the application of radioactive emanations for medical treatment throughout almost a whole century, the 20th century, was justified in spite of the possible health consequences of the exposure to radiation emitted by emanations and their decay products? Should one look upon the application of emanations for medical treatments as beneficial use or perhaps as misuse, though a well meant-one? Finding a comprehensive answer to these questions exceeds the author's humble qualifications.

The auther thanks W. den Hollander for his help in preparing the document and the figures.

References

[1] W. C. R. Röntgen: Sitzungsberichte der Physikalisch - Medicinischen Gesellschaft zu Wüzburg, (1895) 132.

[2] H. Becquerel: C. R. Acad. Sci., Paris, **122** (1896) 501.

[3] H. Becquerel: C. R. Acad. Sci., Paris, **123** (1896) 855.

[4] H. W. Kirby, *The Early History of Radiochemistry*, AEC Research and Development Report, Monsanto Research Corporation for the U. S. Atomic Energy Commission, MLM- 1960, September 22, 1972, 126 pp., p. 10.

[5] G. C. Schmidt: Verhandlungen deutsh. phys. Ges. Berlin, **17** (1898) 14.

[6] Sklodowska Curie: C. R. Acad. Sci., Paris, **126** (1898) 1101.

[7] P. Curie and S. Curie: C. R. Acad. Sci., Paris, **127** (1898) 175.

[8] P. Curie, Mme. P. Curie, and G. Bémont: C. R. Acad. Sci., Paris, **127** (1898) 1215.

[9] H. Günterová and M. Surová: *JÁCHYMOV St. Joachimsthal*, 2nd edition, Léčebné lázně a.s. Jáchymov, Jáchymov, 1997.

[10] M. Kraus: *Das staatliche Uranpecherzbergbaurevier bei St. Joachimsthal in Bömen*, Wien, 1916.

[11] A. Zeman and P. Beneš: Radiochim. Acta, **70**/**71** (1995) 23.

[12] E. Curie: Madame Curie; *Haar leven en werk* (authorised adaptation of the book: La vie de Madame Curie from the French into Dutch language by W. Corsari) 2nd printing, H. P. Leopold's Uitg.-Mij. N. V., The Hague, 1937, illustration between p. 192 and 193.

[13] E. Rutherford and R. B. Owens: Trans. R. Soc. Can., 2nd series, **5**, Section 3 (1899) 9.

[14] E. Rutherford: Phil. Mag., 5th series, **49** (1900) 1.

[15] E. Rutherford: Phil. Mag., 5th series, **49** (1900) 161.

[16] F. Soddy: *Radio-Activity*, "The Electrician" Printing & Publishing Company, London, 1904, p. 103.

[17] A. Romer: *Radiochemistry and the Discovery of Isotopes*, Dover Publications, New York, 1970, illustration between p. 8 and 9.

[18] P. Curie and M^{me} M.-P. Curie: C. R. Acad. Sci., Paris, **129** (1899) 714.

[19] J. Danne: *Le Radium; Sa préparation et ses propriétés*, Librairie Polytechnique Ch. Béranger, Paris, 1904, p. 54.

[20] E. Dorn: Abhandlungen der Naturforschenden Gesellschaft zu Halle, **23** (1901) 1.

[21] E. de Haen: Ann. Phys. und Chem., Leipzig, **68** (1899) 902.

[22] F. Giesel: Ber. deutsch. chem. Ges., **35** (1902) 3608.

[23] F. Giesel: Ber. deutsch. chem. Ges., **36** (1903) 342.

[24] F. Giesel: Ber. deutsch. chem. Ges., **37** (1904) 1696.

[25] Reference [4] pp. 24-25.

[26] H. Becquerel: *Recherches sur une Propriété nouvelle de la Matiére, Activité radiante spontanée ou Radioctivité de la Matiére*, Typographie de Firmin-Didot et C^{ie}, Paris, 1903.

[27] Reference [17] p. 192.

[28] G. Pfennig, H. Klewe-Nebenius, W. Seelmann-Eggebert: *Karlsruher Nuklidkarte* (Karlsruhe Chart of the Nuclides), 6th Edition, Forschungszentrum Karlsruhe, Karlsruhe, Germany, November 1995.

[29] A. C. Wahl: in *Radioactivity Applied to Chemistry* (A. C. Wahl, Editor and N. A. Bonner, Assistant Editor), John Wiley & Sons, New York and Chapman & Hall, London, 1951, p. 284.

[30] O. Hahn and O. Müller: Z. Electrochem. **29** (1923) 189.

[31] J. Porstendörfer: *Properties and behaviour of radon and thoron and their decay products in the air*, Tutorial Session, Fifth International Symposium on Natural Radiat. Environment, NRE V, Salzburg, Austria, September 22-28, 1991, 77 pp.

[32] B. Wahlström: *Radiation, Health and Society*, International Atomic Energy Agency, Vienna, 1997, 56 pp.

[33] G. v. Hevesy and F. Paneth: Z. anorg. Chem., **62** (1913) 323.

[34] H. Levi: *Georg de Hevesy, Life and Work*, Rhodos, Copenhagen, 1985, p. 32.

[35] G. Hevesy and F. Paneth: *A Manual of Radioactivity*, (Translated by R. W. Lawson), Oxford University Press, London, 1926, p. 209-210.

[36] Lipliawsky and H. Lungwitz: *Die Radioelemente in der Heilkunde*, Adler-Verlag, Berlin, 1913.

[37] Reference [36], p. 178-180.

[38] Reference [35], p. 178-179.

[39] Reference [16], first p. after p. 214.

[40] Reference [36], p. 162.

[41] Is. Zeehandelaar JBz.: *Het radium en de geneeskunde*, (Radium and the Medicine), H. Meulenhoff, Amsterdam, 1912, illustration between p. 60 and 61.

[42] Reference [41], p. 61.

[43] Reference [36], p. 150.

[44] G. Mutschlechner: *Die Entdeckung der Radioaktivität in den Gasteiner Kurmitteln und ihre Bedeutung*, Sonderabdruck aus dem Badgasteiner Badeblatt (1959, p. 2), Nr. 2 und 3/ 1965, Verlag der Kurverwaltung, Badgastein, 1965, p. 4.

[45] Reference [36], p. 241-248.

[46] Reference [36], p. 108.

[47] Reference 36, p. 244.