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A BRIEF HISTORY OF LORD RUTHERFORD'S RADIUM

by

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In this paper I give a brief summary of what is known about the acquisition, use and fate of the radium sources that were in the possession of Lord Rutherford during his lifetime. The account is written in two parts, corresponding to the periods from the discovery of radium in 1898 until his death in 1937 and then from 1937 until recent times. The history of Rutherford's radium closely shadows the history of radioactivity, the evolution of nuclear physics, the race for the bomb, and the development of the nuclear industry.

Keywords: radium; Rutherford; radioactivity

Radium stands out among the naturally occurring isotopes for its radioactive potency, the degree to which it captured the public imagination after its discovery, and its unique role in the development of science and medicine. For these reasons the broad history of radium is of considerable interest, and several previous works have given an account of this.[1] The background to the present work is an interest in the origin and fate of radioactive substances in the possession of Rutherford that led to contamination in his laboratories in Manchester, and which was recently the subject of a review. Although a preliminary account of this has been provided as part of the review,[2] given Rutherford's unique and central role in the development of modern physics there is a particular interest in tracing specifically the history of the radium in his possession that was accumulated during his career. This account follows on from a recently published description of the role of the Royal Society in these affairs.[3]

DURING RUTHERFORD'S LIFE

Montreal, 1898–1907

During the period when radium was discovered, isolated and made commercially available, Rutherford worked at McGill University in Montreal as Macdonald Professor of Physics. Much of his important work there was done in collaboration with the chemist Frederick Soddy between 1900 and 1902.[4] Together they worked out the theory of successive transformations to explain the nature of radioactivity, but most of this work was done with thorium. His earliest work using radium was performed with preparations, variously

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obtained from Julius Elster and Hans Geitel in Germany and the Curies in Paris,[5] that were in the form of the barium radium chloride salt. These sources were, however, rather weak and limited in supply.

The situation changed markedly when more concentrated samples produced by the German chemist Friedrich Giesel became readily available. In the spring of 1903 Soddy moved to London to work with William Ramsay at University College London (UCL) on the production of helium from radium. Shortly after arriving in London, Soddy chanced upon Isenthal's shop on Mortimer Street, about 20 minutes walk from UCL, which was advertising Giesel's radium:

> Our trouble here was the same as in Montreal. We had a quite insufficient amount of radium for our investigations. Then, by the most extraordinary chance, the whole future prospect was changed. ... I was walking along Mortimer St off Upper Regent Street, in London one day when, casually looking through Isenthal's window, I saw advertised something I could not credit to be true: 'pure radium compounds on sale here'. ... I learnt from the salesman that Professor Giesel in Germany ... had started manufacture of radium compounds on a commercial scale in the Chinin Fabrik at Brunswick. He used residues left after the extraction of uranium from the Pitchblende found in the old state silver mine of St Joachimsthal in Bohemia. Isenthal had quickly taken advantage of this supply. At that time one could buy radium only from the French factory by favour of the Curies. Here it was to be bought in a London shop at some eight shillings a milligram of pure radium bromide.[6]

Soddy purchased 20 mg of radium bromide, which was delivered to Ramsay's laboratory sometime in early July.[7] Sometime shortly after this, in the summer of 1903, Rutherford visited Soddy in London.

> I told him immediately on his arrival at UCL about my find at Isenthal's and together we lost no time in walking round to Mortimer St. I must say he experienced the same astonishment and joy that I had experienced when confronted with radium bromide for eight shillings a milligram some weeks before. He was absolutely bowled over and became as excited as a school boy over the coming holidays. With thirty precious milligrams of pure radium bromide we bounded back to Gower Street and we both immediately repaired to the dark room with some metal foils and a bit of X-ray screen. The effect was terrific; it was like a person born blind suddenly being given sight, for though R had made a special study of the Becquerel rays, this was the first time he had ever seen them. All his work had been done by the ionization method with substances far too feebly radioactive to light up the X-ray screen. Now he had a visual demonstration of what he had found out in the dark—so to speak.[8]

Before his return to Montreal, Rutherford lent his 30 mg of radium to Soddy and Ramsay as a contribution to the helium experiments, the apparatus for which is shown in figure 1. Soddy wrote to Rutherford on 13 July as follows:[9]

> If you received my wire you will have heard the results of the second experiment with your radium. ... It is a veritable triumph and we are very grateful to you for making it possible. Your radium weighed 31.8 mg and I hope soon to hear from the manufacturers when I shall get my 30 mg from them.

It is apparent from the subsequent letters between Soddy and Rutherford during autumn 1903 that Isenthal's reputation had been damaged by the selling of poor-quality preparations

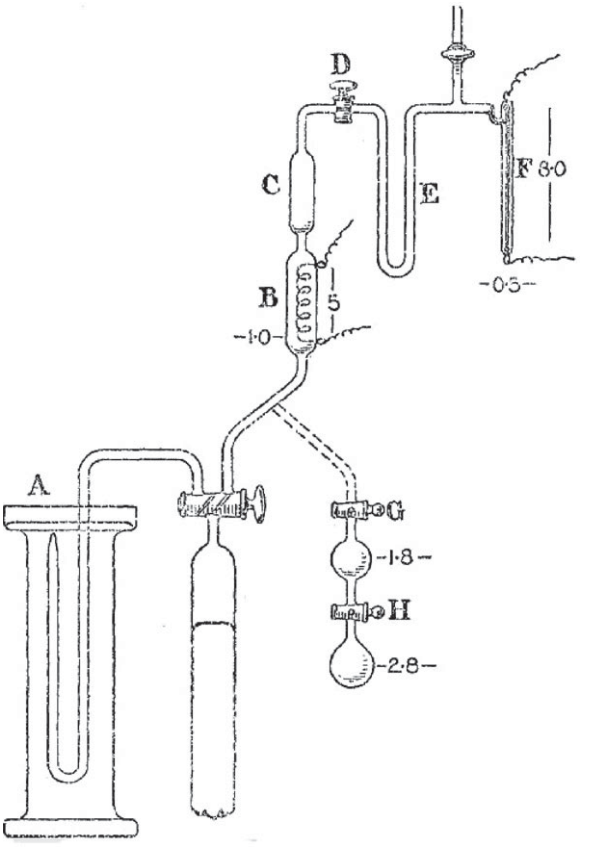

Figure 1. Apparatus used by Ramsay and Soddy to measure the spectrum of helium from radium purchased from Isenthal. The gases could either be introduced at inverted siphon A in a mercury trough from a separate apparatus (Toepler pump & radium bulb), as was done with Soddy's 20 mg, or introduced directly from bulb H, as was done with Rutherford's 30 mg. (Adapted from figure 2 in Sir William Ramsay and Frederick Soddy, 'Further experiments on the production of helium from radium', *Proc. R. Soc. Lond.* **73**, 346–358 (1904).)

and he was forced to withdraw an advertisement in *Nature* in September 1903. On 18 September Soddy wrote:[10]

> Isenthal's stuff is I regret to say to all appearances identical to the 30 mg I got recently from Müller-Uri [a German glass-blower and agent for Giesel]. This in light of the advert he has in this week's *Nature* is somewhat serious. However, I believe he is acting in good faith. He wrote to me by return today after I had sent him the unfavourable report … until he has my definitive report he will not repeat his advertisements. … Isenthal had about 0.1 gram of the rotten stuff for which he paid 90% more than the old & sells it I believe at £6.10 for 5 mg.

Consequently Ramsay visited Giesel in Brunswick, and thereafter further preparations were sent direct to London. At least two of these were forwarded to Montreal, including compensation for the 30 mg. On 12 October Soddy wrote:[11]

> We are in luck for Giesel sent us his pure $RaBr_2$ this morning & fine stuff it is too. He only charged us 12 Marks per mg which is less than you paid Isenthal. Tomorrow when I go to the lab I will weigh out 31.8 mg and send this to you with this letter . . . .

It would appear from Soddy's letters that Rutherford had tried to obtain at least 50 mg more, but there was already a limit on the supply. On 27 October Soddy wrote:[12]

> with regard to the 50 mg I had ordered from Buchler for you, they write me that as you have already some on order from Müller-Uri, they cannot book this order, as they wish the very small amount of the preparation to find its way to as many . . . as possible.

On 19 November:[13]

> With regards the radium, I hear Müller-Uri has dispatched his 30 mg which I have not yet received, but should today. I paid £22.10 for it and enclose the receipt. This is what you originally paid Isenthal and Müller-Uri tells me the price is right. If you will take this lot you are welcome and I will send you immediately . . . . For the future your best plan is to deal directly with Buchler & Co, Chinin-Fabrik, Braunschweig. I have been in contact with them and they say that they would prefer to deal direct with scientific men and not with dealers. But if you want more you should order it at once.

On 4 December:[14]

> I sent you . . . last week from Dundee 30 mg radium in a registered letter that I hope you received.

From the above we may infer that in addition to his first 30 mg from Isenthal's he had received another 30 mg sent by Soddy and an unknown amount ordered from Müller-Uri referred to in Soddy's letter of 27 October. A reference is made to the Montreal acquisitions in the 1902–03 McGill Council Reports[15] by Professor John Cox:

> 80 milligrams of this rare substance have been secured, the total existing amount being probably not more than one gram; and this will be employed for further research.

It is not clear whether Cox's report includes all of the consignments received by Rutherford but we may guess that he had between 80 and 160 mg of the bromide. It is apparent from other parts of Soddy's letter of 4 December that Buchler were already experiencing a shortage of ore.

> Do you know I have a shrewd suspicion that Curie has knobbled the Austrian government & secured the monopoly of the Joachimsthal mine, the only practicable source of radium, damn him. The Austrian government have closed down all supplies of the residues & Curie is now reported to live in Vienna. The German people write me that the residue is not to be had. Things will soon look bad, unless they find some in your part of the world.

The brief era of cheap radium was soon to come to an end, and within a matter of months price inflation set in. In 1902 the price of a milligram was about 10 Marks, but in 1904 the price increased rapidly from 37 to 93 Marks. By 1910 it had reached 500 Marks and only in 1914 settled at around 800 Marks.[16] Undoubtedly supply and demand were a major factor in driving the price, not least the attempt by the Austrian government to monopolize the supply (see the next section), although Rutherford contended that this was to a certain extent artificial.[17]

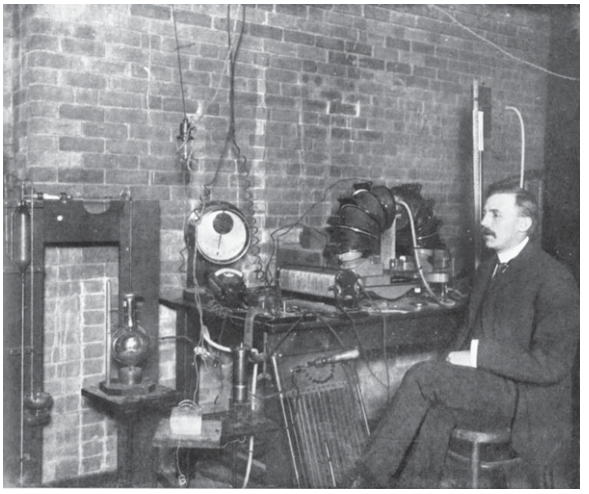

Figure 2. Rutherford in the basement of the Macdonald Laboratory at McGill University in 1905. On the left is shown one of the Toepler pumps made by Müller-Uri with a bulb attached containing radium salt in solution.

It is also apparent from Soddy's letters that, in addition to radium, two Toepler mercury pumps, with accessories, produced by the glass-blower and instrument maker Müller-Uri, were also shipped over from London in October 1903. In his letter of 18 September 1903 complaining about the poor-quality radium, Soddy had continued:

> I saw the pumps Müller is making for you. They have a rather narrow main tube which will make them slow in action, but otherwise they are perfect. But he had taken you to mean that you wanted the [?] thing complete with the arrangement for filling vacuum tubes attached to pumps as used at Univ. Coll. [see figures 1 and 4*b*], and this, although it would no doubt be useful, makes the pumps stand about twice as high. So I took upon myself to tell him to send you one complete mounted pump, & not to mount the other pump but send it as a spare one, without any of the accessories, by itself. You will be able to fit the stand then if you want it mounted. I also told him to send a glass trough for mercury [that?] we use & four gas test tubes, as you would be down if you did not have these, if you want to use it for filling vacuum tubes. I also told him to enclose a little of the tap grease which our lab man has made for you.

The significance of this communication is that it is clear that Rutherford adopted Ramsay's apparatus and method of manipulating the gases evolved from radium, which he had developed over the years he had been working on the noble gases. One Toepler pump would be used to keep the radium in solution (see figure 6*a* below) and the other would be used for purifying emanation and filling vacuum tubes (as shown in figures 1 and 6*b*). From this time onwards he kept his radium apparatus in his basement laboratory at the Macdonald Laboratory, as can be seen in a photograph from 1905, reproduced in figure 2.

After the acquisition of the Giesel radium in 1903, the vast bulk of Rutherford's research was performed with this supply. Some 20 publications on radium appeared in 1904 and 1905 concerned variously with the heating effect of radium, the nature of the slow transformations of radium (Pb/Bi/^{210}Po) and the nature of the α-particle.[18] When Rutherford moved to Manchester in 1907, the Giesel radium probably remained in Montreal.

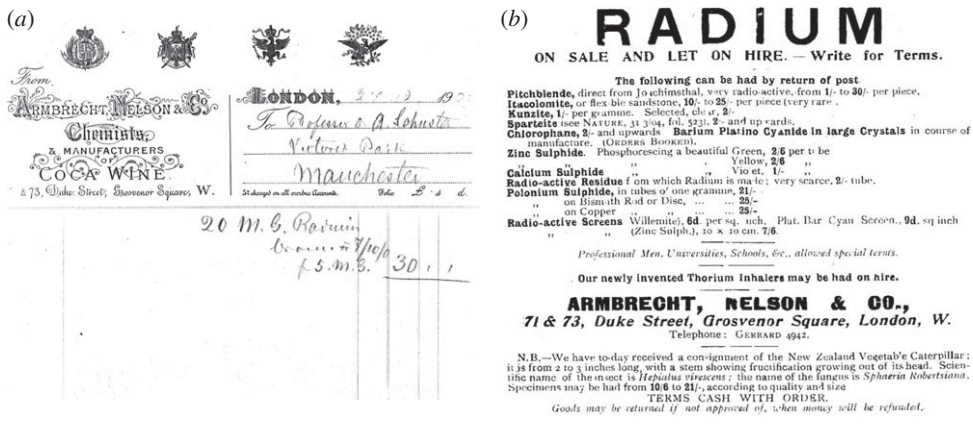

Figure 3. (*a*) Receipt for 20 mg of $RaBr_2$ from Armbrecht to Arthur Schuster dated October 1903.[72] (Reproduced by courtesy of the University Librarian and Director, the John Rylands University Library, The University of Manchester.) (*b*) An advertisement by Armbrecht for radium, dating from 1904.

Manchester, 1907–19

Before Rutherford's arrival in Manchester, Arthur Schuster in 1903 initially obtained 20 mg and subsequently a further 40–50 mg of radium bromide, so that by 1906 he had in total about 60–70 mg. His first 20 mg was purchased from Armbrechts, Nelson & Co. of Duke St, London, who at that time were advertising the sale of a variety of radioactive substances (figure 3). Several experiments were conducted with this radium by Walter Makower, Sidney Russ and others.

As noted above, in about 1905 the dominance of Buchler & Co. was challenged when the Austrian government set up a radium production plant, and two years later it embargoed all exports of uranium ore and products in an attempt to monopolize the market. This continued until about 1913, when other sources could be exploited, for example from Cornwall, Portugal and Colorado. Between 1905 and 1906 the Austrian Academy of Sciences in Vienna had obtained about 4 g of radium chloride, and in 1908 the Institute for Radium Research was founded and directed by Stefan Meyer.[19] Thus, when Rutherford moved to Manchester in 1907 Austria was the only place to obtain significant quantities of radium.

One possible route by which he might have acquired radium at this time was through the Royal Society, which, as has been documented, established a radium research fund and committee in 1903/04 and obtained in the autumn of 1906 the outputs of the treatment of 500 kg of Austrian pitchblende residues. This work was performed by the French chemist Armet de Lisle at Nogent-sur-Marne near Paris, and resulted in 412 g of partly worked-up radium barium chloride, and three other fractions containing actinium, radio-lead and polonium. The radium fraction was lent to the Government Chemist for purification, so it would not then have been available. However, Rutherford did obtain, in July 1907, 40 kg of actinium-bearing material, which he arranged to have processed by H. C. Greenwood at Thomas Tyrer & Co. of London,[20] resulting in 160 g of precipitate, and later in 1910 by the chemist Bertram Boltwood at Manchester, to produce 1.8 mg of thorium oxide, which included 5 mCi of ionium (^{230}Th, the parent of ^{226}Ra) and 10 g of preparation that contained 30 mCi of actinium (^{227}Ac). It seems likely that Rutherford also acquired the radio-lead

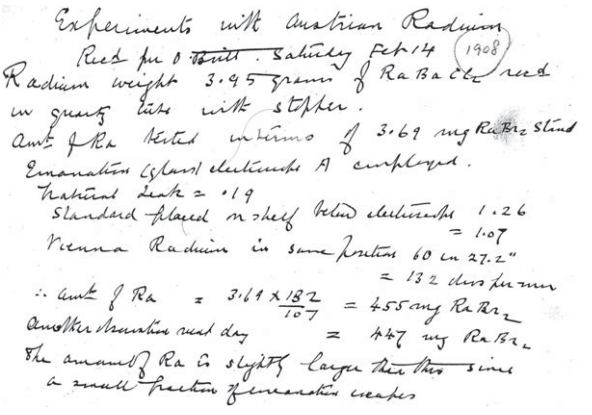

Figure 4. Rutherford's laboratory notes recording the arrival of the Austrian radium in February 1908. (Reproduced from AD 7653/PA 182, by courtesy of the Syndics of Cambridge University Library.)

fraction, which the chemist Georg von Hevesy was asked to extract the ^{210}Pb from; his failure to do so led eventually to the discovery of isotopes and the method of tracers.

Although undoubtedly pleased with the residues, Rutherford was keen to obtain radium. and he would have been aware of the efforts to obtain Austrian radium from contacts at the Royal Society. However, the loan of 300 mg $RaBr_2$, when it came, was made jointly to both Ramsay and Rutherford, and it was delivered to Ramsay's laboratory at UCL. This led to a dispute between Ramsay and Rutherford over how to divide its use. Ramsay wished to keep the whole together at London with his Giesel radium and send draws of emanation from the aggregate up to Manchester, whereas Rutherford wanted the 300 mg at Manchester. I do not intend here to go over once again the already well-told sequence of letters that were exchanged by the two protagonists in November 1907. It is worth noting, however, that there are at least two versions of the story, Rutherford's champion being the biographer Arthur Eve,[21] and Ramsay's being the biographer Morris Travers.[22]

Clearly, however, Rutherford was not happy with this arrangement, because he had made direct representations for his own radium. His notes headed 'Experiments with Austrian Radium'[23] record the arrival and are reproduced in figure 4, the text of which reads:

> Rec^{d} from O Brill. Saturday Feb 14 [1908].
> Radium weight 3 .95 grams of $RaBaCl_2$ rec^{d} in quartz tube with stopper.
> Am^{t} of Ra tested in terms of 3.69 mg $BaBr_2$ stand.
> Emanation (glass) electroscope A employed.
> Natural leak = .19 [divisions per minute]
> Standard placed in shelf below electroscope, 1.26 = 1.07
> Vienna Radium in same position, 60 [divisions] in 27.2″ = 132 divs per min
> ∴Am^{t} of Ra = 3.69 × 132/1.07 = 455 mg $BaBr_2$
> Another observation next day = 447 mg $BaBr_2$
> The amount of Ra is slightly larger than this since a small fraction of emanation leaks.

These laboratory notes are interesting in several ways. During the early days of radioactivity one of the most important instruments was the gold-leaf electroscope. After it had been charged up with static electricity it was possible to measure an ionizing

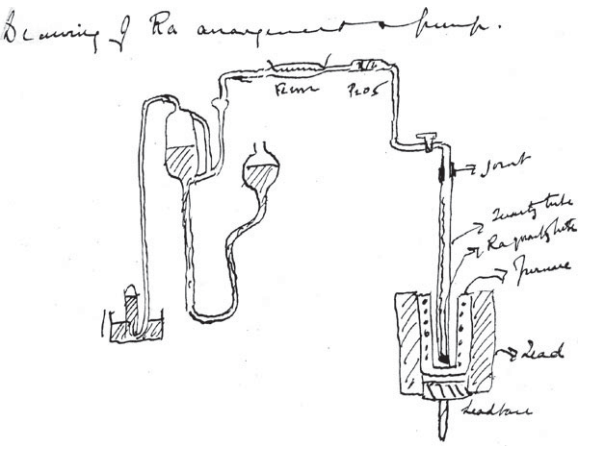

Figure 5. Rutherford's apparatus for obtaining emanation by heating radium in a quartz tube, used between February and May 1908. (Reproduced from AD 7653/PA 182, by courtesy of the Syndics of Cambridge University Library.)

source by the rate at which it caused the electroscope to leak. Note that although the preparation was actually radium barium chloride, its strength was estimated in terms of the equivalent radium bromide, which is a source of confusion in the literature. This radium was delivered personally by Otto Brill of the Vienna Academy, who had also delivered the shared radium to Ramsay and was then also doing some work with the latter.[24]

Ramsay to Rutherford, 17 February 1908:

> Brill told me of your present, and I congratulate you on it. If I were you I should dissolve it in acid—HCl for choice—and transfer it to a bulb of say 50 ccs capacity. You have only 0.375 gram of solid and that would be ample to contain it. Seal it to a pump like mine: if you want any particulars, only write, and I shall be delighted to give you all I know. All the same, if you want any large quantity of emanation, just let me know, and we will send a draw, as before. We can easily spare it from time to time . . . .

In fact Rutherford had already immediately embarked on an extensive series of experiments to measure the emanation given off when heating radium salt to 500–900 °C. The radium was placed in a thin quartz tube enclosed within a larger quartz tube in a furnace; the evolved gases were led to a Toepler pump (see figure 5). After four months of such experiments, however, on noting on 7 May that 'continued heating had cracked a new quartz tube'[25] he abandoned this method and returned to the method of dissolving the radium salt in hydrochloric acid in a 50 cc bulb attached to a Toepler mercury pump, as recommended by Ramsay (see figures 2 and 6). Hans Geiger later recalled this change:[26]

> Rutherford's work got off to a quick start in 1908 as a result of a loan of 250 mg of radium from the Academy of Sciences in Vienna—a very considerable quantity at that time. The radium salt was not highly concentrated and in general was of no direct use as a source of radiation. Rutherford decided that the best thing to do was to devise a method that would yield regular amounts of emanation, so that the emanation could be used whenever intense radiation was required. At first, attempts were made to extract the emanation by heating the salt, but this proved unsatisfactory. Rutherford then dissolved the salt and pumped

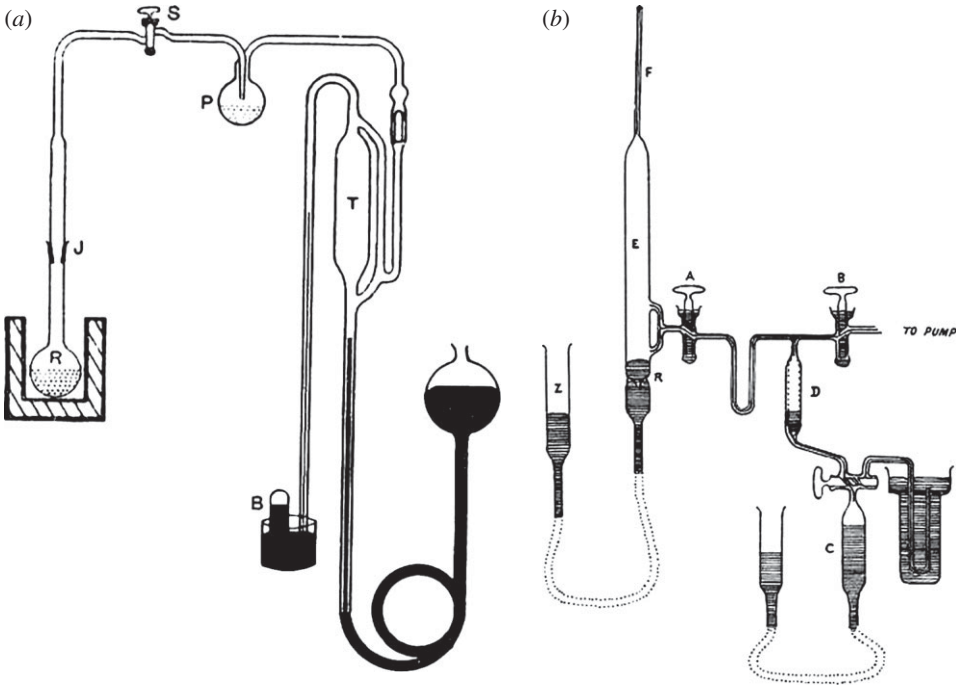

Figure 6. (*a*) Apparatus for keeping radium in acid solution and for drawing emanation, adapted from Fig. 107 of *Radioactive substances and their radiations*.[73] (Reproduced with permission from Cambridge University Press.) (*b*) Apparatus for the purification of emanation and transfer to 'emanation tube' F, adapted from Fig 1 of E. Rutherford, 'Experiments with the radium emanation. I. The volume of the emanation', *Phil. Mag*. **16**, 300–312 (1908). (Reproduced with permission from Taylor & Francis.)

> off the emanation with the gaseous products. The emanation was subsequently cooled and condensed in liquid air, and passed into special containers, mostly thin small tubes.

This apparatus was thereafter kept in a 'radium room' on the top floor of the Schuster Laboratory, the same room where Rutherford and his student Thomas Royds performed an experiment to determine the helium nature of the α-particle. This room had the advantage of having two windows, one being a large bay window, which would in principle allow radon to be ventilated outside the building rather than diffusing through the building, as it had done in the Macdonald Building.

The basic principle of Ramsay's Toepler pump set-up was that it would allow small quantities of radon to be 'milked' off the radium and then transferred over mercury to other apparatus. The radon could then be purified and compressed into a small glass tube to use as an α-particle source or used to transfer the active deposit, the decay products of radon, onto a metal button to use as a radium-C (^{214}Po) α-particle source.[27] Before World War I, much work was conducted in the basement where Hans Geiger, Ernest Marsden and Henry Moseley had their laboratories.[28] It was therefore necessary to carry the sources down the five flights of stairs from the top floor, often in a hurry. In the earliest days at Manchester, work on the extraction of radium emanation in the radium room was

performed by a laboratory assistant, William Lantsbury.[29] During World War I, at the time that Rutherford did his work on transmutation of nitrogen by bombardment with α-particles, much of the source preparation was done by William Kay in the preparation room at the other end of the top floor.[30] After World War I he had some help from Ernest Marsden and James Chadwick.[31]

Cambridge, 1919–37

After the end of World War I, in 1919, Rutherford was appointed Cavendish Professor and at that time he took with him a large amount of apparatus and his supply of radium, after evaporation to dryness, along with all the glassware.[32] He redissolved the radium and set up the Toepler pump in rooms at the top of the Cavendish Tower.[33] He set up his own laboratory room at the east end of the Cavendish wing on the ground floor,[34] where he kept most of his old Manchester apparatus, and set Chadwick to work in the basement under the 1896 South Wing,[35] close to the bottom of the tower, to continue the transmutation work started at Manchester. It is apparent from the notebooks of late 1919 and early 1920[36] that there were in fact two pumps working in the tower rooms to feed the Rutherford and Chadwick experiments in the basement with Ra-C (^{214}Po) and Ra-F (^{210}Po) sources.[37] The sources at this time were prepared by George Crowe, who had been sent to Manchester to learn from Kay how to handle radioactive substances.

It is certain that the Cavendish had purchased radium before Rutherford's arrival, because J. J. Thomson, Walter Makower, Lord Rayleigh (R. J. Strutt) and others had done work there with it, although dates of purchase and amounts are not clear. Thomson and Rayleigh, along with William Ramsay, were also members of the Executive Committee of the UK Radium Institute.[38] Later records show that the Cavendish owned about 408 mg of hydrated bromide or 218 mg of equivalent radium element. In 1921 the Medical Research Council (MRC) made a loan of radium to Rutherford and the records show that this amounted to about 493 mg of hydrated bromide or 264 mg of equivalent radium element. Thus, by this time he had acquired a substantial quantity of radium, which included the old Manchester batch as well as the Cavendish radium, totalling about 1356 mg of equivalent hydrated radium bromide or about 723 mg of element. In addition to three large Manchester, Cavendish and MRC sources, Rutherford also acquired some smaller sources, including 10 mg from the Vienna Radium Academy.

For the next decade or so this large source was tapped regularly to supply experiments in the basement laboratory and throughout the Cavendish by the large number of research students who came to work with Rutherford. At peak output Crowe would be expected to produce several sources per day, which over years took its toll (one amputated finger and skin grafts on several others). This was to change, however, after the *annus mirabilis* of 1932 when the era of the particle accelerator began and the neutron was discovered. One of the last of Rutherford's students at the Cavendish, Samuel Devons,[39] recalled this change:

> For decades this radium—there was approximately one gram of radium bromide in the Cavendish—had provided the basis for much of Rutherford's research, but now it was hardly in demand at all. Cockroft and others were busily exploiting the high voltage equipment he had built, and newer, better such equipment had been ordered from Philips in Holland. Rutherford had built for himself a smaller equipment of some 250 kV, which he and Oliphant used; a cyclotron had already been planned and construction was about to begin; Chadwick had left Cambridge and was busy with similar preparations at

> Liverpool. Sealed radium-beryllium sources were still used for the new neutron investigations, but the old treasure, the 1000 millicurie source—in solution—was freely available even to a humble graduate student. ... For more than a year I enjoyed a virtual monopoly of the radium source—perhaps the first (and last) student to be so privileged.

Several items can be found in the archives from the last few months of Rutherford's life that illustrate the changes since the early days of radium, and which point to the future. One of these was a letter from Rutherford to the secretary of the Cavendish dated 22 April 1937:[40]

> Some time ago the Union Miniere of Brussels loaned me 100 mgm. of radium for neutron work, and this source is invaluable to us for a number of experiments. As we shall want this source permanently, I think it undesirable to be under an obligation to the Belgian firm in this matter, and with your consent I would propose to purchase the 100 mgms. at the low rate of £4 per mgm. If you agree, I presume this should be paid from the Austin fund.

The Joachimsthal mine had long ceased to be the main source of uranium ore when other deposits were found around the world. One of the largest sources was the Katanga mine in the Belgian Congo, and as owner of the mines Union Minière du Haut Katanga (UMHK), after opening a uranium refinery in 1922, enjoyed a near monopoly in the supply until the outbreak of World War II. Rutherford's letter also indicates that radium was now needed primarily for neutron sources, as does an invoice dated 10 May 1937 for £395 from the Chemical Services Co. Ltd of 43 Parker St, London WC2:[41]

> RADIUM
>
> 98.75 mgrms Radium Element in the form of chloride mixed with Beryllium and contained in one brass tube No. 1-84476, at £4 per mgrm. element.

Although radium had become redundant as a source of energetic particles for nuclear reactions, mixed radium–beryllium neutron sources were now in demand in sealed containers. What, then, was to become of his old sources in solution in the tower rooms?

It is fortuitous for the history of science that in 1927 the ownership of the MRC radium was transferred to the Department of Scientific and Industrial Research (DSIR) under the management of the Government Chemist, because this allows us to keep detailed track of the fate of this batch. The MRC in 1927 had wanted the radium back for other purposes; at that time there was a large demand for radium to supply the hospitals. During the negotiations over the radium, H. T. Tizard of the MRC recorded in a handwritten footnote a conversation he had had with Rutherford:[42]

> Sir Ernest said all, or nearly all his radium was dissolved for the purposes of extracting radium emanation. It would mean a considerable upheaval of his organisation if he had to extract it all again in solid form and replace it with impure material.

It was therefore resolved that the DSIR would take ownership of the MRC radium on loan to Rutherford in exchange for another source to be transferred to the MRC after purification by UMHK. As part of the arrangement, so that the government could keep track of its radium stock, the Cavendish was required to sign an annual certificate thereafter. The exact wording of the certificate was as follows:[43]

> I certify that I hold, on behalf of the Department of Scientific and Industrial Research, radium compound estimated by the National Physical Laboratory to contain 493 $\pm$ 3% milligrammes of radium bromide ($RaBr_2 + 2H_2O$) (equivalent to 264 milligrammes of

> radium element) the property of H.M. Government, and that I am liable to return this if required in case of emergency or if no longer needed by me for scientific purposes.

Every year from 1928 until 1937 Rutherford signed the annual certificate. The last certificate was signed on Thursday, 14 October 1937,[44] when dealing with laboratory correspondence at home in the morning. That evening he had his last direct contact with the Cavendish through a phone call from Norman Feather.[45] On the next day his illness was diagnosed, followed by surgery that evening. Tragically for the scientific world, Rutherford died four days later, on Tuesday, 19 October.

After Rutherford's death

Storage in the observatory cellar

After Rutherford's death it was natural for the Government Chemist and the DSIR to enquire who was now responsible for the radium. The DSIR wrote to J. D. Cockcroft; he passed the letter on to E. V. Appleton, who replied on 17 November 1937:[46]

> Cockroft has handed onto me your letter of 15th November, and in reply I have to say that, as acting Director of the Laboratory during the interregnum, I am responsible for the radium you have lent the laboratory.

The Government Chemist and the DSIR were then exercised to determine whether the radium was still being used and if not whether it should be recalled for other purposes. Appleton replied on 3 June 1938:[47]

> With reference to your letter of 31st May I have to tell you that the radium which you lent us is in active use in a number of ways in the Laboratory here. It is, for example, our standard neutron source, and also is used as a γ-ray source in the Mond Laboratory for heating materials at low temperatures. I therefore do hope that the loan to us can be continued.

After William Lawrence Bragg had been appointed to the Cavendish Chair, he signed the certificate for 1938 and for 1939 but along with the 1939 certificate he added a note dated 13 October:[48]

> I return the certificate. At the moment the radium, which was in solution, has been reduced to its solid and placed in a 'dungeon' at the observatory where a suitable pit was available. We hope to keep a little research going, however, and use it from time to time.

It was government policy at this time that all the hospitals, government and research institutions that possessed radium should prepare boreholes for safe storage as protection against bombing by the Luftwaffe.[49] The decision by the Cavendish staff to put their radium in the observatory cellar was consistent with the national policy, but it is not clear what proportion of the total was stored and what remained in the tower rooms.

In 1940 Bragg signed the certificate as usual. Then in the spring of 1941 discussions took place between Sidney Russ, who at that time was working at the Middlesex Hospital as a radiologist, and Norman Feather, who was then acting Director of the Cavendish in Bragg's absence on war duty. Russ put in a request to have use of the DSIR radium

because it was required for their medical programme. Feather's reply of 2 April[50] is instructive because it clearly indicates the aggregation of previously separate sources:

> I am sorry I have been so long answering your letter about radium, but the position is rather complicated. At the beginning of the war we evaporated all our radium solutions in a single concentrate. Part of it belongs to the University (about 218 milligrams element), and part to the DSIR (264 milligrams element); the second portion we have had on loan for a large number of years.
>
> It is clear that permission will have to be obtained from both the DSIR and from the University if this radium, which is now not in use, is to be loaned for the purposes you mention.

After a sequence of exchanges between Feather, the DSIR (now headed by Appleton) and the Government Chemist, the way looked clear for radium to be transferred to Russ. Then the university discovered that a Dr J. S. Mitchell,[51] an Elmore student working on radiotherapy, had need of radium and requested that the university not be asked to return the 264 mg now aggregated as part of 482 mg of element. Then it appeared that the DSIR might require the radium for Admiralty use, but subsequently an alternative source was found for this purpose, and it was agreed that the university could keep the radium for research purposes if needed. The university was all set to make an arrangement with Russ and Mitchell, because the form of the aggregate was not suitable for Mitchell's purpose, but then in July Russ revealed that he had found an alternative source. The aggregate remained in the observatory cellar and Bragg signed the 1941 certificate, followed a year later by the 1942 certificate.

Tube Alloys, 1940–46

The archives contain a very considerable number of declassified papers relating to Tube Alloys work that was conducted at the Cavendish during the war. The history of this period has been expertly told elsewhere,[52] but in brief there were at the Cavendish two major teams: a 'slow-neutron' group centred on the French team who had escaped Nazi occupation and led by Hans von Halban, and a 'fast-neutron' group led by Norman Feather. The slow-neutron group was performing research that would lead to a nuclear reactor or 'uranium boiler'; the fast-neutron group was performing research that would contribute to the development of a 'uranium bomb', including work on the use of element 94, plutonium. In the early years of the war this work was directed by the Maud committee, chaired by G. P. Thomson. After July 1941, when the Maud committee submitted its reports, this work came under the management of the Directorate of Tube Alloys within the DSIR.

As well as large quantities of uranium and moderating materials such as graphite and heavy water (deuterium oxide), large quantities of radium were needed to provide neutron sources. Feather and von Halban obtained at least a further 3 or 4 g for this purpose, which was coordinated by the National Radium Commission (NRC) and the National Physical Laboratory (NPL). This radium came down from Edinburgh, where the NRC had a centre at the Royal Infirmary, and was packaged by Johnson Matthey at Wembley.

In late 1942 the slow-neutron Tube Alloys work was transferred to the Montreal Laboratory in Canada under the directorship of von Halban. After the signing of the Quebec Agreement on 19 August 1943 there was a general exodus of Tube Alloys scientists to the USA to take part in the Manhattan Project. However, some fast-neutron Tube Alloys work was allowed to continue at the Cavendish under the direction of

Feather, whose work had been primarily concerned with the emission of neutrons from fission fragments, and methods for the detection of neutrons. It seems that the 3–4 g of radium above must have been either returned to the NRC or transferred to Canada with the slow-neutron group, because there was now a shortage of radium at the Cavendish. On 20 September 1943 Feather put in a request to the Government Chemist at the NPL to have Rutherford's aggregated radium converted from the bromide to a sulphate form and sealed in metal containers for the purpose of Tube Alloys research:[53]

> I am writing to you about the radium which this Laboratory has had on loan from the DSIR for a number of years. This radium, about 264 milligrammes element, has been in solution as bromide. At the beginning of the war it was evaporated, together with a similar solution containing about 218 milligrammes element belonging to the University. The combined sources have been in safe storage since that time.
>
> We now wish to convert all this radium into the form of sulphate and enclose it in a small metal container for use in research which is being carried on here under my direction for the Tube Alloys Department of the DSIR. I am writing therefore to enquire whether you would give permission for that fraction of the radium which is the property of your Department to be treated in this way.
>
> The chemical treatment would be carried out under Dr Grove of Mesers. Thorium Ltd. at their laboratories, and we should of course hope, as soon as the radium was no longer required for the use which we have in mind, to have it reconverted into bromide and separated again into the part which belongs to the DSIR and that which is the property of the University. I should be glad to have your answer as soon as possible, since the work we have in mind is of an urgent character.

The request was accepted and the aggregate was sent off. Thorium Ltd was at this time based in Chilcote House, Amersham, which had been purchased for them in April 1940. Work on radium had progressed steadily from that time and it had become the national centre for radium, as recorded by detailed monthly progress reports from 1940.[54] In the report by Grove for November 1944 the following was noted:

> At present we have at Amersham one of the chemists from the Cavendish Laboratory, who has come to carry out the preparation of some experimental Neutron sources .... In this connection we have prepared for the Cavendish Laboratory approximately 40 mgms of Radium which is of nearly 100% purity.

In the December 1943 report:

> A start has been made on the refining of approximately $\frac{1}{2}$ gramme of Radium owned by the Cavendish Laboratory, Cambridge, which has been stored by them during the war and is now to be put into use once more. This work will be complete during January.

In the January/February 1944 report:

> The refining of 460 mgms of Radium for the Cavendish Laboratory has now been completed, and the pure Radium Bromide [should say Sulphate] has now been delivered to Johnson Matthey and Co. for loading into metal containers.

When the request came through for the 1943 certificate Bragg had to reply in November 1943 and again in February 1944 that the radium was still with Thorium Ltd. It was not until May 1944 that the converted radium arrived back at the Cavendish from the Johnson Matthey laboratory at Wembley, where it had been sealed up in two separate containers. The two

containers had been certified by the NPL to contain 234.5 and 232 mg of radium element. According to the July 1944 report from Thorium Ltd,[55] Johnson Matthey closed their Wembley radium laboratory shortly after completing the work for the Cavendish as a result of the threat from flying bombs:

> Owing to the danger of spreading radioactive materials by blast from flying bombs Johnson Matthey have closed their Wembley radium laboratory. We have assisted them by undertaking the storage of one gramme of radium which could not be accommodated in their own bore hole.

In view of the fact that the radium was no longer in bromide form it was agreed to change the wording of the 1928 certificate as follows:

> I certify that I hold, on behalf of the DSIR, radium compound equivalent to 264 milligrames radium element, the property of H M Government, and that I am liable to return this if required in case of emergency or if no longer need by me for scientific purposes.

This was duly signed for 1943, and then again by Bragg for 1944 and 1945. From June 1944 until August 1945 Feather submitted 15 reports to the Tube Alloys directorate, eight of which were neutron-related. It is likely that Rutherford's radium was used for this purpose. Feather's last Tube Alloys report was submitted in August 1945, shortly after which he accepted an appointment at the University of Edinburgh.[56]

A research establishment had long been in the planning by a Consultative Council,[57] and on 29 October 1945 the RAF Harwell site was officially announced as the main experimental facility for the Atomic Energy Research Establishment (AERE).[58] Tube Alloys work was gradually transferred from the Montreal Laboratory, the universities and other locations. Responsibility for Tube Alloys was moved from the DSIR to the Ministry of Supply, and in 1946 as part of this reorganization Thorium Ltd was purchased by the government and named the Radiochemical Centre (RCC), intended to be the 'store-keeper' of all government-owned radium.

Decommissioning the tower rooms, 1958

Although Feather had departed in 1945 and Harwell was being established, some work with neutron sources did continue at the Cavendish, although much of this was done with the cyclotron, and it is not clear whether this involved Rutherford's radium. Nevertheless, Bragg continued to sign for the aggregated DSIR radium from 1946 up to 1951. After Bragg's resignation from the Cavendish to take up the directorship of the Royal Institution in 1953, the certificate was signed in 1954 and 1955 by E. S. Shire. At this point there is a break in the records that follow the DSIR radium until the early 1960s.

Although we lose track of the DSIR radium in the mid 1950s, there are records of decontamination work that took place in September and October 1958. The work was performed on request by the Cavendish Secretary, Mr Dibden, by a team from Harwell consisting of members of the Health Physics Group and the Industrial Chemistry Group (ICG). In the Health Physics Reports of 23 September 1958[59] by D. Fox and J. Stephenson it was noted that

> Agreement had been reached between EMR, Industrial Chemistry Group and Cambridge University that a room in the Cavendish Laboratory in which experiments involving radioactive materials had been carried out over a number of years should be decontaminated

> before re-use. The laboratory had not been used for several years and it was expected by the Cavendish Laboratory that only minor contamination would be present.

From what follows in the report, the investigators must have had a bit of a shock when they entered the tower rooms:

> The laboratory was situated at the top of a flight of narrow stairs and constituted two small rooms, both of which contained a large amount of old equipment, benches, cupboards, etc. The whole was extremely dirty, dust lying thickly over all articles; in addition, large quantities of mercury had been spilt in various places on the floor.
>
> Several large radium sources (about 480 mg) were on a wooden window sill in the inner room with very little shielding … and there were a number of smaller sources—probably radium and thorium—scattered around both rooms.
>
> Before a contamination survey was carried out it was necessary to remove all known sources and an air sample at floor level in the inner room showed 1700 dpm/m^3. The rapid decay of this sample indicated the presence of radon and daughters. A first survey of the area showed levels of activity on chairs, benches, equipment, floor and walls of up to several thousand counts per second.

All of the known sources were removed before the decontamination was carried out. A later analysis indicated ^{230}Th (25%), ^{226}Ra (35%) and ^{210}Po (40%). It is almost certain that these sources included the remainder of Rutherford's radium, after excluding the aggregated DSIR and university radium. The ^{230}Th was probably the ionium that had been purified by Boltwood in Manchester. The high ^{210}Po count is suggestive that the radium was close to equilibrium with its decay products, and by 1958 the Austrian radium would have been 50 years old. At the time of the 1958 decommissioning, George Crowe was still working at the Cavendish and was the main contact for the Harwell team, who seemed to have been astounded that Crowe 'actually' worked in the rooms:[60]

> Our main contact at the Cavendish Laboratory was Mr Crowe who had been connected with the laboratory for some 30 years and was now a laboratory steward. He appears to have spent much of his early days actually working in the rooms in question. Mr Crowe suffered radiation injuries to his fingers (one finger amputated and considerable damage to the others) some 20 years ago after working for about 6 years preparing several 'button' sources of RaD, E and F a day. For some considerable time the rooms have been used mainly as store rooms but during the past 7 or 8 years a small amount of work, particularly with polonium (probably prepared from the decay products of radon) has been carried out.
>
> Two known incidents had taken place in the rooms. One involved a spill of radium solution which, it was claimed, was properly cleaned up. In the other, a solution containing some 200 mg of radium was evaporated to dryness on a hot plate in the open laboratory. (The names of the two persons concerned in the latter incident are available.)

According to the associated ICG report of 23 September:[61]

> On Wednesday 24th September 1958, the party returned to Cavendish with appropriate active waste containers, protective clothing, breathing apparatus and decontamination kit, complete with tools for breaking laboratory benches etc. Four days work was required to strip the rooms of contents. Glassware and general trash was broken and sealed in large fibre drums. … All bottles containing solutions were packed in vermiculite and sealed in fibre drums. A three ton Bedford and 15 Cwt Ford Van were escorted on the return journey to AERE with this load of active waste on 27th September 1958.

At present no records of the arrival of the tower waste can be located in the archives, although there are records of general activities of the ICG and Health Physics Division. The Health Physics Group at this time was based in Building 364 of the Harwell campus, as it is today, and the ICG was based in Building 175 in the north part of the campus, very close to where Nirex UK and the Nuclear Decommissioning Authority (NDA) are currently located. In an interview conducted in 2010 with Lord Flowers, who in 1958 was the Scientific Director at Harwell, he could not recollect the arrival of this radium, but this may have been because at that time he was transferring to the University of Manchester.

Return of the DSIR radium to the Government Chemist, 1958–64

It is apparent that some enquiry had been made to the RCC in 1958 as to the cost of restoring the Government source to its original weight so that it could be returned because it was no longer required. The RCC reply was that it would be cheaper if the Cavendish were asked to pay for the difference in weight, about 26 mg, between the original amount and the divided aggregate. This arrangement seemed agreeable to all sides. At a meeting in March 1962 the Cavendish changed its mind:[62]

> Since Mr Dibden wrote to you on 13th June 1958, there have been substantial changes in the research activities in these laboratories, with the result that I feel it would be foolish for us to return the radium source which has been so kindly loaned to us over such a long period.
>
> I should like therefore, to formally request that the Government Chemist should allow us to retain this source for a further period of time, during which time we would expect to satisfy him that we still hold the source in the amount granted to us. I fully realise that should we be allowed to do this, we should be liable to return the source in case of emergency needs.

The DSIR position was, however, that as the original Treasury Pool was to be disbanded, now that the RCC had taken the role as the national 'store-keeper', that no further loans or extensions of loans could be anticipated. Any current holders should be asked to either pay for it or return it to the Government Chemist. The Government Chemist took a different view in a communication to the DSIR in June 1962:[63]

> I am happy with the present arrangement that the Ra stays on loan to the Cavendish Lab. If we ask them to buy it, they might well refuse to do so and then we would be in the embarrassing position of having to provide proper (and expensive) storage for it. By all means examine the position with the Cav. Lab., but please bear this contingency in mind.

The DSIR then sought to obtain from the RCC a price for the radium on loan and settled on £4 per milligram, giving a total price of £1060. This offer was put to the Cavendish on 25 July 1962. On 13 August Dr Nicol, the Secretary, informed the DSIR:[64]

> After making full enquiries about our likely future needs of such a source, I find it would in fact not be advisable for the Department to purchase and I am therefore writing to confirm that we should return the source which is now estimated at 260.5 milligrams, letting you have cash payment in respect of the difference between this value and the original value of the source ...

After an enquiry was made to determine the exact strength of the two sources held by the Cavendish, Nicol then made an offer to hand over both sources.[65]

Table 1. Rutherford's stock of radium, 1900–1937.
(Amounts are expressed in equivalent bromide, if known.)

location and dates	form	origin	date	amount (mg)
Montreal, 1898–1907	chloride	Estler & Geitel	1900	?
	chloride	P. de Haen, Hanover	1901	?
	chloride	Paris?	1902	?
	bromide	Giesel, Buchler & Co.	1903	80–160
Manchester, 1907–19	bromide	Armbrechts, Nelson & Co.	1903	20
	bromide	Schuster?	1903?	40–50
	bromide (standard)	?	1907	3.69
	chloride	Vienna	1908	455
	chloride (standard)	Vienna	1912	7
Cambridge, 1919–37	chloride?	Vienna	1921	10
	bromide/sulphate	MRC/DSIR	1921/27	493
	bromide/sulphate	University?	1920s?	408
	bromide?	Union Minière	1936?	100
	chloride with Be	Chemical Services Co.	1937	98.75

> We are prepared to return both the sources designated D.S.C.1 of 234.5 milligrams and D.S.C.2 of 232 milligrams, in settlement for the original 264 milligrams loaned to Lord Rutherford from the Treasury Pool in 1928. ... If this suggestion is not acceptable, we are quite prepared to return the larger ... and make a cash settlement to cover the difference between the source which was originally loaned and the ... value of DSC 1.

On 9 November the DSIR turned down the offer to take both sources but confirmed that the Cavendish should return the source D.S.C.1, along with £110 payment for the difference from the original source, to be delivered to the Laboratory of the Government Chemist (LGC), then at 13 Clement's Inn Passage. The payment of £110 was duly made on 20 November 1962, but a complication arose as the LGC was shortly to move to new premises at Cornwall House, Stamford Street. The Cavendish were requested to keep D.S.C.1 in storage until 1963 when the move had been completed. The records break off again at this point, but it must be presumed that D.S.C.1 was transferred to Cornwall House some time shortly after January 1964, when the move actually took place.

Summary and discussion

The history of Rutherford's radium closely parallels the history of radioactivity, nuclear physics, the race for the bomb and the development of atomic energy establishments. Starting with the discovery of radium in 1898, the use of radium changed from being a substance of intrinsic interest for studies on the nature of radioactivity, to being a primary source of α-particles for nuclear experiments, to being primarily one of sources of neutrons when combined with beryllium. By the time of his death, Rutherford had accumulated about 1 g of radium element, which included both the Manchester and Cambridge sources (see table 1).

In 1939, at the outbreak of World War II, about half of Rutherford's accumulated stock was evaporated, aggregated and put into storage in the observatory. In 1943 this portion was sent off for reprocessing at Amersham and used for Tube Alloys work for the rest of the war.

After the war we can keep track of at least the portion owned by the DSIR up until 1964. There are no records of the other half until 1958, when the team from the UK AERE investigated the tower rooms and removed all the sources to Harwell. It seems likely that these included Rutherford's old Austrian radium from 1908, which had remained in the tower rooms for almost four decades from 1919, including throughout World War II, and had been practically forgotten about after the war, and probably before the war. Samuel Devons's recollection was that by 1935 he had almost sole use of it because there was no longer any interest in the old radium. In his 1941 letter to Russ, Feather talks about 'all our radium' when referring to the aggregated and evaporated radium stored in the observatory. When the Harwell team entered the tower rooms in 1958 they had been led to believe that only 'minor contamination' would be found.

It is of interest that some 20 years after the 1958 decommissioning, a further remediation was required in 1977 when the Department of Physics had moved to the new site. This caused some interest in the media at the time (not unlike the interest shown by the Manchester remediation). The *Cambridge Evening News* reported comments on this work by Norman Feather, who at that time was an emeritus professor at Edinburgh:[66]

> It is nonsense to say any experiments were carried out in the room. No experiments took place there. The room was only used for the preparation of radiation sources .... There was a fair amount of contamination, it was almost inevitable with what was known about the subject in those days, but the room was not locked when Rutherford died. I was responsible for the room and it was still being used when I left Cambridge in 1945, but no one spent more time than they had to inside the room.

We may only speculate on the fate of the radium removed to Harwell, but there are three possibilities: storage, dumping and reprocessing. In 1958, Building 462 off Rutherford Avenue in the western area of Harwell was the main site for storage and waste management.[67] This area is still used today for waste storage and it is possible that Rutherford's old radium is sealed up in a bunker somewhere under Rutherford Avenue. In the late 1950s there was also a programme of dumping waste in the Atlantic Ocean; it is possible that it lies incarcerated deep in the mid-Atlantic trench.

Although storage or dumping is possible, it is more likely that the radium was dispatched to Amersham for reprocessing. There was in the late 1950s a huge demand for radium, primarily as radium–beryllium neutron sources. For example, the Zeus prototype fast breeder reactor at Harwell required 10 g of radium.[68] There was also a demand from Aldermaston for the production of polonium to be used as bomb neutron initiators, although some of this demand could be met from the irradiation of bismuth at Windscale. Between December 1958 and September 1959 the RCC had on order no less than 55 g.[69] It would have seemed profligate indeed to have thrown away a good half-gram of the stuff in 1958. Thus it is quite likely that the 1908 Austrian radium was used to make a neutron source, either for a reactor or for a bomb.

We can be reasonably sure that the source D.S.C.1, which was exchanged for the original MRC radium, was delivered to the LGC in 1964. The Radiochemical Department of the LGC was based on the fourth floor of Cornwall House,[70] and presumably D.S.C.1 was stored there before being sent elsewhere, possibly to the RCC at Amersham. The RCC was privatized in 1982 and currently trades as GE Healthcare Ltd. The LGC moved again in the 1980s to Teddington, the same location as the NPL, and was privatized in 1996, trading now as LGC UK. The fourth floor of Cornwall House, renamed the

Wilkins–Franklin Building, is now occupied by King's College Faculty of Life Science. In 1964 source D.S.C.2 was still owned by the Cavendish and in their keeping. Interest in nuclear physics continued at the Cavendish during the tenure of Otto Frisch after 1948, although he himself did not publish any papers that made use of radium–beryllium neutron sources. It seems unlikely, however, that the source would have been kept beyond the move to West Cambridge in 1974, and it was presumably sent on to either Amersham or Harwell.

Epilogue

Ultimately all portions of Rutherford's radium are probably reunited now as part of the UK Radioactive Waste Inventory (UKRWI), and currently awaiting 'geological disposal'.[71] Both GE Healthcare Ltd, Amersham (site 1A), and the NDA, Research Sites Restoration Ltd, Harwell (site 5C), are listed as waste custodians. Further research will be required to identify which of their individual waste streams may contain the portions (see UKRWI Annex 4).

Acknowledgements

The primary sources for the present work are documents held in the National Archives and in the libraries of Cambridge, Manchester and McGill universities. I am grateful to the staff within these institutions for their help and in making the documents available.

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