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Standards and Measurements of Ionizing Radiations in the 20th Century at the National Bureau of Standards/National Institute of Standards and Technology 1901 – 1921

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Abstract

This report describes the development of standards for radioactivity and x rays in the US during the first two decades of the 20th century. The first standards for radium were pioneered by chemists in industry and academia. The US national standards were established at the National Institute of Standards and Technology on receipt of a radium standard from Marie Curie and the International Radium Standards Commission in 1913. During World War I, NIST began development of standards for self-luminescent dials and watches containing radium paint, and standards for x-rays to support the US industry for protective materials against x rays and radium. Marie Curie's 1921 visit to NIST to receive a gram of radium funded by donations from the women of America is described.

Keywords

Alpha particles; emanation; gamma rays; ionization chambers; luminescence; Marie Curie; mesothorium; radioactivity; radium; standards

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Preface

This is the first complete account of the development of standards for radioactivity and x rays in the US in the first two decades of the 20th century. This may seem unusual given the vast literature devoted to the discoveries of x rays and radioactivity. There are two main reasons for this oversight. First the federal institute known as the U.S. Bureau of Standards was not founded until 1901, and the work at that institute related to x rays and radioactivity did not begin until the second decade. Second, the principal investigators at "the Bureau" as it was known during the period 1910 -1921, Noah Ernest Dorsey, Edward Bennett Rosa and Samuel Wesley Stratton, were not engaged in this work after 1921. None of the three, nor the historian Rex Cochrane in his book *Measures for Progress* captured the history of radiation standards in the early years. The next generation of physicists at the Bureau including Leon Francis Curtiss and Lauriston Sale Taylor came in the mid-1920s and their historical accounts of the standards were mainly devoted to later work.

Absent contemporary reports on the history of standards from the staff at the Bureau, the present account draws on three main sources. The first of these are the NIST archives which take the form of digital archives, photographs and records, that have been scanned by the NIST Library, as well as folders of papers specific to both topics and individuals. The files on N.E. Dorsey are particularly valuable. The second category is the staff records from 1913 -1921. The names of sections, groups and divisions changed many times but the mandate for work of the Radioactivity and Dosimetry Groups has changed little over the past 100 years. These records, miscellaneous paper files of the radiation physics divisions over 90 years, are slightly contaminated with radium-226. Many of these have been photographed, photocopied and scanned and this material has become a part of the formal NIST archives. A complete set of the contaminated records are now safely stored such that they are available to future researchers. The third category is published reports from the time by academic scientists and those at the U.S. Bureau of Mines. A particularly valuable resource in this last category are letters in the Bertram Boltwood archives at Yale University. Boltwood was the official US representative to the International Radium Standards Commission and the Yale archives contain 18 letters he exchanged with N.E. Dorsey at the Bureau between 1913 and 1921.

Acknowledgments

We would like to express our appreciation to several colleagues who have kindly provided numerous leads and specific references; first and foremost, Ronald Collé in the NIST Radioactivity Group. We are much indebted to a group of colleagues who share our enthusiasm for the history of radium including Joel Lubenau, Edward Landa, David Allard and Paul Frame. We would like to express thanks also to Laura Atwell and David Shauer of the National Council on Radiation Protection and Measurements, to Aimee Slaughter and Kristen Frederick-Frost, and to our colleagues in the Radioactivity Group Jeffrey Cessna and Lisa Karam. And a special thanks to Cheryl Levey in the NIST Standards Coordination Office who has provided outstanding assistance in preparation of numerous drafts of the manuscript.

1. Introduction

Following Henri Becquerel's discovery of the phenomenon of radioactivity from uranium containing materials in 1896 and Marie and Pierre Curie's success in separating radioisotopes of radium and polonium, the investigation of radioactivity became one of the leading threads of science for the next decade. The rapid advances in science came on two fronts, first in chemistry and then in physics. Marie Curie was able to identify several minerals containing various amounts of uranium and thorium, and working with 8 tons of pitchblende from St. Joachimsthal, Bohemia (present Czech Republic) she was able to separate weighable amounts (decigram quantities) of radium-226 (Curie, 1903). The metrology of radium depended on chemically pure separations of radium halides (chlorides and bromides) containing only trace amounts of barium.

Ernest Rutherford, a native New Zealander, led an extraordinary research effort in the physics of radioactivity in Montreal, Canada and Manchester, United Kingdom. Rutherford's first book *Radioactivity* was published in 1904. With his colleagues and guest scientists at McGill University in Montreal, he carried out pioneering research on the nature of radioactive decay and the use of alpha particles from radioactive sources as probes of the atomic nucleus (Eve, 1939, Brown, 1997). The pace of Rutherford's work accelerated when he took a post in physics at Manchester University in 1907.

For their early work, the Curies and Becquerel were awarded the Nobel Prize in physics in 1903, while Rutherford was awarded the Nobel Prize in chemistry in 1908 *"for his investigations into the disintegration of the elements, and the chemistry of radioactive substances"*. Marie Curie would be honored for her work in chemistry as well with the Nobel Prize in chemistry in 1911. The history of radioactivity is covered in many reviews, including first person accounts from Marie Curie (Curie, 1903, 1912, 1923) and Ernest Rutherford (Rutherford, 1904, 1912; Rutherford et al., 1930). The role in radium metrology played by the National Bureau of Standards (NBS)[1] in the first decade of the century was slight. In 1901, initial staffing at NBS included one man identified as a physicist, Edward Bennett Rosa, who came to NBS from the physics department at Johns Hopkins University. In 1904, Rosa observed an NBS staff member, Llewelyn G. Hoxton, reading Rutherford's book *Radioactivity*. He asked to borrow the book and returned it the following day (Cochrane, 1966; Eisenhower, 1980; Coursey, 2018). It was too soon for NBS to undertake development of standards for radioactivity.[2] Rosa and his colleague Noah Ernest Dorsey were engaged in research that would soon lead to the world's most accurate measurement of the speed of light (Rosa and Dorsey, 1907).

In the first decade of the new century, there were three closely related drivers for the development of measurement standards for radioactivity: science, medicine, and industry. The scientists required agreement on both terminology and reproducible radioactivity measurement methods that would allow them to compare results that would be acceptable for publication in scientific literature (Randall, 1912; Badash, 1979). When radium was found to offer potential in the treatment of cancer, physicians needed standards: for their radium sources, for instruments to

[1] The U.S. Bureau of Standards was founded in 1901. In 1934 the name was changed to the National Bureau of Standards, and in 1987 to the National Institute of Standards and Technology. In this work, the institute will be referred to as NBS, except in direct quotes from the period.
[2] During the early years, NBS would host the April annual meeting of the American Physical Society (APS) in Washington, DC. While Rutherford was in Montreal, he would come to Washington for the APS meetings. He first reported meeting the NBS physicists, Samuel Wesley Stratton, Director and Edward Bennett Rosa, Physicist, in April 1906. He probably also met Noah Ernest Dorsey. Dorsey also came from the Johns Hopkins University physics department, but he spent a year at the Department of Agriculture, before joining NBS (NIST Archives, 2023). Dorsey was 12 years junior to Rosa, his direct supervisor.

measure the sources, and yardsticks for accessing the efficacy of treatments (e.g., Abbe, 1904). By 1905 it was evident that the commercial value of radium for medical applications would support an international market for the separation of radium from ores, and production of materials for medical purposes (Landa, 1982). The three leaders of the international community were the French, English and German speakers Marie Curie, Ernest Rutherford and Stefan Meyer. By the end of the decade, they would each head a major institute for the study of radioactivity.

The interrelationships of all the entrepreneurial researchers, physicians and industrialists are captured here in a series of tables. Table 1 lists the principal mining sites for uranium bearing ores (Landa, 1982). Table 2 lists 18 international chemists who had some role in the investigations of radioactivity that led to a foundation for physical measurement standards. Four of these received a Nobel Prize for chemistry (Ramsey, Richards, Soddy and Hahn). Table 3 lists 15 academic physicists who led the investigations in early nuclear science and developed the instruments for quantitative measurements of radioactive decay products. Five of the physicists would also win Nobel Prizes (Marie and Pierre Curie, Rutherford, Chadwick and Hess). Stefan Meyer did not win a Nobel Prize, but he holds the record for the person who recommended the most winners of the Physics Prize (13).[3] Many of these 33 scientists formed collaborations with physicians in nearby hospitals. A select list of some of the first and most active medical specialists who began therapies with radium are shown in Table 4. Most of these men were established physicians in their 50s who saw the potential for radium to augment their traditional therapies. Table 5 lists several of the companies that were formed to prepare and sell radium products. A complete account of the mining industry for uranium and radium is given in Robison, 2015 (Robison, 2015). Many of these enterprises were unsuccessful and folded after a few years. The more successful ventures were subsidiaries of corporations that already had facilities and trained industrial chemists and engineers to produce and market products, such as uranium (St. Joachimsthal), vanadium (Pittsburgh), thorium (Gloucester), or organic and specialty chemicals (Berlin and Braunschweig).

Table 1. Principal mining sites for ores containing uranium/radium in early 20th century (Frondel, 1958, Landa, 1982, Robison, 2015). Pitchblende is used to define a common variety of uraninite (Frondel, 1958).

Site	**Ore**	**Chemical formulas**
St. Joachimsthal, (Jáchymov) Bohemia	Pitchblende (Uraninite)	Mixed Uranium oxides: UO_2 and U_3O_8
Paradise Valley, Colorado/Utah	Carnotite	$K_2(UO_2)_2(VO_4)_2 \cdot 3H_2O$
Urgeiriça and Canas de Senhorim (Viseu), Portugal	Chalcolite	Copper-Uranium phosphate
Autun, France	Autunite	Calcium-Uranium phosphate
St. Austell, Cornwall	Pitchblende (Uraninite)	Mixed Uranium oxides: UO_2 and U_3O_8
Haut Katanga, Belgian Congo (Democratic Republic of the Congo)	Pitchblende (Uraninite)	Mixed Uranium oxides: UO_2 and U_3O_8
Great Bear Lake, Canada	Pitchblende (Uraninite)	Mixed Uranium oxides: UO_2 and U_3O_8

[3] Additional details on Stefan Meyer and the Institute of Radium Research are available in Meyer, 1949, 1950 and Reiter, 2001, 2011.

Table 2. Selected list of chemists engaged in radium research in early 20th century

Chemists	Dates	Home Institute	Visiting status	References
Bertram Borden Boltwood	1870 - 1927	Yale University	Manchester University	Kovarik, 1929, Badash, 1969, 1979
Otto Brill	1881 - 1954	University of Vienna	Standard Chemical Company, Pittsburgh With William Ramsey in London With Pierre and Marie Curie in Paris	Lubenau and Landa, 2019
André-Louis Debierne	1874 - 1949	Curie Institute Paris		
Ellen Gleditsch	1879 - 1968	University of Oslo	Curie Institute Yale University	Badash, 1979
Otto Hahn	1879 - 1968	Kaiser Wilhelm Institute for Chemistry, Berlin	McGill University, Montreal & University College London	Hoffman, 2001; Sgantzos et al., 2014
William Francis Hillebrand	1853 - 1925	US Geological Survey & US Bureau of Standards	Ph.D. University of Heidelberg	Clarke, 1925
Otto Hönigschmid	1878 - 1945	Prague Polytechnical University	Institute for Radium Research Vienna	Meyer, 1945
Glenn Donald Kammer	1888 - 1927	Standard Chemical Company, Pittsburgh	University of Pittsburgh	Silverman, 1927, 1950; Lubenau and Landa, 2019
Henry Titus Koenig	1891 - 1934	Standard Chemical Company, Pittsburgh	University of Missouri Curie Institute, Sorbonne	Silverman, 1934, 1950
Samuel Colville Lind	1879 - 1965	University of Michigan U.S. Bureau of Mines	Curie Institute	Laidler, 1998
Herbert Newby McCoy	1870 - 1945	University of Chicago		Badash, 1979
Richard Bishop Moore	1871 - 1931	U.S. Bureau of Mines	University of London	Rentetzi, 2008
William Ramsay	1852 - 1916	University College London		Soddy and Worthington, 1916
Theodore W. Richards	1868 - 1928	Harvard University	Göttingen University	Badash, 1979 (p 203)
Herman Schlundt	1869 - 1937	University of Missouri	University of Leipzig	Gibbons, 2013
Sabin Von Sochocky	1883 - 1928	US Radium Corporation, New Jersey	Sorbonne, Prague, Vienna	Wall, 1976; Coursey, 2021
Frederick Soddy	1877 - 1956	Glasgow University	McGill University, University College London	Howorth, 1958
Charles Herman Viol	1886 - 1928	Standard Chemical Company, Pittsburgh	University of Pittsburgh	Lubenau and Landa, 2019

Table 3. Selected list of physicists engaged with measurements and standards of radium

Physicists	Dates	Home Institute	Visiting status	References
James Chadwick	1891 - 1974	Manchester University	Physikalisch-Technische Reichsanstalt	Brown, 1997
Marie Curie	1867 - 1934	Sorbonne		Curie, 1961
Pierre Curie	1850 - 1906	Sorbonne		Curie, 1961
Noah Ernest Dorsey	1873 - 1959	U.S. Bureau of Standards		NIST archives, 2023
William Duane	1872 - 1935	University of Colorado	Curie Institute	Bridgeman, 1936, Brucer, 1993
Julius Elster	1881 - 1920	Gymnasium Wolfenbüttel		Fricke, 1992
Arthur Eve	1862 - 1948	McGill University		Foster, 1949
Gioacchino Failla	1891 - 1961	Memorial Hospital	Curie Institute	Marinelli, 1962; Atomic Heritage, 2023
Johannes Wilhelm (Hans) Geiger	1882 - 1945	Physikalisch-Technische Reichsanstalt	Manchester University	Shampo et al., 2011
Hans Geitel	1882 - 1945	Gymnasium Wolfenbüttel		Fricke, 1992
Victor Francis Hess	1883 - 1964	Institute for Radium Research	US Radium Corporation, New Jersey, US Bureau Mines	Lubenau and Landa, 2019
George William Clarkson Kaye	1880 - 1941	UK National Physical Laboratory	Cambridge University	Griffiths, 1941
Stefan Meyer	1872 - 1949	Institute for Radium Research		Reiter, 2001, 2011
Ernest Rutherford	1871 - 1937	Manchester University	Cambridge University	Eve, 1939
Egon von Schweidler	1873 - 1948	University of Innsbruck	Institute for Radium Research	Paneth, 1949

Table 4. Selected list of physicians engaged in radium therapy in early 20th century.

Physician	Dates	Institute	Sources of Radium for Therapy	References
Robert Abbe	1851 - 1928	Roosevelt Hospital, New York, NY	Curie Institute Paris & Institute for Radium Research Vienna	Slaughter, 2013
Curtis Field Burnam	1877 - 1947	Kelly Clinic, Johns Hopkins Hospitals, Baltimore, MD	Multiple suppliers	Burnam, 1936; Evening Sun, 1947
Henri-Alexandre Danlos	1844 - 1912	Hospital St. Louis, Paris	Curie Institute Paris	Badash,1979 (p.136)
Henry Harrington Janeway	1873 - 1921	Memorial Hospital, NY	National Radium Institute	Robison, 2015
Howard Atwood Kelly	1858 - 1943	Kelly Clinic, Johns Hopkins Hospitals, Baltimore, MD	Institute for Radium Research, Vienna & Standard Chemical Company, Pittsburgh, PA	Badash, 1979, Slaughter, 2013, Burnam, 1936

Table 5. Selected list of companies extracting radium in early 20th century

Company	Location	Staff and consulting chemists	Reference
Auergesellschaft	Berlin, Germany	Siegfried Merzbacher	Joe Dunthorne, 2020
British Radium Corporation	London, United Kingdom	William Ramsay	Landa, 1982
Buchler Corporation	Braunschweig, Germany	Friedrich Giesel, Julius Elster, Hans Geitel	Robinson, 2015
Carnotite Reduction Company	Chicago, Illinois	Herbert Newby McCoy	Badash, 1979
Eugen de Haen & Company	Hamburg, Germany	Friedrich Giesel	Badash, 1979
National Radium Institute	Denver, Colorado	Charles Parsons, Richard Moore, Samuel Lind, Herman Schlundt	Parsons et al. 1915, Landa, 1987,
Radium Company of Colorado	Denver, Colorado	Henry Koenig, Willy A. Schlesinger	Landa 1982, Robison, 2015
Rare Metals Reduction Company	Buffalo, New York	Stephen T. Lockwood	Parsons, 1913, Lockwood, 1921
Standard Chemical Company	Pittsburgh, Pennsylvania	Otto Brill, Charles Viol, Glenn Kammer, Henry Koenig	Lubenau and Landa, 2019
Société Centrale de Produits Chimiques	Paris, France	André-Louis Debierne	Lubenau et al. 2020; Blanc 2009
Sociétié Minière de Radium de St. Joachimsthal	St. Joachimsthal, Bohemia	Stephan Meyer, Victor Hess, Otto Hönigschmid	
Union Minière de Haut Katanga	Oolen, Belgium	M. Clérin, M. Boulanger, Henry Koenig	Silverman, 1934, Mukanda, 1967, Landa, 1982
Welsbach Thorium Mantle Company (radium-228)	Gloucester, New Jersey	Harlan S. Miner, Howard Barker, Herman Schlundt	Badash, 1979 (p.80), Gibbons, 2013

2. Early US Standards 1900 - 1910

An ideal standard would be a mass of pure radium or a stable radium compound such as radium chloride. But such a standard was not realizable for several reasons. First, a pure source of the elemental radium metal was not produced until Marie Curie and André Debierne in 1910 succeeded in preparing a small amount by electrolysis of radium chloride (Curie & Debierne, 1910). Second, there were only a few chemists, including Marie Curie and Otto Hönigschmid, with access to large enough amounts of raw material to separate and weigh significant quantities of anhydrous radium chloride. They both used purified samples of radium chloride to determine the atomic weight of radium (Hönigschmid, 1911, 1934; Curie, 1912). And finally, mass standards were out of the question for shorter-lived radioactive isotopes of polonium and bismuth that could not be separated in weighable amounts.

Earlier investigators who had access to only small amounts of radioactivity turned to other materials and experimental methods. The three chief measurement methods used by the chemists, physicists and mineralogists were based on emanation (radon-220 and radon-222), alpha-particle emission rate, and gamma-ray emission rate (Randall, 1912; Parson et al., 1915).

NBS was not engaged in radioactivity work during that first decade. The best accounts of the early standards work are provided by the 1912 publication by Merle Randall in the Transactions of the American Electrochemical Society (Randall, 1912) and the critical analysis of the early work by the historian Lawrence Badash in his 1979 book **Radioactivity in America** (Badash, 1979). Randall, a physical chemist born in Missouri, is best known for his classic textbook **Thermodynamics** with G.N. Lewis (Lewis and Randall, 1923). Randall received his B.A. and M.A. degrees at the University of Missouri with his research under the direction of Professor Herman Schlundt. For his M.A. thesis Randall reviewed all the work on standards of radioactivity up until 1908. (He left Missouri in 1909 to pursue a Ph.D. degree at the Massachusetts Institute of Technology.) Randall chose as the title for his research paper **The Boltwood Standard of Radioactivity** (Randall, 1912). In his introduction, he acknowledges the work of several investigators in the US and Europe as shown here in Table 6.

Table 6. Early standards of uranium compounds (Randall, 1912)

Investigators	Material(s)	References
R.J. Strutt	Crystal uranyl nitrate	Strutt, 1903
J. Elster and H. Geitel	Uranyl potassium sulfate	Elster and Geitel, 1904
G.A. Blanc	Uranium nitrate	Blanc, 1905
S. Allen	Uranium oxide	Allan, 1903
H.N. McCoy	Urano-uranic acid	McCoy and Ashman, 1908
B.B. Boltwood	Uranosa-uranic acid – thin film	Boltwood, 1908

Not all these "standards" were easy to replicate in other laboratories. Randall and later Badash (Badash, 1979) considered the most valuable of these to be the two proposed by the US chemists Bertram B. Boltwood at Yale University (Boltwood, 1908) and Herbert N. McCoy at the University of Chicago (McCoy and Ashman, 1908). Boltwood and McCoy were independently carrying out research to identify the radioactive decay products in the uranium and thorium series. They were both using leaf electroscopes to measure ionization from uranium sources and to measure separated radium preparations, allowing for ingrowth of the radon progeny. What Randall referred to as the "Boltwood Standard" was an emanation standard.

2.1. Boltwood emanation standard

The emanation methods took advantage of the strong signal in an electroscope from the emissions of radon and its progeny. The noble gas radon could be collected from mineral samples as well as from radium preparations and solutions in different stages of purification (Boltwood, 1904; Hess, 1913; Lind, 1915).[4] When the gas was introduced into a gold-leaf electroscope, the ionization produced by the alpha and beta emissions of the radon and progenies caused a deflection in the leaf with a drift rate that was proportional to the activity of the gas. Under ideal conditions one could detect radium to a limit of 17 picograms (Badash, 1979 p.87).

Badash makes the case that the earliest critical work on standards came from Rutherford's collaborations with Boltwood and several other chemists (Badash, 1969, 1979). Physicists often

[4] Emanation was later understood to be the noble gas radon, which had three different isotopes associated with the three natural decay chains of uranium-238, thorium-232 and uranium-235 (the actinium series). The three isotopes were named radon (radon-222; $T_{1/2}$ 3.8 days), thoron (radon-220; $T_{1/2}$ 52 s) and actinon (radon-219; $T_{1/2}$ 3.92 s).

use a quote attributed to Rutherford "In science there is only physics; everything else is stamp collecting" (e.g., Randall, 2015). This may be intended to be humorous, but it does not reflect the close collaboration and friendship that Rutherford had with three of the leading chemists of his time: Frederick Soddy from Oxford University, Bertram Boltwood from Yale, and Otto Hahn from the Kaiser Wilhelm Institute in Berlin. Soddy, the English chemist, spent a year at McGill University with Rutherford in 1902 -1903. The Rutherford and Soddy work on the Radioactive Decay Law led to Rutherford's Nobel Prize in chemistry in 1908 (Rutherford and Soddy, 1902). The Nobel citation mentions the crucial collaboration on chemistry with Soddy, who would later receive his own Nobel Prize in chemistry for his role in discovery of isotopes. Soddy's work with William Ramsay in London confirmed that helium-4 atoms in minerals were alpha particles from radium decay (Ramsay and Soddy, 1903). Following up on the Rutherford-Soddy discoveries, Hahn and Boltwood would carry out detailed chemical investigations on the decay products of uranium-238 and thorium-232. Working with Ramsay in London, Hahn in 1904 discovered the isotope radium-228 in the decay chain of thorium-232. Hahn called this "mesothorium" and it was soon marketed as "German Radium" (Sgantzos et al., 2014). In 1905 Hahn moved to McGill to continue his studies with Rutherford. Boltwood had been investigating the radium content of different minerals as early as 1898 or 1899 (Badash, Letters p. 25, 1969).

The genesis of "US standards for radioactivity" came from Rutherford's visit to Yale University in April 1904 to deliver the Silliman Lectures. He met Boltwood when he gave his lecture on the current status of work in the new field of radioactivity. Boltwood wrote to him on May 11, 1904. *"Dear Prof. Rutherford, You will perhaps recall meeting me when you were here in New Haven and talking over the question of the determination of the relative proportions of uranium and radium in natural minerals, a subject on which I was at the time working* (Badash, 1969)." And, of course Rutherford remembered their conversation as the radium to uranium ratio was one of the most critical parameters in the new field of radioactivity. Their meeting and this first exchange of letters would lead to correspondence over two decades that record their personal reflections and opinions during the critical period of the development of the field of radioactivity. The historian Badash collected and edited their correspondence and added valuable endnotes from the academic literature and university archives (Badash, 1968, 1969, 1979, 1989). Boltwood's studies in the period 1899 – 1904 parallel those of Marie Curie in that he investigated the radioactivity content of a series of minerals. Boltwood had a private practice as a consultant to mining interests that provided him access to a large number of ore samples from around the world. Marie Curie used an apparatus connected to an electrometer to measure the air ionization from a thin sheet of powdered minerals (Curie, 1903). She tested many of the same minerals as Boltwood and measured the ionization from samples, with currents of the order of picoamps. Boltwood's studies transmitted to Rutherford in May 1904 went further. He made great efforts to completely dissolve the minerals in acid to allow him to collect all the emanation in the apparatus shown in Figure 1.

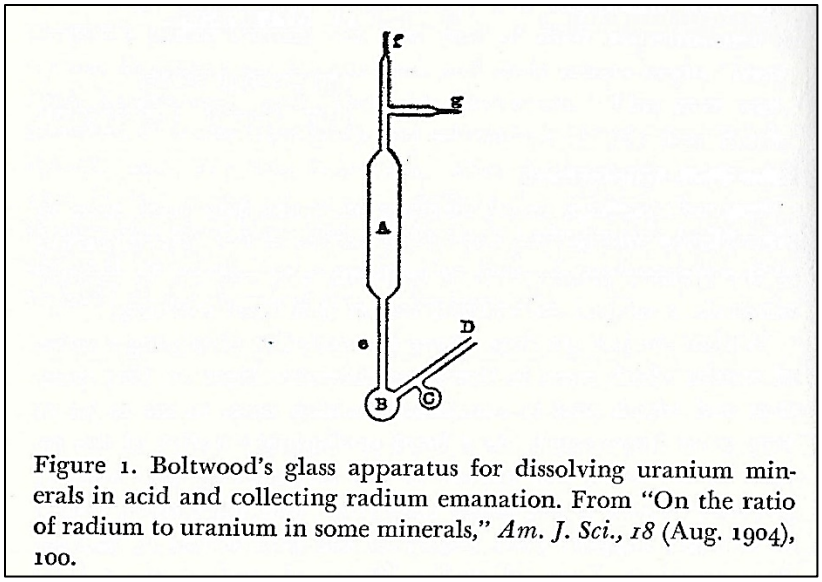


Figure 1. Boltwood's glass apparatus for dissolving uranium minerals in acid and collecting radium emanation. From "On the ratio of radium to uranium in some minerals," *Am. J. Sci., 18* (Aug. 1904), 100.

Figure 1. Boltwood's glass apparatus for capture of radon gas in Am. J. Sci. **18** (1904a).

After allowing sufficient time for radon to come to equilibrium in solution, the gas was collected and sealed in Bulb A. The radon was then introduced into a sealed chamber as part of the gold-leaf electroscope shown in Figure 2. Boltwood constructed this instrument based on a design by C.T.R. Wilson at the Cavendish Laboratory in Cambridge (Badash, 1979 p.77). The drift time for the gold leaf, in divisions per minute, provided the relative strength of the radioactive source. This was a major improvement over the use of the powdered samples that were subject to alpha-particle attenuation in minerals of different composition. Boltwood was completely measuring the radioactivity of radium-226 progenies radon-222 and its short-lived progeny.

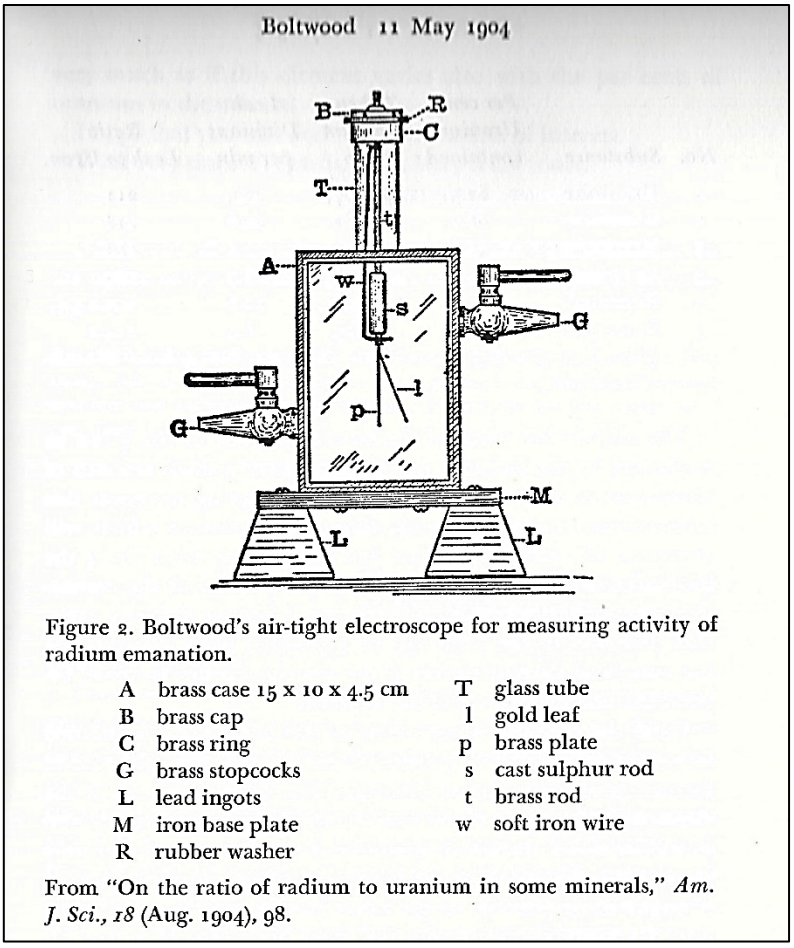


Figure 2. Boltwood's air-tight electroscope for measuring activity of radium emanation.

A	brass case 15 x 10 x 4.5 cm	T	glass tube
B	brass cap	l	gold leaf
C	brass ring	p	brass plate
G	brass stopcocks	s	cast sulphur rod
L	lead ingots	t	brass rod
M	iron base plate	w	soft iron wire
R	rubber washer		

From "On the ratio of radium to uranium in some minerals," *Am. J. Sci., 18* (Aug. 1904), 98.

Figure 2. Boltwood emanation electroscope in Am. J. Science, 18, Aug. (1904a)

Boltwood described his "standard" for radioactivity in a later issue of American Journal of Science that same year (Boltwood 1904b).

> "The standard here suggested and employed is the quantity of radium emanation set free when a known weight of uranium in the form of a natural mineral is dissolved in a suitable reagent. The mineral that has been used is a pure uraninite from North Carolina. It was dissolved in aqua-regia, the solution diluted with water and the gas removed by boiling."
>
> "The rate of the leak as determined at three hours was taken as the basis of calculation, and was considered as equal to the activity of the radium emanation associated with 0.0100 gram uranium. The leak was equal to 1.76 division per minute. A fall in the gold leaf equal to 0.001 division per minute was therefore equivalent to 5.68×10^{-6} grams uranium."

The subsequent collaboration with Rutherford on the ratio of radium to uranium was probably the chief reason that Boltwood would be selected to represent the US on the first International Radium Standards Commission (to be discussed in detail later). Boltwood and Rutherford set out over the next two years to establish the Ra/U value. This account draws heavily on the books by Badash (Badash, 1969, 1979). Their progress was interrupted briefly by the *war of standards*, which was probably more like a "battle" than a "war". Boltwood, an accomplished analytical

chemist, could accurately measure the uranium content of mineral samples. With his gas-tight electroscope he could accurately measure the radon-222 associated with each sample, and accounting for decay he could compute the equilibrium content of radium-226 in the mineral. But, for a reference measurement, he required a reference sample of radium-226.

Rutherford's collaborator in the McGill physics department, Arthur Eve, prepared a *radium standard* solution of pure radium bromide. Rutherford provided Boltwood in 1905 with an aliquot from his McGill standard solution to use in calibrating the electroscope at Yale. Sixty days later (allowing more than sufficient time for equilibrium concentration of radon-222) Boltwood boiled the solution and used the radon-222 to calibrate his electroscope. For the uranium measurement he used a North Carolina uraninite ore. In 1905 they found *"the quantity of radium associated with one gram of uranium in a radio-active material is equal to approximately 7.4 x 10^{-7} gram."* (Rutherford and Boltwood, 1905). By January of 1906 Rutherford and Eve began to question some of Boltwood's measurements of radioactivity in uraninite samples. This led to a brief *war of standards* which was resolved when Boltwood cautioned Rutherford that their radium standard solutions should be made acidic (with hydrochloric acid) to prevent the radium from sticking to the walls of the glass containers. In April of 1906 Rutherford wrote *"I am glad to hear that the "war of standards" is now over but I am sorry that I have been indirectly at fault in the mistake that has arisen".* They published another paper in July of 1906 (same title) with a revised value of 3.8 x 10^{-7} grams of radium per gram of uranium (Rutherford and Boltwood, 1906).[5]

Their first reference to the U.S. Bureau of Standards (NBS) came on April 28,1906. Rutherford reported to Boltwood that he was down in Washington for a meeting of the Physical Society. He reported that he was dead tired and that the meeting was "largely attended by physicists from the Bureau of Standards" (Badash, 1969 p.134).

2.2. W.F. Hillebrand, Chief Chemist of NBS, and the Age of the Earth

At that April APS meeting, Rutherford probably met the physicists Stratton, Rosa, Wolff, Vinal and Dorsey, but did not mention a chemist from the US Geological Society (USGS) who may have attended the same meeting. William Francis Hillebrand was the Chief Chemist at the USGS who in 1906 was the president of the American Chemical Society. In 1909 Professor Stratton recruited Professor Hillebrand to be the NBS chief chemist and named Rosa as the chief physicist (Cochrane, 1956).[6]

Although Rutherford did not mention Hillebrand, it is interesting that, during the same exchange of letters in 1904, Boltwood referenced Hillebrand's analyses of uranium and other chemical

[5] This value would be revised again when Rutherford had access to one of the international standards. A current estimate based on the ratio of half lives is 3.58 x 10^{-7}.

[6] Both scientists have major awards given in their name: The annual Hillebrand Prize of the Chemical Society of Washington (CSW) is awarded for original contributions to the science of chemistry by member(s) of the local section of the American Chemical Society. A number of prominent NBS/NIST chemists have received the Hillebrand Prize. The Edward Bennett Rosa award of the National Institute of Standards and Technology is awarded for significant contributions to measurement sciences.

elements in minerals from around the world.[7] In April 1905 Boltwood sent Rutherford a table of Hillebrand's values for lead, nitrogen, and uranium in 12 minerals. Rutherford would not have been aware of the detailed chemical analyses of minerals by Hillebrand as the latter's results were mostly published in bulletins of the US Geological Survey (Hillebrand, 1891, 1892, 1893) and the American Journal of Science which focused on geological studies (Hillebrand and Ransome, 1900). The ratio of Pb to U was highest in the oldest mineral and lowest in the youngest. Boltwood noted: *"If lead can be shown to be a disintegration product of uranium, will it not necessarily follow that all the lead existing on the globe originated in this way? I think that the deductions that can be made from this assumption will make even the metaphysicians dizzy."* Boltwood was sharing his ideas with Rutherford while at the same time submitting papers to the American Journal of Science (Boltwood, 1904a, 1904b, 1905, 1907). In addition to uranium and lead, Hillebrand had found entrapped gases in his samples, which he assigned to nitrogen. Ramsay and Soddy later showed that several uranium-bearing minerals contained helium and argon, and Ramsay questioned Hillebrand's finding of nitrogen. Hillebrand and Ramsay cleared up the discrepancy in an 1895 series of personal letters (Clarke, 1925). Hillebrand's Connecticut uraninite did in fact have nitrogen in the entrapped gas along with argon and helium. Hillebrand acknowledged that he simply missed the spectroscopic analyses of the other gases. In November 1905, Boltwood wrote to Rutherford with more data on Pb/U ratios from 12 minerals analyzed by five chemists including Hillebrand and reported "lead and helium are the disintegration products of uranium only." (Boltwood Nov. 1905).

Boltwood credited Rutherford with the novel idea that these quantitative ratios of the uranium decay products could be used in radioactive dating, and these would in turn lead to new estimates of the age of the earth (Boltwood, 1907a, Badash 1968, 1979 p.92). Using their measured mass ratio of Ra/U of 3.8×10^{-7} and Rutherford's value of the radium half life of 2600 years, Boltwood calculated that about 10^{-10} grams of radium is produced per gram of uranium. The fraction of uranium decaying each year is also about 10^{-10}. And thus, the quantity of lead produced each year from a gram of uranium would also be about 10^{-10} grams. Boltwood then divided the Pb/U ratios by 10^{-10} to obtain the age of the mineral.

$$\text{Age} = \text{Pb/U} \times 10^{10} \text{ years}$$

He found that the minerals dated from 410 million years (Connecticut uraninite) to 2200 million years (Ceylonese thorianite). A summary of Boltwood's 1907 data is given in Figure 3.

[7] In 1898, Pierre Curie had written to Hillebrand asking his assistance in identifying American ores that might contain polonium radioactivity. Hillebrand provided information and arranged for the Smithsonian Institution to provide Curie with a 500 g sample of uraninite from Colorado (Blanc, 2009).

In the table which follows (Table VI) the ages of the minerals included under Table I have been roughly calculated in accordance with the method outlined above. The ages of the minerals in years are obtained by multiplying the average value of the ratio 10^{10}. The general plan of calculating the ages of the minerals in this manner was first suggested to the writer by Prof. Rutherford.

TABLE VI.

Locality.	Age of minerals in million years.
Glastonbury (Portland), Conn.	410
Branchville, Conn.	535
Spruce Pine, N. C.	510
Marietta, S. C.	460
Llano and Burnet Co., Texas	1800
Douglas Co., Colorado	1900
Moss District, Norway	1300
Annerod, Norway	1700
Sabaragamuwa Prov., Ceylon	2200
Galle District, Ceylon	860

The actual values obtained for these ages are, of course, dependent on the value taken for the rate of disintegration of radium. When the latter has been determined with certainty, the ages as calculated in this manner will receive a greater significance, and may perhaps be of considerable value for determining the actual ages of certain geological formations.

Figure 3. Table VI and text from Boltwood showing ages for 10 minerals from different locations. American Journal of Science, Vol. 134, p.87, 1907.

Boltwood noted “The number of such analyses to be found in the literature is not large, and what is still more unfortunate, with the exception of those made by Hillebrand and a few others, cannot be considered as particularly accurate.” Hillebrand’s analytical data for lead and uranium were used for all the samples except the two from Ceylon for which Boltwood used his own analyses. Boltwood noted at the time that the exact ages would be known with greater accuracy with a better value of the half life of radium. He would then turn his attention to finding the parent nuclide for radium (Boltwood, 1907b) and the radium half life (Boltwood, 1915).

The estimated age of the earth at the time based on thermodynamic measurements by Lord Kelvin was about 100 million years with some values of the order of 20 million years (Badash, 1968, 1989). But, in the early part of the century there was a problem noted with Kelvin’s model which assumed a uniform cooling rate of the earth; radium decay was shown to be adding heat to the mineral layer of the earth which would extend the life beyond 100 million years. Boltwood’s paper, in a journal concerned with geological studies, was not immediately accepted in the larger scientific community. The English physicist R.J. Strutt continued similar work to Boltwood considering both helium and lead concentrations in other minerals, and his efforts, expanded by the English geologist Arthur Holmes, led to general acceptance of the lead/uranium method for dating the age of the earth’s crust (Strutt, 1908; Holmes, 1911; Badash 1989). The current estimate of the age of the earth is 4.543 billion years, although the earliest rocks amenable to dating by radiometric methods were formed about 3.8 billion years ago. In conclusion one could say that the idea of dating the age of the earth came from Rutherford, the exposition was provided by Boltwood, using the data of Hillebrand and Rutherford.

2.3. The McCoy Alpha-particle standards

In parallel to the work using emanation standards, early investigators used solid sources of uranium compounds to calibrate their electroscopes (see Table 7) (Randall, 1912; Badash, 1979). Apart from the Boltwood standard, the standard proposed by Herbert Newby McCoy was the most used in the US. It is interesting to look at the differences and similarities between these two US chemists (Badash, 1979).

Boltwood was independently wealthy and was able to attend university in Germany and New Haven. McCoy came from a working-class background, obtained his undergraduate degree at Purdue University, and worked in industry before graduate studies at the University of Chicago. Boltwood worked mainly alone in the laboratory while McCoy mentored many capable research students.

There were many similarities in the period 1900 – 1908. Both were working with commercial mining interests to develop chemical procedures to extract radium from ores. They were engaged in parallel research investigations of uranium and thorium radioactive decay products. And they were both proficient in constructing electroscopes, and in quantitative measurements of alpha particles from pure uranium and thorium, and from radium-226 and progeny. Although they did not collaborate directly, they did study and reference each other's papers, and had some limited conversations in addition to letters in journals with comments on each other's work (Badash, 1979).

McCoy and his students were also extracting radium-226 from minerals and making emanation measurements with electroscopes. But McCoy proposed a simpler method that would not require complete acid dissolution of the minerals (McCoy 1905). He proposed as a standard the emission of alpha particles from a square centimeter of a powdered specimen of U_3O_8 (McCoy & Ross, 1908).

> "The standard of activity was a thick film of the oxide U_3O_8, weighing about 0.025 g. per sq. cm. It was deposited on a flat copper plate 7.00 cm. in diameter. The activity due to each sq. cm. of such a film is taken as the unit of activity. The total activity of 1 g. of any substance, in terms of such a unit, may be called its specific activity.
>
> The specific activity of pure uranium oxide, U_3O_8, was redetermined from measurements of a series of new films on flat plates, the new electroscope being used with the films at 8.5 cm. from the electrode. The results, calculated by the method of graphical extrapolation previously used gave for the specific activity of the oxide, 676. Since the oxide contains 84.85 per cent, of uranium, the specific activity of uranium is 796. This value differs but little from the value, 790, found earlier."

This standard works fine for pure U_3O_8, but initially there was a problem when a value for a mineral was assigned some multiple of the 796 value for uranium. For a mineral containing uranium and thorium in equilibrium with their decay products, the ionization is caused by alpha particles of different ranges from multiple radionuclides. Some of these are entrapped in the mineral, so the ionization does not represent the complete set of alpha-particle emitting constituents. Boltwood pointed out this shortcoming in a letter to Rutherford (Badash, 1969) and mentioned he would discuss it with McCoy. There is no record of their conversation, but McCoy did make the correction in his 1908 paper (McCoy & Ross, 1908). To correct for the entrapped

gas, he subjected a mineral sample to a total dissolution and collected all the radon, such that his values of U + Ra and progenies/U were in agreement with Boltwood (Badash, 1979).

McCoy and Ashman applied several corrections to their data and arrived at the value of the ionization current from one gram of uranium of 4.61×10^{-10} amperes. Boltwood also prepared films of different thickness and plotted ionization current versus source mass to extrapolate to zero mass (Boltwood, 1908). The two US chemists reported that their results were in good agreement (McCoy and Ashman, 1908).

2.4. The C.G.S. System International (SI) Standards

Randall made the interesting observation that the US standards referred to above were in general agreement, while the Europeans used the C.G.S. standards (centimeter – gram – second) (Randall, 1912). He was referring to results reported in SI units of current (in amperes) and charge (in stat-coulombs). For example, in Marie Curie's doctoral thesis she reported measurements on several uranium compounds and "represented the intensity of the current in amperes by the letter *i*…" (Curie, 1903)

Uranium compound	*i* (picoamperes)
Metallic uranium (containing a little carbon)	23
Black oxide of uranium, U_2O_5	26
Green oxide of uranium, U_3O_8	28
Hydrated uranic acid	6
Nitrate of uranium	7

She noted at the time that the ionization from a uniform source of uranium powder did not change appreciably with thickness. She found that a 0.5 mm layer of uranium oxide produced a current of 27 pA, while a 3.0 mm layer produced a current of 30 pA. Marie Curie's measurements were repeated shortly after by both McCoy and Boltwood, and the protocol for the McCoy standard builds on Curie's earlier measurements.

Another standard in use in Europe was proposed by Mache (Mache, 1904a,b). He used a galvanometer similar to the one used by Curie to measure the ionization current due (primarily) to radon and progeny in a measured volume of air. Mache proposed a unit for radon activity concentration called the Mache-Einheit where 1 Mache Einheit (ME), corresponded to 13.5 Bq/L (364 pCi/L).

In his master's thesis at the University of Missouri, Randall set about to reconcile all the proposed "standards" up to 1908 (Randall, 1912). To accomplish this, he assembled or constructed a set of electroscopes of the designs used by the various investigators. He also constructed emanation collection apparatus that would match the systems used by Boltwood and others. Randall pointed out that in none of the electroscopes in use did the alpha particles achieve the full range in air. And since the radon progeny (Radium A, B and C) are absorbed on the walls of the electroscope, only half of their alpha particles contribute to the air ionization. His summary included 14 observations he made from his comparative study of the various standards. His first three observations are given here (Randall 1912):

1. It has been experimentally shown that in general no constant relation exists between the Boltwood emanation standard and the C. G. S. electrostatic standard as ordinarily used.
2. The maximum ionization current produced in the ordinary electroscopes by the radium emanation and its products from an old mineral containing one gram uranium was calculated as 6.18 x 10 $^{-10}$ amperes, or 1.854 C. G. S. units.
3. For small electroscopes of the same design, the observed ionization current is nearly proportional to the volume of the ionization chamber.

This third point presages the concept of exposure standards based on charge per unit volume.

2.5. Gamma-ray emission rate standards

A secondary method of standardization – that is, one that required comparison with a primary standard – was to compare an unknown material with a sample of known activity by gamma-ray measurement. Eve had actually proposed this method as early as 1906 when he suggested gamma-ray measurements against 1 kg of thorium nitrate in a thin-walled cylindrical glass vessel 16 cm in diameter with the radiation filtered through 1 cm of lead (Eve 1906). This introduced the ideas of a reference material in a standardized geometry with filtration to remove extraneous lower-energy radiations. The gamma-ray method became more widely used by the end of the first decade of the century as reliable standards of mass of radium chloride and radium bromide became available from Paris (Marie Curie) and Vienna (Stefan Meyer).

By 1908 the x-ray physicists and physicians in the English Roentgen Ray Society proposed as a standard comparison of the gamma-ray emission rate of a sample with 1 milligram of pure radium bromide filtered through 1 cm of lead (Butcher, 1908). The committee that made this recommendation included Rutherford and Soddy (Smith, 1975). This was ideal for industry as they were soon able to obtain standards in milligram quantities from Paris and Vienna. The committee also included x-ray physicists including Sir William Crookes and G.W.C Kaye of the UK National Physical Laboratory. Gamma rays from radium became the standard for x rays as well, since the x-ray tubes of the day could not produce a stable, reproducible current (Smith, 1975). The stage was now set for the introduction of an international standard for radioactivity.

3. Standards: 1910 - 1915

3.1. The International Radium Standards Commission

The first formal action on establishing an international standard for radioactivity came 12 – 15 September 1910 at the International Congress on Radiology and Electricity in Brussels. A special committee was appointed to report to the Congress on the best means to establish an international standard (Rutherford, 1910), Frame (1996), Boudia (1997). It is interesting to look at the members selected to serve on the International Radium Standards Commission (Table 7).

Table 7. The International Radium Standards Commission of 1910.

Marie Curie	Sorbonne University, Paris
André-Louis Debierne	Sorbonne University, Paris
Ernest Rutherford	Manchester University, Manchester
Frederick Soddy	Glasgow University, Glasgow
Otto Hahn	Kaiser Wilhelm Institute for Chemistry, Berlin
Hans Geitel	Gymnasium Wolfenbüttel, Braunschweig
Stefan Meyer	Academy of Sciences, Vienna
Egon R. Von Schweidler	University of Innsbruck, Innsbruck
Arthur S. Eve	McGill University, Montreal
Bertram B. Boltwood	Yale University, New Haven

The three principal members; those who signed the certificates were Curie, Rutherford, and Meyer. Perhaps not surprisingly, since they spoke three different languages, the other seven members of the international commission were French, English and German speakers who were collaborators with the principals. Curie included her chief collaborator, the chemist Debierne. Four of the others, Soddy, Hahn, Eve and Boltwood, were collaborators with Rutherford at McGill and Manchester. Geitel and von Schweidler were two more German speakers who were associated with the radium research in the Austro-Hungarian empire. Hahn the German chemist in Berlin had spent time with Rutherford in Montreal, with Ramsay in London and with Meyer in Vienna. Given the oversize importance that the US was soon to play in this field, it is noteworthy that the US had only one representative, Boltwood, a chemist with a joint chair in the physics department at Yale University. He was the official US delegate to the 1910 International Congress on Radiology and Electricity at the time the Congress appointed members of the commission. Marie Curie reported that the United States had two members of the commission: Boltwood and Eve (Curie, 1912). She apparently did not make a distinction between Canada and the United States. Rutherford was named president with Marie Curie and Stefan Meyer from Vienna as the principal members (with Meyer serving as secretary).

Frame has described the interesting discussions of that first meeting in Brussels with French, English and German speakers each trying to make their case for the quantity and unit to describe radioactivity (Frame, 1996). Boltwood and probably others accepted the idea that the named unit of activity would be the *Curie* in honor of the founders, but they wanted the unit quantity of activity to be some small amount close to the quantities usually measured by the emanation techniques. But Marie Curie objected that the quantity honoring her late husband should not be such an infinitesimal value. Marie Curie would prevail, and a standard of radioactivity (1 Curie) was defined as that quantity of radium emanation (radon-222) in equilibrium with one gram of radium element (radium-226) (Curie, 1912). She agreed to prepare a radium standard. The Paris

Standard that she prepared in 1911 contained 21.99 mg of radium chloride. The glass ampoule contained a thin platinum wire extending beyond the seal which was thought at the time to be necessary to prevent a charge buildup in the ampoule. Subsequently, five standards were prepared in Vienna by the chemist Otto Hönigschmid (at the time at the Prague Polytechnical University) having masses of 10.11, 31.17, 40.43, 236.91 and 680.50 mg of radium chloride (Meyer and Hess, 1912). The Paris standard and the three smaller Vienna standards were sealed in Thurigian glass tubes having a wall diameter of 0.27 mm.

In March of 1912 seven members of the commission traveled to Paris to compare the two standards (Curie, 1912; Hahn et al., 1912). Boltwood, Eve and Geitel did not attend. Only the 31.17 mg Vienna standard was used in the comparison. Rutherford described the measurements and the electroscope used from the British side (Rutherford, 1912; Rutherford and Chadwick, 1912). James Chadwick was Rutherford's undergraduate student at Manchester University at the time and this was one of his first published papers (Brown, 1997). Marie Curie describes the electroscopic method used by André Debierne in her paper (Curie, 1912). Hahn et al. described the measurements from the German perspective (Hahn et al. 1912). The two standards agreed to within 0.2 % (Curie, 1912). These radium sources were to serve as "the primary international standard" for the next quarter century.

Marie Curie wanted to retain the Paris standard in her laboratory, but Rutherford objected that the international standard should not be retained in a working research establishment, but rather it should be stored in an institute devoted to metrology. Thus, he elected that the British standard should be maintained at the National Physical Laboratory in Teddington, the national measurement institute for the United Kingdom. This was probably a politically wise move as universities in Manchester, London, Cambridge and Glasgow could all make a case for keeping the UK standard. Boltwood, the US member of the Commission, had a similar discussion with Rosa at NBS and Rosa recommended that the international standard be retained at the Bureau International des Poids et Mesures (BIPM) in Sevrés, France. Boltwood conveyed this suggestion to Rutherford who convinced Marie Curie that she should allow the BIPM to retain the physical standard (Badash, 1969). Marie Curie agreed in part because of a personal friendship with Charles-Edouard Guillaume, then deputy director of the BIPM (Ariouet, 2021). She also agreed because the institute in Vienna provided her with an equal amount of radium to replace that in the standard. At the time, the BIPM did not have a program in radioactivity but agreed to keep the standard. Marie Curie and André Debierne travelled from the Sorbonne to Sevrés whenever they needed the "Paris Standard" for measurement purposes (Ariouet, 2021). The replacement cost of the Austrian radium was paid by the Sir George and Lady Beilby, the in-laws of Frederick Soddy (Badash, 1979).

3.2. International Secondary Standards

The next step for the International Radium Standards Commission was to commission the chemist Hönigschmid to prepare secondary standards for distribution to several national standards bodies and larger industrial facilities. The main activities of the commission were carried out by the secretary Stefan Meyer in Vienna in coordination with Hönigschmid and the Austrian government. Hönigschmid used radium chloride prepared from the pitchblende residues at St. Joachimsthal. Recipients had to arrange payment in advance from the Austrians. The US State Department purchased the US standard. Dr. G.T. Beilby, Soddy's father-in-law,

paid the 354 pounds (British) (about $35,000 in 2023 USD) for the UK standard (Smith, 1975). Seven secondary standards, prepared in October 1912 and July of 1913, are listed in Table 8 (Meyer and von Schweidler, 1916).

Table 8. From a table by Meyer and von Schweidler 1916 giving the disposition of the initial seven secondary standards. As indicated, these standards were sent to France, Germany, England, the United States, Sweden, Japan and Portugal. Each was directly compared to the Paris and Vienna primary standards.

Land	ein-geschmolzen am	Wiener Messung $RaCl_2$ (mg)	Pariser Messung	Gewähltes Mittel
Frankreich	4. Okt. 1912	22.47	22.42	22.45
Deutsches Reich	4. Okt. 1912	19.73	19.74	19.73
England	4. Okt. 1912	21.10	21.16	21.13
Ver. Staaten Amerika	1. Juli 1913	20.29	20.28	20.28
Schweden	1. Juli 1913	9.74	9.71	9.73
Japan	1. Juli 1913	9.80	9.80	9.80
Portugal	1. Juli 1913	9.07	9.11	9.09

The sealed ampoules were measured first in Vienna, probably by Hess and von Schweidler (Meyer et al., 1912). Hess was carrying out this calibration work during the same period that he discovered cosmic rays – from an elevated signal in his electroscopes – for which he was to receive the 1936 Nobel Prize in physics (Hess, 1912). Separate arrangements were required to ship the ampoules to the Curie laboratory in Paris where Ellen Gleditsch reported that the standards measurements were carried out by Debierne (Boltwood letter to Rutherford (Badash, 1969)).

3.3. NBS Preparation for Receipt of the US National Standard

Prior to 1913 there were already a few standards in the United States as the principal radium supplier in the US, the Standard Chemical Company of Pittsburgh, had purchased standards from both Paris and Vienna (Viol, 1914). These will be discussed in a later section on the US radium industry. The radium standards program at NBS began in the Spring of 1913 when Rosa charged N.E. Dorsey with preparation for receipt of the US standard. Late in 1911 Dorsey had traveled to Europe to compare electrical standards with “other standardization laboratories” (NIST archives, 2023). These visits probably included NPL, to meet with G.W.C. Kaye in the Physics Department, and other laboratories on the continent. On his return home Dorsey had the same problem familiar to many NBS/NIST researchers. The customs officials questioned his possession of government owned scientific equipment without proper paperwork. He and his wife had to scrape together $20 (about $620 in 2023 USD) to pay the customs fees (NIST archives).

Dorsey did not publish on his early work in radioactivity although some of his notebooks are available in NIST archives. Unsurprisingly, the few papers remaining are slightly contaminated with radium-226. These have been sealed and saved for future reference. A few excerpts from these notebooks and other records will be included in this section. Fortunately, Boltwood, as the US representative to the International Radium Standards Commission, maintained his extensive correspondence with Dorsey over the next decade, and these are collected in the archives at Yale University (Yale, 2023). The first 8 letters between May and December 1913 are reproduced here along with commentary.

3.4. Boltwood – Dorsey Correspondence: May – December 1913

From the Yale University archives with explanatory notes (Yale, 20213). The typeset of the letters, complete with misspellings, is intended to replicate the original correspondence.

Letter of May 14, 1913 from N.E. Dorsey - Handwritten on stationery from Cosmos Club

Bureau of Standards
Washington, D.C.
May 14, 1913

Prof. B.B. Boltwood,
Yale University
New Haven, Ct.

Dear Prof. Boltwood: -

When you were in Washington a few weeks ago Prof. Rosa told you, I believe, that I wished, and the Bureau desired me, to visit your laboratory some time before vacation.

Will it be convenient for you for me to make the proposed visit the later part of next week? If this time is not convenient I shall defer the trip until later.

Trusting that I may soon have the pleasure of meeting you, I remain

Very truly yours
N. Ernest Dorsey

This is the first correspondence between Dorsey and Boltwood. The recent meeting between Professor Rosa and Professor Boltwood probably occurred at the annual meeting of the American Physical Society. Rosa, Stratton and Dorsey were all members of the Cosmos Club, which explains why Dorsey wrote the letter on club stationery.

Letter of June 4, 1913 from Dorsey – handwritten

Bureau of Standards
Washington, D.C.
June 4, 1913

Dear Professor Boltwood:-

After visiting New Haven I spent several days in New York visiting the hospitals and physicians. Among others I met Dr. Robert Abbe who has been using radium in sealed tubes for the treatment of cancer. He has been in the work for about ten years and seems to be wonderfully successful. Do you know anything about him? He tells me that he has frequently had the sealed (glass) tubes to explode. To avoid danger to the patient in case of such an explosion he always places the sealed tubes inside of celuloid or quill case. Have you noticed anything of the sort? I think you said that quartz tubes would go to pieces but that glass would not. Abbe intercompares his salts photographically and has thus discovered that the French preparations run very short.

I also met some very live and interesting x-ray men, and find that there is plenty of work ahead of me along that line.

Yesterday I met a Mr. Turner of the American Vanadium Co. His work is bibliographical. He tells me that the Co. obtained a gram of radium bromide, 95 +% pure, in April. And late in May it appeared that they would have another gram by the end of May. They expect to found a bank for the renting of radium salts. They expect to make applicators of various kinds, radioactive ointments, muds, and waters, emanators, etc. I understand that no printed announcement of those plans have yet been prepared.

I can not tell you how much I enjoyed and profited by my trip to the Sloan Laboratory. It greatly clarified my ideas on the subject of radioactivity work. I shall be still more indebted to you if, while abroad, you will ascertain for me the European opinion as to the best methods of intercomparison of sealed salts, and the accepted value of the "Mache einheit".

Trusting that you may have a very pleasant trip, I remain

Very sincerely

N.E. Dorsey

Robert Abbe, a New York physician, is generally credited with being the first in the US to use radium in therapy (e.g., Abbe, 1904; Slaughter, 2013; Robison, 2015). His first treatments were with milligram amounts that he obtained from Marie Curie. Several of his artifacts are now in collection of the National Museum of American History of the Smithsonian Institution in Washington (Smithsonian Institution, 2023). Dorsey was also assigned responsibilities for x-ray standards. The x-ray work will be described later. The American Vanadium Company was part of the network of companies owned by the Flannery brothers of Pittsburgh that included the Standard Chemical Company soon to become one of the largest suppliers of radium for the worldwide market. Lubenau reports that Mr. Turner was a marketing man from one of Flannery's other companies and was not in a position to provide details on radium production (Lubenau, 2023). Boltwood never replied to the question about the "Mache Einheit". Dorsey was referring to the proposed unit of radioactivity by the German Heinrich Mache (Mache, 1904a, 1904b) (described previously in section 2.4).

Letter of December 1, 1913 from Dorsey (typed)

Bureau of Standards,
Washington, D.C.,
Dec. 1, 1913.

Prof. B.B. Boltwood,
New Haven, Conn,

My dear Prof. Boltwood:-

I fear that you think I am a very unappreciative kind of chap. Really though I did greatly appreciate your sending me the separate of Rutherford's paper, and fully intended to

tell you so long ago. At first I did not know where to find you, so decided to wait until your return; then I simply delayed. Pray excuse me.

Things have gone very slowly. The standard ordered last March has not yet arrived. We have however bought two sealed specimens of radium from the Pittsburgh people, -- a one, a four mg. These have been used to try out the apparatus. We have built a few emanation electroscopes, and have a complete Rutherford setup. The results by the two methods are in close agreement.

I am rather pleased with the Rutherford method. At present the leads are not so good as I desire, but hope to have them better in a few days. Varying the shorter distance in the ratio of 7 to 10 does not change the observed ratio of the two specimens. Have also measured them through lead screens of various thicknesses from 3mm to 23mm; The observed ratio of the large specimen to the small one is slightly smaller for the thicker screens. I believe this is a spurious effect, and trust that it will vanish when I get my new leads installed.

I visited the laboratory of the Standard Chemical Co. last September. They had quite a little radium on hand, and I saw figures that seemed to show that they had prepared about a gram of the element during August. Of course this is not saying that they have mined that much ore; I understand that the Bureau of Mines is confident that they are not mining nearly so rapidly. The men I met there seemed to be very fair kind of fellows.

Do you know any method of detecting the presence of meso-thorium in sealed specimens of radium. I am told that such adulteration is becoming quite common. What are the European laboratories doing about it?

Please remember me to Bumstead, Uhler, and any other friends who may think of us.

Sincerely yours,

N. Ernest Dorsey

Dorsey refers to the two radium specimens that he purchased from the Standard Chemical Company. The salts were provided as sulfates, and both had significant residual barium. The larger was certified to contain 4.10 mg of radium on September 5, 1913. The smaller one was 1.04 mg of radium with a certificate of the same date. Certificates were signed by Lester Walker, Chief Physicist and Charles Viol, Director. One of the certificates is shown here.

Figure 4. Certificate for radium specimen supplied to US Bureau of Standards (NIST archives).

Dorsey's cryptic reference to his conversation with the US Bureau of Mines (USBM) is interesting. The USBM chemists, Charles Parson and Richard Moore, were in competition with the SCC to produce radium (Parson, 1913) and may have been discounting their competitors.

Four-page letter of December 3, 1913 from Boltwood

December 3, 1913.

Mr. N. Ernest Dorsey,
Bureau of Standards,
Washington, D.C.

My dear Mr. Dorsey:

Your letter of the first of December just reached me and I must acknowledge that I was wondering how you were getting along in regard to the testing of radium preparations at the Bureau of Standards. As I have had a good deal to occupy me since I got back to New Haven in September it is unnecessary to say that I have not been looking for trouble in any particular direction as I might have been otherwise if I had had spare time on my hands.

I am surprised to learn that the standard ordered last March from Vienna has not yet reached you. The trouble must be due to delay in Paris because I had the standard in my

hands early in September and it was then being measured with the expectation of having the work completed by the end of the month. I think it is perfectly justifiable for you to write to Marie Curie's laboratory in Paris and ask them to send it to you as soon as possible. It is unnecessary to emphasize the importance of having the standard in this country at the earliest possible date.

I am much interested in what you tell me about the experiments you have made using the Rutherford method for comparing specimens of radium salts by the gamma rays. I am glad to learn that you have obtained such good agreement which I understand is one of the advantages of this method according to the Manchester people.

From what I learned during my trip abroad I can say that the Rutherford method is the one which was used in Manchester for comparing radium specimens. At the Radium Institute in Vienna they used the same method as that employed in Paris; a circular, horizontal, thick lead plate supported on a tripod with a brass ionization vessel below it containing an insulated disc to which a Wolf quartz fiber electroscope is attached. The radium preparation is laid directly on the center of the lead plate and the apparatus, from the standpoint of simplicity of construction and manipulation, has many obvious advantages. Professor Meyer assured me that he would be very glad to supply you with a full detailed description of the construction of the apparatus. He also said that if you desired he would have constructed for you in his workshop a duplicate of the apparatus which they have there. I think, if you can afford to invest in one of these instruments, you would find it very convenient for the general measurement of radium specimens. I should not judge that the expense would be very considerable, probably something like $150.

I also had a talk with Dr. Geiger at the Technische Reichsanstalt and learned from him that he was using a simple lead gold leaf electroscope in a closed lead case for making the comparisons and was getting thoroughly satisfactory results by this method. He told me incidentally that the charge they were making for such measurements there was 50 marks for each specimen examined and certified.

I am interested in what you tell me of the Standard Chemical Company and especially so in regard to their statements concerning the amount of radium which they separated during the month of August. I am a little skeptical in regard to their claims as to the quantities of this material which they are preparing and I doubt very much whether their average per month is anything like the figure which you mention. I am glad to know that the people there made a good impression on you. The specimen of their material which came into my hands for testing a few weeks ago was certainly quite up to the standard which was claimed for it.

In regard to the adulteration of radium preparations by adding mesothorium, I can say that I doubt very much whether this can be considered as at all common. Mesothorium is now selling at a price considerably higher than radium of equivalent activity, so the adding of mesothorium under these conditions would scarcely be profitable. The best method I know of for testing a radium preparation (it must be either crystalline bromide or chloride) is one which Rutherford described to me as having been used in his laboratory. The tube containing the radium salt is notched at the end with a glass-cutting file and cracked by touching with a fiber of molten glass. The tube is then placed in a small electrical furnace and heated (in a good hood or else remote from the testing room) to a temperature of about 350°. This heating replaces practically all of the radium emanation. The measurement of the gamma ray activity

of the preparation for several hours after the heating should show the fall in activity to a minimum value and by extrapolation of the curve showing the variation of the activity it can easily be learned whether the activity is due solely to radium. That is to say, it can be shown that the preparation is free of mesothorium since the gamma ray activity of this is practically unaffected by the heating process while the gamma ray activity of radium falls to a value which is only a few percent of the maximum gamma ray activity. This heating process can only be carried out satisfactorily once on any given specimen since after it has once been heated (and dehydrated) the second heating does not remove the emanation. The only general method for testing for mesothorium is to dissolve a known fraction of the radium salt in dilute hydrochloric acid, dilute this solution to a convenient strength and then test for radium by the emanation method. In the case of insoluble radium salts, like the sulphate which is I believe much used in medical preparations, this method (or any method for that matter) is likely to be quite troublesome. Dr. Geiger tells me that he makes no tests for the presence of mesothorium and specifically states in his certificate that the presence of mesothorium is not excluded.

If there are any other matters along these lines in which I can be of assistance to you please do not hesitate to let me know. I shall be much interested to learn when you get the standard and I hope you will let me know when it arrives.

With kind regards and best wishes

Yours sincerely,

Boltwood provides a great deal of useful information to Dorsey in this letter. It seems that Boltwood felt some responsibility as the sole US representative to the International Radium Standards Commission to make sure that the US standard made its way to NBS. He provides a brief update on his Summer of 1913 in Europe. He had visited Rutherford at Manchester University in the UK, Hans Geiger at the Technische Reichsanstalt (precursor to the Physikalisch Technische Bundesanstalt (PTB)) in Berlin, and Stefan Meyer at the Institute for Radium Research in Vienna.[8] Meyer was the secretary of the Commission. Otto Hönigschmid had prepared three standards on October 4, 1912, that went to France, UK and Germany. He prepared a second set of four standards on July 1, 1913, that were destined for the US, Sweden, Japan and Portugal. When Boltwood said that "he had the standard in his hands in September" he didn't say whether he was in Vienna or Paris. But it still had to be measured and certified in Paris before shipment to the US.

This letter has a good brief description of the apparatus in use in Manchester, Paris, Vienna and Berlin. It is interesting that Stefan Meyer was willing to fabricate a detector for NBS for about $150 (Boltwood's estimate), which would be about $4600 in 2023. Dorsey had already prepared a suitable apparatus for the comparisons, but he followed up with a letter asking Meyer for details of the system in Vienna. One could review the original papers to see the minor differences, but they all seemed suitable for comparing the gamma-ray emissions from two radium specimens for the purposes of measuring a ratio of the activities. The Paris system is described by Marie Curie (Curie, 1912).

[8] Boltwood must have also met Hönigschmid during this trip since the latter sent him a very interesting "Radium" postcard to commemorate their visit (Lubenau et al. 2020).

It is interesting that Dorsey and Boltwood are comparing notes and opinions on the operations at the Standard Chemical Company in Pittsburgh. Dorsey had made a very fruitful visit to Pittsburgh in September 1913 and met with Charles Viol and others. The company was making great strides towards record production of radium from carnotite ores from Paradise Valley in Colorado/Utah, and in a few years would become the world's largest radium supplier. In January 1914, Viol would testify before Congress and mention his connections with Dorsey at NBS.

Boltwood did not agree with Dorsey that contamination of radium with mesothorium was a common problem. Frederick Soddy in 1913 had discovered that radioelements of differing mass are isotopes of the same element; hence mesothorium was in fact radium-228. Boltwood should have heard about Soddy's work by December 1913. A comparison of the decay schemes for radium-226 and radium-228 helps explain Boltwood's method for heating to eliminate the radium emanation (3.6 day half-life radon-222). This eliminates the higher-energy gamma rays from the radon progeny which are the main contributors to the response of the ionization apparatus. Boltwood was aware of the danger of releasing the radon in the testing room and suggested use of a good hood.

Boltwood also passed on the information from Berlin that the counterparts to the US standards laboratory (the German PTR) were charging 50 marks for measurement and certification of a sample submitted for test. In 1913 the exchange rate was 4.2 DM per USD. Dorsey would check back with Boltwood later as he set the calibration fees for NBS.

Typed letter of December 11, 1913 from Dorsey

Bureau of Standards,
Washington, D.C.,
Dec. 11, 1913.

Prof. B.,B. Boltwood,
Yale University,
New Haven, Conn.

My dear Prof. Boltwood:-

To say that I greatly enjoyed your letter of a week ago is putting it very mildly. I was especially interested to learn that our standard had reached Paris so early as September; the only excuse that I can see for the long delay since then is a possible disagreement between the Paris and Vienna values. We have written Prof. Meyer in regard to it. We have also asked him to send us details of his apparatus: I should like to, eventually, have copies of the apparatus that is used in the various standardizing laboratories.

Speaking of discrepancies in the measuring of radium, you may be interested to hear of two cases that have come to my notice. The most striking is that of one of the standards belonging to the Standard Chemical Company. They have three standards: -1. A two mg of Austrian radium, certified by Meyer; 2. A six mg of their own salt, certified by Marie Curie; 3. A 19 mg of the same salt as was used in the second, certified by Hess. The Curie and Hess standards are concordant, but in terms of these the Meyer is about 2% too high. That the certified value of the Meyer is too high is also borne out by the fact that their "pure" salts run nearly 102% pure, they tell me, when referred to this standard. When I was in Pittsburgh I intercompared their standards, using their apparatus, and found the same discrepancy that they had found.

The other case is the standard that Dr. Kelly has. This is supposed to be known in terms of Rutherford's. As I have not yet been able to see the data I do not know what uncertainty there may be in the reduction to the International Standard; but Dr. Kelly tells me that the assumed value of his standard is too high. Dr. Viol of the Standard Chemical Company had already told me the same thing; he told me that their "pure" salt which Dr. Kelly bought ran very appreciably over 100% when compared with this standard. Dr. Burnam does all the measuring for Dr. Kelly and keeps the data regarding the standard. He is now in Europe, When he returns I hope to get his records, and shall then endeavor to find out where the trouble is.

(2)

I interpret the sentence at the bottom of page 2 in your letter to mean that the standards (i.e. The Curie and Hess standards) of the Standard Chemical Company agree with yours. This gives and indirect comparison of our two specimens with yours. Would you mind telling me the order of magnitude of the discrepancy you found? Was it as great as 1 /2 %?

Some time ago Prof. Rosa told me that he intended to write you in regard to the possibility of the Bureau borrowing your standard for a short time. I do not know if he has done so yet, he has said nothing more to me about it. Entirely unofficially, I am now going to ask some questions on my account; if you have already answered them to Prof. Rosa please do not bother to answer them again. First, will you please tell me the size of your standard, or standards; and with what accuracy they are known in terms of the International Standard. Second, while awaiting our standard from Austria, would you be willing (and under what conditions) to lend us one of your standards for a short time, should a necessity for it arise? The necessity may never arise, and the Burau may disapprove of any such proceedings, but it is evident that there might come a time when it would be very desirable to have a well known standard in addition to the specimens that we now have.

Regarding fees, I have already seen the P.T.R. schedule in the Zeits. Für Instk.: on the Hess certificate in the possession of the Stand. Chem. there is a receipt for a fee of 100K. Do you know their schedule? It seemed to us fair to increase the fee as the size of the specimen increases, both on account of the increased difficult in screening, etc. and on account of the need for larger standards. We have adopted provisionally the following:- Less than one mg, $5; one mg or more, but not exceeding ten mg, $10; over ten mg, $1 per mg. These apply to sealed specimens only. I should like to know what you think of them.

I am very glad to know there is but little likelihood of radium being adulterated with mesothorium. I had been told that it is common; and the prices of mesothorium that were quoted to us last spring were about ¾ of that of radium of equal activity, this made such a state of affairs seem plausible.

Please excuse blunders in the typewriting, I'm picking this out on my own machine, and am far from an expert in the art.

With all good wishes, I remain

Very sincerely yours,

N. Ernest Dorsey

Dorsey's letter contains several strikethroughs and insertions; he does acknowledge the blunders in his typing. A few sections are underlined that are not clear. He referred in the second paragraph to an eighteen mg standard, and then crossed it out and changed it to 19 mg. In the second paragraph where he says "they tell me" he had moved that remark in the text. Presumably "they" refers to Viol and company in Pittsburgh. In the first paragraph on page two, it is not clear if he is asking whether the discrepancy is "so great as" 2%, or ½%. He probably meant 2%, but it can be read either way.

The mention of a "Hess certificate" is interesting. This refers to the Austrian physicist Victor Hess, who won the 1936 Nobel Prize for discovery of cosmic rays. Hess got his degree in Vienna and assisted Stefan Meyer at the Institute for Radium Research. The mention of "Curie and Hess standards" seems to be shorthand for the" Paris and Vienna standards". Hess travelled back and forth between the US and Europe and in 1920s was the physicist for the U.S. Radium Corporation in New Jersey.

It is interesting that Dorsey mentions his conversations with "Dr. Kelly". Howard Kelly had a private practice in Baltimore and by that time was one of the leading US physicians using radium in therapy. Partnering with James Douglas in New York, he set up the National Radium Institute under a cooperative agreement with the US Bureau of Mines (to be described later). His clinic became in time associated with Johns Hopkins University. As Dorsey was developing calibration fees, he considered what his counterparts in Germany were charging as well as the extra burden of handling larger sources.

Three-page typed letter of December 15, 1913 from Boltwood

December 15, 1913

Mr. N. Ernest Dorsey,
Bureau of Standards,
Washington, D.C.

My dear Dorsey:

Your letter of December 11th reached me on Saturday. I think the delay in the delivery of the U.S. Radium Standard is probably due to the fact that the summer vacation of the Paris people does not end until the middle of November and that they are usually away until that time. Moreover, Madam Curie has, I understand been spending some time in Russia and probably did not return until some time this month. I understand that Debierne really makes all the measurements for intercomparisons of the standards but the certificates have to be signed by the Madam. This would explain the delay. I got these facts from a Miss Gleditsch who is working here this winter and who has spent a number of years in Marie Curie's laboratory. I doubt whether there is any disagreement between results obtained in the Paris and the Vienna measurements.

What you write me about the three standards of the Standard Chemical Company is interesting and I wonder whether the explanation of the disagreement between these is not merely due to the fact that most of the specimens are measured merely for sales purposes, in which case only a moderate degree of accuracy is aimed at and the general tendency in Vienna is to supply more radium than is actually charged for rather than the opposite. In other

words, some of these preparations are not real Standards at all, any more than a kilogram of some chemical preparation purchased from a reputable dealer is considered to be a standards kilogram.

N.E.D. 2

In regard to Dr. Kelley's standard, it strikes me that this is also in all probability merely an approximate standard and that its value was determined by a rapid comparison with Rutherford's standard without any definite idea of great accuracy. Rutherford's standard is actually five per cent too low; that is to say, a given quantity of radium expressed in terms of Rutherford's standard is to be reduced by 5% in order to bring it into accord with the International Standard. The assumed value of Rutherford's standard is therefore approximately 5% too high.

As to the relation of the Standard Chemical Company's standard with mine:- The measurement that I made was merely to determine whether a certain customer was getting full value in radium for what he was paying the Standard Chemical Company. The comparison of his specimen with my standard was therefore carried out quickly and the results obtained were only approximate. They indicated that he was getting about 2% more radium than he was paying for. This was about the order of the limit of experimental error.

Now as to my "Standard"; let me give you a brief account of this. In 1910 while I was working in Rutherford's laboratory with a quantity of radium of the order of 200 milligrams, I had occasion to measure this with a fair degree of accuracy. Since Rutherford's standard is of the order of only 3.6 milligrams $RaBr_2$, a direct comparison of this standard with the large radium quantity was undesirable. So I took a known fraction of the solution of the radium salt, evaporated this to dryness and finally enclosed the resulting salt in a small glass tube. This small specimen, containing radium approximating 1.5 milligrams, was compared somewhat carefully with Rutherford's standard. When I came back to this country in 1910 I brought the specimen with me and compared it with a specimen of impure radium salt, several centigrams in weight, sealed up in a glass tube and purchased from the Brunswick

N.E.D. 3

Company. This specimen is what I have called my "Standard" and the value in radium which I assume is probably within 2% of the correct value in terms of Rutherford's standard. It isn't a very satisfactory standard but it served its use in the lack of a better one. It contains approximately 1.3 milligrams of radium and I reduce it to terms of the International Standard by making the 5% correction that I have already referred to. I doubt whether this standard would be of any particular value to the Bureau but if you should need it at any time I would be glad to lend it to you.

As to the schedule of fees which you have adopted, it seems to me that the charges for the smaller are quite reasonable, but I don't think that the rate of one dollar per milligram for quantities of over twenty five milligrams is altogether justified. It doesn't seem to me that the increased difficulty of measuring quantities of the order of 100 milligrams and more is quite as great as the increased charge would suggest, but of course, you would be the best judge of that matter and I don't think that the schedule is at all extortionate.

I hope this covers the various matters mentioned in your letter and with assurances of kind regards, I am

Yours sincerely,

(B.B.Boltwood)

There is a lot of good information in that first paragraph. It is late in the year and Dorsey wants to understand the delay in shipment of the US standard. Marie Curie was 3 years older than Boltwood and was probably a little condescending. Boltwood was a life-long bachelor and could be condescending himself. He mentions that André Debierne – Curie's colleague who discovered actinium – made all the intercomparison measurements on the standards – but the Madam signs the certificates.[9]

Two-page typed letter of December 16, 1913 from Dorsey

Bureau of Standards,
Washington, D/C.,
Dec. 16, 1913.

Prof. Bertram B. Boltwood,
Yale University,
New Haven, Conn.

My dear professor Boltwood:-

I am certainly indebted to you for the information in regard to your Standard. We were of the opinion that you probably had your standard compared directly with the International Standard. In such a case a comparison of ours with it would be of great value to us while awaiting the U.S. standard. As things stand it would probably be more advisable to make the comparison after our standard arrives. However I'm just as much obliged to you as if you had loaned me a 50 mg standard known to 1/10%.

Your suggestion regarding Dr. Kelly's standard seems very plausible. If the Vienna certificates state what they mean, your explanation of the Standard Chemical Company's standard can scarcely hold. The certificate states:-"Dasselbe wurde als 'Secondärer Standard' an den Wiener Etalons geeicht. Der γ -Strahlung nach ist im Jahre 1913 äquivalent 2.58_3 Milligram Radium-Chlorid ($Ra\ Cl_2$). (Die jährliche Abnahme Beträgt etwa 0.4 Pro-mille.) Unter Zugrundelegung de Atomgewichte von 226.0 für Radium, 35.457 für Chlor, 79.916 für Brom, entspricht dies:- 1.96_6 Milligram Radium-Element--------- Die Genauigkeit dieser Angabe wird auf zumindest 0.5 Prozent für gesichert gehalten." This certainly appears to eliminate the expectation of 2% error. After our Vienna standard comes, I expect to take the

[9] Boltwood obtained that information from Ellen Gleditsch who was with him at Yale and had previously worked in Curie's laboratory. Boltwood mentioned in a letter to Rutherford that Gleditsch had applied to Yale but was not accepted. Then he found that she had obtained a scholarship and was on a boat to New York. While at Yale, she published a paper on the half life of radium (1686 years). Boltwood was eager to share her early results with Rutherford by letter. Gleditsch was very effusive in praise of Rutherford and Boltwood for their guidance on the work (Gleditsch, 1916). She later became a professor of radiochemistry at Oslo, Norway and an international leader for women in science (Badash, 1979).

matter up with them again. If they have not found a satisfactory explanation of the discrepancy by that time, I shall endeavor to get them to send the standard to us to be remeasured.

Am glad that you think the proposed schedule of fees is not extortionate. The more I think of it the more I am of your opinion, that the fee for 100 mg, or so, is rather out of proportion; at least until we get a distinctly larger standard, and are able to do more accurate work.

Please give my regards and the season's greetings to Bumstead and Uhler.

Trusting that you may have many happy Christmases and prosperous New Years, I remain,

Sincerely,
N. Ernest Dorsey

The certificate that Dorsey quotes from at length in German has not been found in the NIST archives. It is impressive that he retyped the German text from the certificate without visible errors and cross outs. It is not clear why the atomic mass of bromine is listed in the certificate since the standard was a radium chloride.

One-page typed letter of December 26, 1913 from Dorsey

Bureau of Standards,
Washington, D.C.,
Dec. 26, 1913.

Prof. Bertram B. Boltwood,
Yale University,
New Haven, Conn.

My dear professor Boltwood,

The long expected radium standard has at last arrived. We received it last Saturday. A preliminary comparison with it indicates that our 1 mg specimen is 1 /2 % larger than the value certified by the Standard Chemical Co. The 4 mg is about 1 % larger. Next week I hope to get other comparisons. Shall let you know if anything especially interesting shows up.

With best wishes for the New Year, I remain

Yours truly,
N. Ernest Dorsey

Christmas day in 1913 was on a Thursday. Dorsey had received the Standard the previous Saturday and made measurements that week to compare Standard No.6 with the two specimens from Standard Chemical Co. He wanted to notify Boltwood as soon as he had made measurements to confirm that everything seemed in order.

He must have been writing in haste on Friday December 26 because this is the first instance where he failed to write out Company – and abbreviated with Co.

3.5. US Secondary Standard Number 6

Dorsey was understandably excited about having opportunities to make measurements with the international standard. That last week in December of 1913 he started a small notebook – later contaminated, but still available – and a transcription of the first page of hand-written notes is given here:

Page 1
Ra6
15.44 mg

Description of Standards

Primary Standard. Known as "Secondary Standard No.6" and hereafter referred to as "Ra 6". It was bought from the Austrian Government. The Dept. of State was requested on March 29, 1913 to arrange for the purchase. The specimen was received Dec. 20, 1913.

It was prepared under the auspices of the "Commission des Internationale Etalons de Radium" from the pitchblende of St. Joachimstal. It is certified by the Commission to contain 21.50 mg of radium chloride, was sealed July 1,1913 in a tube of Thuringian glass 0.27 mm wall thickness, exterior diameter 3.2 mm, length 22 mm; a thin platinum wire is fixed into one end of the tube. Γ- ray measurements at Vienna and Paris give 20.28 mg $RaCl_2$ in 1913. If the atomic weights are 226 for Ra, 33.459 for Cl, 79.916 for Br. This corresponds to 15.44 mg Ra element

20.28 mg $RaCl_2$
26.36 mg $RaBr_2$

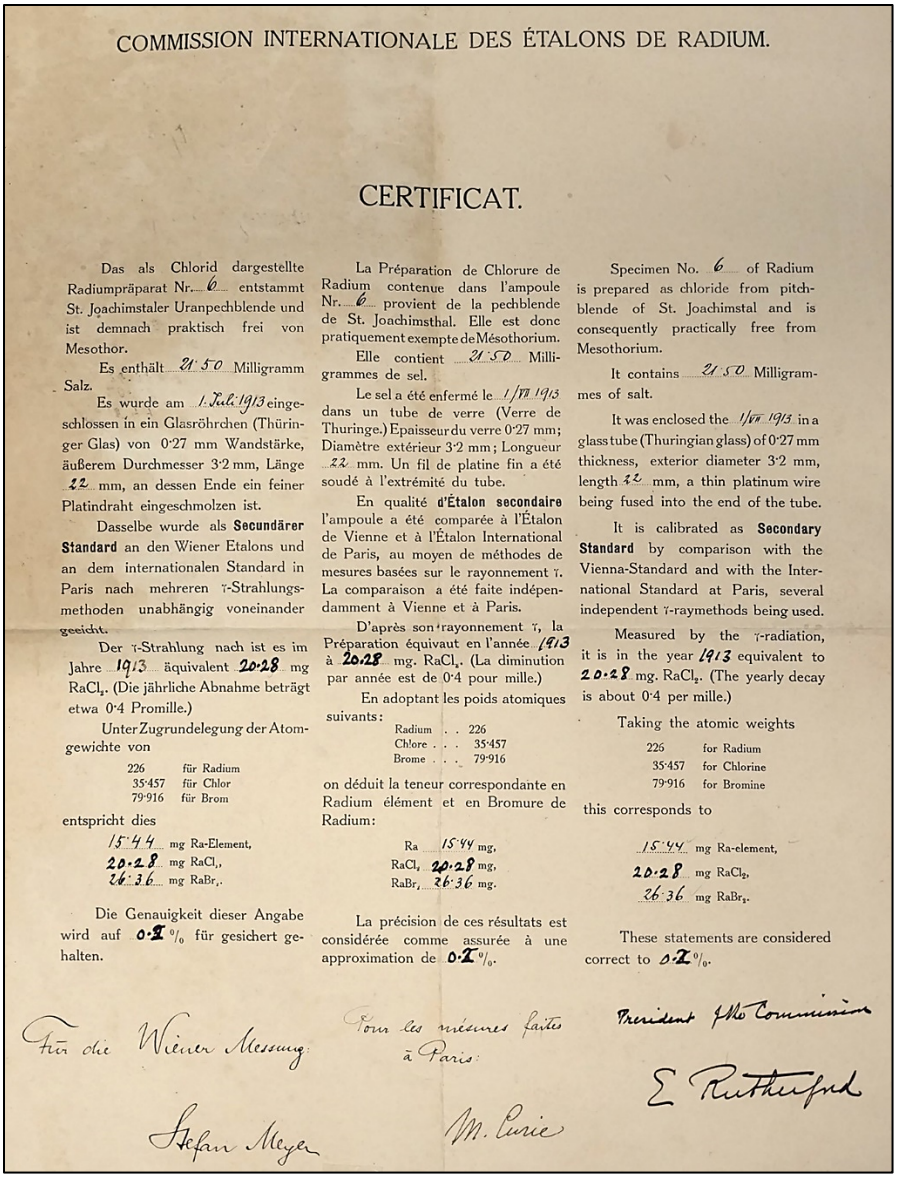
"These standards are considered correct to 0.2%"

Bureau of Standards Description is as follows.

Tube---- length 22.4 mm, to lip of wire 22.74 mm

Diameter wire enc(luded) Min 3.59 mm Max 3.60 mm

The particulars in Dorsey's notebook match the information on the certificate provided by the International Radium Standards Commission. But these notes are the only indication that the US Department of State actually paid for the standard with funds to the Austrian government. The actual certificate provided for Secondary Standard No. 6 is shown here.



COMMISSION INTERNATIONALE DES ÉTALONS DE RADIUM.

CERTIFICAT.

Das als Chlorid dargestellte Radiumpräparat Nr. 6 entstammt St. Joachimstaler Uranpechblende und ist demnach praktisch frei von Mesothor.

Es enthält 21·50 Milligramm Salz.

Es wurde am 1. Juli 1913 eingeschlossen in ein Glasröhrchen (Thüringer Glas) von 0·27 mm Wandstärke, äußerem Durchmesser 3·2 mm, Länge 22 mm, an dessen Ende ein feiner Platindraht eingeschmolzen ist.

Dasselbe wurde als **Secundärer Standard** an den Wiener Etalons und an dem internationalen Standard in Paris nach mehreren γ-Strahlungsmethoden unabhängig voneinander geeicht.

Der γ-Strahlung nach ist es im Jahre 1913 äquivalent 20·28 mg $RaCl_2$. (Die jährliche Abnahme beträgt etwa 0·4 Promille.)

Unter Zugrundelegung der Atomgewichte von

226 für Radium
35·457 für Chlor
79·916 für Brom

entspricht dies

15·44 mg Ra-Element,
20·28 mg $RaCl_2$,
26·36 mg $RaBr_2$.

Die Genauigkeit dieser Angabe wird auf 0·2 % für gesichert gehalten.

Für die Wiener Messung:
Stefan Meyer

La Préparation de Chlorure de Radium contenue dans l'ampoule Nr. 6 provient de la pechblende de St. Joachimsthal. Elle est donc pratiquement exempte de Mésothorium.

Elle contient 21·50 Milligrammes de sel.

Le sel a été enfermé le 1/VII 1913 dans un tube de verre (Verre de Thuringe.) Epaisseur du verre 0·27 mm; Diamètre extérieur 3·2 mm; Longueur 22 mm. Un fil de platine fin a été soudé à l'extrémité du tube.

En qualité **d'Étalon secondaire** l'ampoule a été comparée à l'Étalon de Vienne et à l'Étalon International de Paris, au moyen de méthodes de mesures basées sur le rayonnement γ. La comparaison a été faite indépendamment à Vienne et à Paris.

D'après son rayonnement γ, la Préparation équivaut en l'année 1913 à 20.28 mg. $RaCl_2$. (La diminution par année est de 0·4 pour mille.)

En adoptant les poids atomiques suivants:

Radium . . 226
Chlore . . . 35·457
Brome . . . 79·916

on déduit la teneur correspondante en Radium élément et en Bromure de Radium:

Ra 15·44 mg,
$RaCl_2$ 20·28 mg,
$RaBr_2$ 26·36 mg.

La précision de ces résultats est considérée comme assurée à une approximation de 0·2 %.

Pour les mesures faites à Paris:
M. Curie

Specimen No. 6 of Radium is prepared as chloride from pitchblende of St. Joachimstal and is consequently practically free from Mesothorium.

It contains 21·50 Milligrammes of salt.

It was enclosed the 1/VII 1913 in a glass tube (Thuringian glass) of 0·27 mm thickness, exterior diameter 3·2 mm, length 22 mm, a thin platinum wire being fused into the end of the tube.

It is calibrated as **Secondary Standard** by comparison with the Vienna-Standard and with the International Standard at Paris, several independent γ-raymethods being used.

Measured by the γ-radiation, it is in the year 1913 equivalent to 20·28 mg. $RaCl_2$. (The yearly decay is about 0·4 per mille.)

Taking the atomic weights

226 for Radium
35·457 for Chlorine
79·916 for Bromine

this corresponds to

15·44 mg Ra-element,
20·28 mg $RaCl_2$,
26·36 mg $RaBr_2$.

These statements are considered correct to 0·2 %.

President of the Commission
E. Rutherford

Figure 5. Certificate for US national standard of radium-226.

The total mass of salts in the standard is 21.50 mg, which indicates about 6% of the mass represent residual compounds from the purification of the radium chloride. The equivalent activity of the standard as a radium bromide is given in case the standard is to be compared to a bromide such as those produced by the Standard Chemical Company. The certificate was folded and contained in a hand-addressed envelope. Both are contaminated with radium-226. The darker quadrant of the certificate has the most activity. It is not possible to say when or where it became contaminated, but, because of its critical importance, it was stored with many of the other early records and likely resulted in slight contamination of them as well.

It is interesting that the US standard was identified as Secondary Standard No. 6 although it is listed as the fourth standard distributed by the Commission in Table 8. This puzzle is answered by reference to a much later paper by Stefan Meyer (Meyer, 1945). Meyer's later table lists 30 standards prepared and measured in Vienna up to 1936. This table lists No. 4 as 24.08 mg of

radium chloride and No. 5 as 2.58 mg of radium chloride. These two were not measured in Paris but were purchased separately in 1912 or 1913 from the Austrians. No. 4 went to W.L.S. Alton the director of the London Radium Institute. Both Soddy and Ramsay were associated with that institute. The smaller standard No. 5 went to the Standard Chemical Company in Pittsburgh (see Dorsey's letter above of December 16, 1913).

3.6. US Radium Industry standards

Table 6 lists several corporations that were providing radium materials for medical and research customers. There are excellent, detailed references to the early radium industry including: Badash, 1979; Landa, 1982, 1987; Robison, 2015 and Rentetzi, 2008, 2022. A most valuable reference is the 2019 book **Radium City** (Lubenau and Landa 2019) recounting the role of Pittsburgh in the production and uses of radium. The present account will focus on only two companies, the Standard Chemical Company of Pittsburgh (Flannery, 1915) and the National Radium Institute in Denver, with special attention to their interactions with Dorsey at NBS.

3.6.1. Standard Chemical Company of Pittsburgh

The origins and operations of the Standard Chemical Company are described in detail in **Radium City** (Lubenau and Landa 2019). What follows is a brief summary of the excellent scholarship from Lubenau and Landa. In the early century the Flannery brothers had developed a very successful corporation for production of vanadium for use in steel alloys for the railway industry. They turned their attention to radium because of their hopes to cure a sister's cancer. In January of 1910 Joseph Flannery went on a 5-week trip to Europe and met Rutherford, Meyer, Hahn and Curie. On August 30, 1910, Flannery wrote to Boltwood at Yale asking for his assistance on radium chemistry. Boltwood recommended a young Yale chemist Roland S. Bosworth, and in June of 1911 Boltwood and Bosworth worked out extraction methods for carnotite and estimated some costs. (In July 1911 Boltwood sailed for England to spend a sabbatical year with Rutherford at Manchester.) Late in 1912 the Austrian chemist Otto Brill arrived in Pittsburgh from the Institute for Radium Research in Vienna. In that same year the Standard Chemical Company hired Charles Viol a student of McCoy's at Chicago to assist Brill. They soon added a third chemist, Glenn Kammer, who received his degree from University of Pittsburgh (Silverman, 1950). Early in 1913 Brill wrote to Boltwood asking for assistance on "standards". In May of 1913 Brill left for Vienna with a stop in Paris for Marie Curie to calibrate a one milligram "dab" of radium. In September of 1913 Brill was recalled to Vienna by the Austrian government. Boltwood reported that in November of 1913 he had a standard submitted to him from Pittsburgh and he found it in good agreement with his own (Boltwood to Dorsey, Dec. 1913 (Badash, 1969)). Owing to his experience with the Austrians, Brill provided considerable value to the project in Pittsburgh, but he was difficult to work with, and the new team in Pittsburgh; Charles Viol, Glenn Kammer and another chemist from the University of Pittsburgh, Henry Titus Koenig (Silverman, 1934), developed the extraction chemistry for a successful large industrial plant. Rentetzi provided an account of their work:

"Viol, Kammer and Koenig directed the final delicate operation of fractional crystallizations. These occurred in small, open porcelain pans in the company's chemistry laboratory. The end product, a tiny pinch of nearly pure radium bromide, was ready five weeks later." (Rentetzi, 2008)

In the Summer of 1913, the Standard Chemical Company received three standards from Europe. One, from the Institute for Radium Research, the Secondary Standard No. 5, was referred to in Dorsey's letter of December 16, 1913. It contained 2.58 mg of $RaCl_2$ (1.966 mg radium element). Two other sources were prepared in Pittsburgh and submitted for calibration in Paris and Vienna. The certified values of these two were 5.83 mg (from the Curie Laboratory) and 19.24 mg (from the Institute for Radium Research) radium element. These arrived in time for Dorsey's visit to Pittsburgh. He had reported his favorable opinion of the Pittsburgh scientists in his letter to Boltwood on December 1, 1913. *"The men I met there seemed to be very fair kind of fellows."* Boltwood commented on Dorsey's assessment of the Pittsburgh operations, which seems to indicate that he had not been in close contact with them in the past two years.

Perhaps the best perspective on the radium standards from the Standard Chemical Company comes from Charles Viol's article in the journal Radium in September of 1913.

> "It is of historical interest to note that the first pure radium salt prepared in the United States from Colorado carnotite ores, has been made by the Standard Chemical Company, in its Radium Research Laboratory in Pittsburgh, Pa. Recently this Company delivered to a well known Baltimore, (Md.) surgeon, one hundred milligrams of radium element in the form of anhydrous radium chloride. The salt, weighing 133 milligrams, was sealed in two tiny glass tubes, each 15 mm. long and 2.4 mm . in diameter. The radium content of this material was determined by a comparison with radium standards by the gamma ray method. The Radium Research Laboratory of the Standard Chemical Company has three radium standards; one prepared and standardized by Professor Stefan Meyer, director of the Radium Institute of the Imperial Academy of Sciences at Vienna, another standardized in Madame Curie's laboratory at Paris, and a third which was also standardized at the Vienna Radium Institute. These standards contain 1.966, 5.83 and 19.24 milligrams of radium element, respectively. These measurements are based on the International Radium Standard which was prepared by Madame Curie, a prototype of the International Standard being in the possession of the Vienna Academy of Sciences." (Viol, 1913)

Viol amplified on these comments in his Congressional testimony in January of 1914 (Viol, 1914):

> Congressman Taylor of Colorado. Does your company give a guaranty?
>
> Dr. Viol. We have these two standards, and then I have a third here. This is a secondary standard prepared for us in Vienna by Prof. Meyer. He, by the way, prepared all of these. He prepared the standard for the National Physics Laboratory in London. He prepared the standard for our own Bureau of Standards here, and he prepared this standard and standardized it for us.
>
> This little tube, after making it up, we sent to Mme. Curie's laboratory, where it was checked up against her standards. This other tube, containing about 19.5 milligrams of radium element, was sent to Vienna. Prof. Meyer, not happening to be in town, his first assistant, Dr. Hess, compared it with their standards there. So the Standard Chemical Co. is in possession of three standards which go right back to either Vienna or Paris, where the original standards were make. All the preparations that leave the laboratory of the Standard Chemical Co. are compared against these standards by suitable methods, and we absolutely guarantee the quantity of radium element in these standards.

> I have here a framed copy of the certificate which we send out with every preparation which leaves the laboratory. In that certificate we make two statements: there is such and such a weight for example, in this tube there are 22.4 milligrams of radium salt, containing 5.9 milligrams of radium element. That is really the vital point. And we unreservedly guarantee our preparations. If there is any error in measurement, as might possibly happen, we make it good. We either send a check for the difference, or take the tube back and make it up. So there is no question in regard to the preparations put out by the Standard Chemical Co. They are full weight and guaranteed.
>
> Mr. Howell. How would a person having a gram of radium and desiring to test it proceed to do so? Where would he apply?
>
> Dr. Viol. Our own Bureau of Standards, here in Washington, is in a position to make these comparisons. Just as if you buy a meter stick or a yard stick, you can send it to the Bureau of Standards and have it checked up.
>
> Mr. Howell. That is the only place in this country?
>
> Dr. Viol. They are the only people in this country, I should say, who have the standards carefully enough standardized so we would be willing to take their word. There are a lot of so-called standards. A man might buy a preparation from us and then undertake to make measurements. Well, we would be inclined to believe his figures, but his preparation is not as carefully calibrated as a standard must be.
>
> Mr. Taylor of Colorado. Where did the Bureau of Standards learn to make these measurements, and who taught them?
>
> Dr. Viol. The gentleman who makes the measurements at the Bureau of Standards had his introduction into this work at our laboratory in Pittsburgh. It came about in this way: The Government ordered some radium from the Austrian Government for a standard a year ago in January or February, and along about May the material was not forthcoming, and they got a little restless. So several months later they came to us and ordered some radium and asked whether we would permit the physicist to come down and see how we made the measurements and what our facilities for that work were. We gladly assented, and Dr. Dorsey, of the Bureau of Standards, came down and spent three or four days in our laboratory, where he was shown all these points in connection with the measurements of the standards, and we together went over the measurement of the tubes which we were preparing for them. Since then the Bureau has received from Austria the tubes which Prof. Meyer prepared and which were calibrated for the committee on the international radium standard."

There is little information about the physicists who directed the calibrations work at the SCC. Lester Walker signed as physicist the 1913 certificates for NBS (Figure 4). Lubenau provides this note on the physicist Arthur Miller: "In 1914, after graduation from Purdue University, Arthur L. Miller accepted an offer to work for the Standard Chemical Company, in Pittsburgh, Pennsylvania, which was then the largest producer of radium. There, he specialized in calibrating radium sources using an electroscope. Since he was familiar with the operation of electroscopes, he was frequently called upon to search for lost radium sources using that instrument." (Lubenau, 1999).

Figure 6 and Table 9 show the relationships between the standards in the possession of the Standard Chemical Company of Pittsburgh and those exchanged between the metrology institutes in Paris, Vienna, London (NPL) and the US (NBS). Viol was confident that the SCC standards would be in good accord with those of NBS.

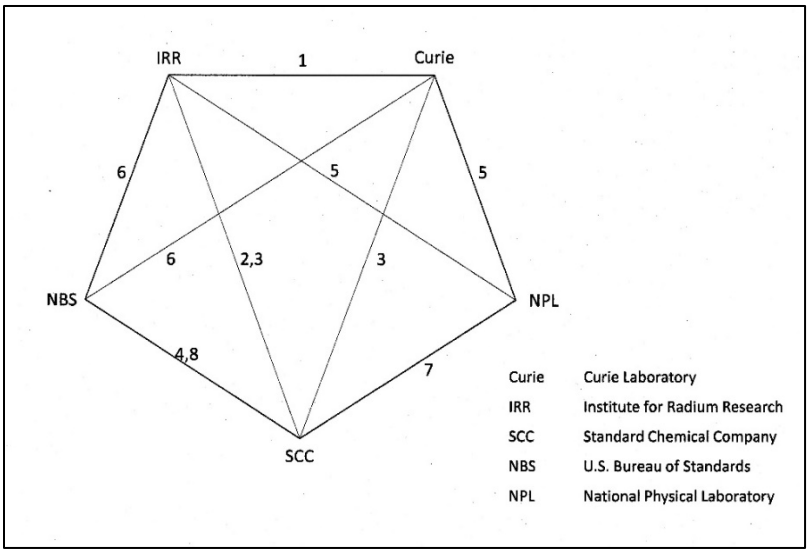


Figure 6. Exchanges of 14 radium-226 standards between key national and commercial laboratories in period 1912 – 1914 (see Table 9 below).

Table 9. Numerals in Column 1 refer to Figure 6. Certificates sometimes presented the mass of radium element, and sometimes mass of the radium chloride. Where both were specified, they are both listed here. The first two rows show the comparison of primary standards in Paris in March of 1912. All the other values shown in the table were based on gamma-ray comparisons by electroscope with primary standards or secondary standards.

	Laboratories	**Date**	**Std. No.**	**Mass Ra (mg)**	**Mass $RaCl_2$(mg)**	**Reference**
1	Curie - IRR	Mar. 1912			21.99	Curie, 1912
	IRR - Curie	Mar. 1912			31.17	Meyer & Hess, 1912
2	IRR - SCC	Summer 1913	No. 5	1.966	2.58	Meyer, 1945, Viol 1913
3	SCC - Curie	May 1913		0.96		Lubenau & Landa, 2019
	SCC - Curie	Summer 1913		5.83		Viol, 1913
	SCC - IRR	Summer 1913		19.24		Viol, 1913
4	SCC - NBS	Sep. 1913		1.04		NIST archives, 2023
	SCC - NBS	Sep. 1913		4.10		NIST archives, 2023
5	IRR - Curie - NPL	Jun. 1913	No. 3	16.08	21.13	Smith, 1975; Meyer, 1945
6	IRR - Curie - NBS	Dec. 1913	No. 6	15.44	20.28	Meyer, 1945
7	SCC - NPL	Dec. 1913		5.3		Smith, 1975
	SCC - NPL	Dec. 1914		27.7		NIST archives, 2023
8	SCC - NBS	Feb. 1914		0.615		NIST archives, 2023
	SCC - NBS	Feb. 1914		0.55		NIST archives, 2023

3.6.2. The National Radium Institute (NRI) Denver

A second major player in the US radium industry during this period was the National Radium Institute (Parsons,1913; Parsons, et al. 1915). This institute represents a short but colorful chapter in the US radium history. The NRI was for a few years at the center of discussions on the US radium industry, medical therapy and policies of the federal government. The overlapping details of these discussions have been described in detail by Badash, Rentetzi and Slaughter (Badash, 1979; Rentetzi, 2008,2022; Slaughter, 2013). Rentetzi and Robison (Robison, 2015) describe this alliance between two prominent, wealthy medical doctors, James Douglas of New York (Langton, 1940) and Howard Kelly of Baltimore. They proposed to partner with the federal government to develop a counterpart to the Austrians (Vienna Institute founded in 1910) and the English (London Radium Institute founded in 1911) with a US institute that would have government subsidies. Kelly was at the time purchasing radium from European suppliers, and he argued that this radium came from Colorado carnotite, and the US should not be dependent on foreign suppliers for radium that was so important for medical uses in the US. Kelly and Douglas found willing collaborators in the US Bureau of Mines. “In 1913, U.S. Secretary of the Interior Franklin Lane announced that three-fourths of the radium produced in the world during 1912 came from American ores” (Rentetzi, 2008). The chemists at the US Bureau of Mines, Charles Parsons and Richard Moore, were very enthusiastic in their support for the proposal. The Chief Chemist Moore had worked with Ramsay in London at about the same time as Brill (Rentetzi, 2008). Their publications describing the cooperative agreement with Kelly and Douglas provide extensive details on establishment of the NRI and the technical activities over the next few years (Parsons, 1913, Parsons et al. 1915).

The Congressional hearing of the Committee on Mines and Mining in January of 1914 provided an opportunity for Kelly of the NRI to make the case for a federal institute to administer mining and radium production in the US. At the same hearings Flannery and Viol objected to a state monopoly on radium (Rentetzi, 2008), that is, government intervention in the marketplace. There were never any federal appropriated funds for the NRI, but Kelly and Douglas provided their own funds ($75,000 each, $2.3M in 2023 USD) to begin mining in Western Colorado and establish a facility for extracting radium from the ores in Denver, Colorado. Under the cooperative agreement the first 7 grams were to go to hospitals directed by Kelly and Douglas, with the remainder to go to the USBM for experimental purposes (Rentetzi, 2022). The USBM provided critical scientific support at costs. They summarized the collaboration in an excellent report: *“Extraction and recovery of radium, uranium and vanadium from carnotite,”* US Bureau of Mines Bulletin 104. Parsons and Moore added the two academic chemists Samuel Lind and Herman Schlundt to their group in Denver during the key operations of the NRI in 1913 – 1915. Lind was on the USBM staff in Denver at the time having just returned from a year with Curie in Paris and Meyer in Vienna. Schlundt from Missouri joined them during the Summer of 1914 (Parsons et al. 1915). By October of 1915 they claimed to have delivered 2.5 grams of radium element in the form of radium bromide to hospitals. As part of Kelly and Douglas’s agreement with the NRI, seven grams of radium would go to hospitals in New York and Baltimore. Burnam reported that approximately 8 grams were delivered to the two hospitals by January of 1917 (Burnam, 1936). This material was delivered to Kelly’s clinic in Baltimore and the General Memorial Hospital for Treatment of Cancer and Allied Diseases in New York, forerunner of the present Memorial Sloan Kettering Cancer Center (Badash, 1979). The NRI closed in April 1917;

the Denver plant was sold in 1919 to the Minerals Recovery Corp. of Denver (Historic Structures, 2023).

The scientific aspects of the NRI work in Denver are particularly valuable, because the government chemists from the USBM provided many technical details on radium chemistry and measurements that were not available from NBS at the time. In 1913 -1915, Moore, Lind and Schlundt had the expertise from their work in the European laboratories, excellent resources from the government, and the impetus to publish their work to promote the institute. They describe essentially the same three methods for measurement of radium as Randall had listed previously (Randall, 1912): alpha-particle measurements of solid powders, emanation measurements, and comparative gamma-ray measurements (Parsons et al. 1915). They included a good description of use of the Lind Electroscope that Lind had published separately during the same year (Lind, 1915). For the gamma-ray measurements, they describe their standard of 10.56 mg radium element that had been compared to the 15.44 mg standard at NBS. There is no record at NBS of this calibration, but there was a close collaboration between the two federal agencies. The NBS Director's Annual Report June 1917 noted *"A 49-milligram tube of radium which was loaned by the Bureau of Mines has been of great assistance in the routine testing during the year as well as in the study of the methods of testing. This tube and a second tube containing about 91 milligrams of radium have been permanently transferred by the Bureau of Mines to the Bureau of Standards, and constitute a very valuable addition to the Bureau's equipment for radioactivity measurements. The radium contained in these two tubes has a market value of about $14,000."* (Directors Annual Report, 1917). It is probably not a coincidence that the two large sources (valued at $336K in 2023 USD) were permanently transferred to NBS at the same time that the USBM concluded their three-year cooperative agreement with the NRI. The NBS Director's reports from previous years had complained that the Radium Section needed a standard of higher mass to allow them to make electroscopic comparisons with the larger samples then being submitted for analysis. This led to the initial loan of the 49 mg source from the USBM.

On the East Coast, the major medical centers were collaborating with medical physicists (new hires and local academicians) to assist the physicians in handling and calibrations of radium sources. Slaughter's thesis at the University of Minnesota, and Robison's book on mining and selling of radium, are the best resources on the radium measurements at the Howard A. Kelly Hospital (Kelly Clinic) in Baltimore, which was the forerunner of the therapeutic radiology department at Johns Hopkins Medical School (Landa, 1987, Slaughter, 2013; Robison, 2015). Kelly was one of the four medical doctors who founded the medical school at Johns Hopkins. Slaughter's account draws in part from the recollections of Curtis Burnam, the chief physician of the Kelly Clinic, who joined the practice in the Spring of 1911 (Burnam, 1936). Kelly had made several purchases of radium in Europe and in 1911 reached out to Boltwood at Yale for assistance in measuring a 100 mg source from a supplier in France. (Burnam recalled that NBS did not yet have a standard available.) Boltwood reported that the sample was 40 mg short, and Kelly asked for and received the missing amount based on Boltwood's assay. In 1912 Kelly travelled to Europe and met many physicians and scientists including Soddy and Rutherford. Rutherford told Kelly "physicians working with radium must have a physicist" and recommended one of his assistants at Manchester, Walter Lantsberry. Lantsberry wrote to Kelly in March of 1914 offering his assistance on measuring some of the Baltimore radium, and subsequently moved to the US to work for Kelly and Burnam. Burnam reported that in 1912 he made visits to Paris, Vienna and Pittsburgh trying to arrange to purchase as much as a gram of radium. He was able to buy 750 mg of radium element in the form of almost pure radium

chloride from the SCC. (See Viol's reference to sales to a "well-known Baltimore MD" in September 1913 (Viol, 1913). This was packaged in 50 mg glass tubes. "A year later" (presumably 1914) Burnam had the measurements rechecked by Dorsey at NBS who reported that they had received about 2% more radium than the SCC assay reports. It is interesting to note that the driver for an accurate assay was the transaction cost rather than the patient dose. Dorsey had mentioned to Boltwood his own discussions with Dr. Kelly in his letter of December 11, 1913.

Douglas, the wealthy businessman in New York did not have the hands-on approach of his partner Kelly in Baltimore (Langton, 1940, Landa, 1987, Robison, 2015). The calibrations of the radium at Memorial Hospital, and those in Boston, were handled by two distinguished physicists who studied with Marie Curie, William Duane and Gioacchino Failla (see Table 4). There is no information in the NBS/NIST archives on the early calibrations for the New York hospitals. But their critical work on emanation plants was coordinated with NBS, and that will be described later.

3.7. Beginning of the NBS Radioactivity Calibrations Program: 1914

By the Spring of 1914 NBS was able to offer calibrations of submitted radium by gamma-ray comparisons with the 15.44 mg international Secondary Standard. The timing proved excellent as Sir Ernest Rutherford from Manchester paid a visit to Washington and delivered two lectures (Coursey, 2018). The first was given on the general topic of radioactivity at the Smithsonian Institution. On Friday and Saturday April 24, 25, NBS arranged a special session of the American Physical Society and the American Institution for Electrical Engineers (later the IEEE). Rutherford gave a talk "On x-ray and gamma-ray spectra". During the session on Saturday Dorsey gave a talk on comparisons of samples of radium salts. Presentation materials from those two talks are not available, but Dorsey likely reported on his gamma-ray comparisons of the two SSC radium sulfate specimens with the US secondary standard. A manufacturer's exhibit at NBS for this session probably included radium samples on display from the SCC.

The nature of the radium industry was soon to change with the outbreak of World War I during the Summer of 1914. The evolution of the US program in radioactivity over the following two years is evident from the Boltwood-Dorsey correspondence (Yale, 2023). Their six letters in 1914 -1915 are reproduced here. The tone of the correspondence changes after the US standard is in place at NBS. Boltwood is now requesting assistance with calibration of his own radium, and they continue discussions on the radium industry as well as technical details of measurements.

3.8. Boltwood – Dorsey Correspondence: 1914 – 1915

Typed letter of November 19, 1914 from Dorsey

Bureau of Standards,
Washington, D.C.,
Nov. 19, 1914.

Prof. B. B. Boltwood,
Yale University,
New Haven, Conn.

My dear Prof. Boltwood:-

Shall be most pleased to measure up your standard to the best of my ability at any time. However, if a few weeks later would be the same to you, I would suggest that you send it about the middle of December. Just now my newly acquired assistant is on leave, and things are a little crowded; never the less, if you wish it done now, send it along and I will work it in.

I understand that a fee is required for all "certificates"; if you do not especially desire a formal certificate, I am sure we can give, gratis, to you an informal report of the measurement.

I believe that your standard contains about 2.1 mg. Ordinarily I would compare it with one of our small tubes, but in the present instance it seems desirable to compare directly with our 15.44 tube. As the ratio is rather large it is very desirable to repeat the measurements on two or three days. Experience shows that this repetition is desirable, but I'm not sure why.

Last June I put in solution a salt which by gamma measurement was found to contain 0.610 mg, and from this prepared a more dilute solution. The concentrations of these solutions are 1.22 micrograms per cc and 12.2 micrograms per cc, respectively. Can we arrange in some way to obtain a comparison of one of these solutions with yours? I understand that your solution was obtained from Rutherford, and so, through his standard, is known in terms of the International Standard.

What do you think of the possibility of cutting loose from standard solutions, making the measurements depend upon the electrical units?

Regards to Bumstead and Uhler.

Very sincerely,

N. Ernest Dorsey

After 18 months of work, Dorsey reports that he has a "new assistant". He proposes to compare the Yale standard of 2.1 mg "with our 15.44 tube." That obviously refers to Secondary Standard No. 6. The jargon is surprising and is yet another indication that Dr. Dorsey is no longer overly ingratiating in his correspondence with Professor Boltwood. He probably has little control over format and signatures for certificates issued by the Bureau. He could provide the results to Boltwood informally by letter, but an official Certificate would be signed by the Director. He

refers to chemistry performed last June – that would be June 1913 – when he opened radium salts and prepared quantitative dilutions. Dorsey was not a chemist and opening 610 microcuries of radium and dissolving it probably spread contamination from the radon and progeny in the laboratory. Dorsey seems keen to compare the US standard with that of Rutherford's Manchester laboratory. The provenance of Rutherford's standard (in late 1914) with respect to the International Standard is not clear. Secondary Standard No. 3 had been in the UK for a year, but Rutherford would have to make arrangements for it to be compared in the NPL laboratories in Teddington.

The question in the final paragraph is quite interesting. It is not clear what Dorsey meant by "cutting loose from standard solutions and making the measurements depend on the electrical units". The trained chemists could obviously assign massic activity based on quantitative dilutions – as Boltwood had already done in Manchester. Any measurements in electrical units, picoamperes per mg, for example, would depend on many factors including the encapsulation and the efficiency of the apparatus, as described by Randall (Randall, 1912).

Typed letter of December 29, 1914 from Dorsey

Bureau of Standards,
Washington, D.C.,
Dec. 29, 1914.

Prof. B. B. Boltwood,
Sloane Laboratory
Yale University,
New Haven, Conn.

My dear Prof. Boltwood:-

During this holiday season our messenger service is rather disorganized, as a result the radium tube did not reach me until late this afternoon. It seems to be in good condition. Owing to its length I am afraid that I can not certify it to a higher accuracy than 1 /2%, but will do the best I can. As I have arranged to be on leave for a few days it is probable that we can not return the tube until some time in the second week of January.

Do you wish the tube returned by registered mail? We usually send them by express, insured at $120.00 per mg. If we send it by mail it will be franked, so I am returning the stamps. That is I think I am enclosing the proper number: the letter is at the Bureau and I am writing in my room, so there may be some mistake in the number. If there is I will correct it later.

Please give my good wishes for the New Year to Bumstead and Uhler. With very best wishes for the coming year, I remain

Yours very truly,

N.E. Dorsey

It is not clear from the typescript whether Dorsey is estimating the limit of uncertainty in the certification of 1 % to 2%. It might be read as ½ %. Express mail delivery between New Haven and Washington seems to have been very efficient in 1914. It isn't clear what Dorsey means by returning the stamps. Perhaps Boltwood had prepaid for the return shipment. It seems that Dorsey intended to take a week off and then measure the Yale standard and return it "the second week" of January.

Three-page typed letter of January 26, 1915 from Dorsey

Department of Commerce
Bureau of Standards
Washington

NED-PFD

Address reply to

Bureau of Standards January 26, 1915.

Prof. B. B. Boltwood,
Yale University,
New Haven, Conn.

My dear professor Boltwood:

I fear you think that we intend to appropriate your standard. I had fully expected to get it off not later than the middle of the month. But several things combined to cause delay. However, the tube is now packed and sealed, and is merely awaiting the routine red tape procedure attendant upon the return of tests.

As the tube is longer, for its contents, than any tube previously measured under more than commercial requirements, it seemed desirable to make a new survey over the two positions at which measurements were made. This was done over a range of ±4 cm from the position occupied by the center of the tube. By least squares the quintic which best represented these observations was determined; the integration of this gives the correction for the length of the tube. It was here that an unexpected, and considerable delay occurred. A full set of these observations were taken by an assistant, and

(2)

on their face appeared to be good; but when I reduced them preparatory to my report it was found that a series which should have checked with a former survey did not do so. I then repeated the entire work myself; the results were very satisfactory. I have been unable to ascertain the cause of the discrepancy mentioned. Only the one series seems to be affected. Enclosed you will find the final graphs, and along each are plotted the corrections as independently deduced from the two absolutely independent surveys.

Assuming the effective length of your tube to be 62 mm the corrections as given by the curves A and B are 0.75 % and 1.21% respectively. (When the correction for the standard is also applied).

The observations at A were against our 15.44 mg standard, and those at B against our 1.052 mg. Three independent sets of measurements were made at A, and two at B. The first of the A sets was rather short and was made by the assistant; the other four were made by myself and were of normal length. The following values were obtained:

	A		B
		wt	
	1.283	1	1.290
	1.297	2	1.287
	1.298	3	
	1.295		1.288
Correction for length	10		16
	1.305		1.304

Had we weighted all of the A sets alike their mean would have been 0.15% less; had we ignored the first set the mean would have been 0.23% greater than that given.

The effect of the brass tube was determined from sets of drifts obtained with the glass tube alternately in and out of the brass tube. The reduction which the brass tube produces in the radiation entering the electroscope was found to be 1.75; 2.02; 3.19; 1.78; 3.26; the mean is 2.00%.

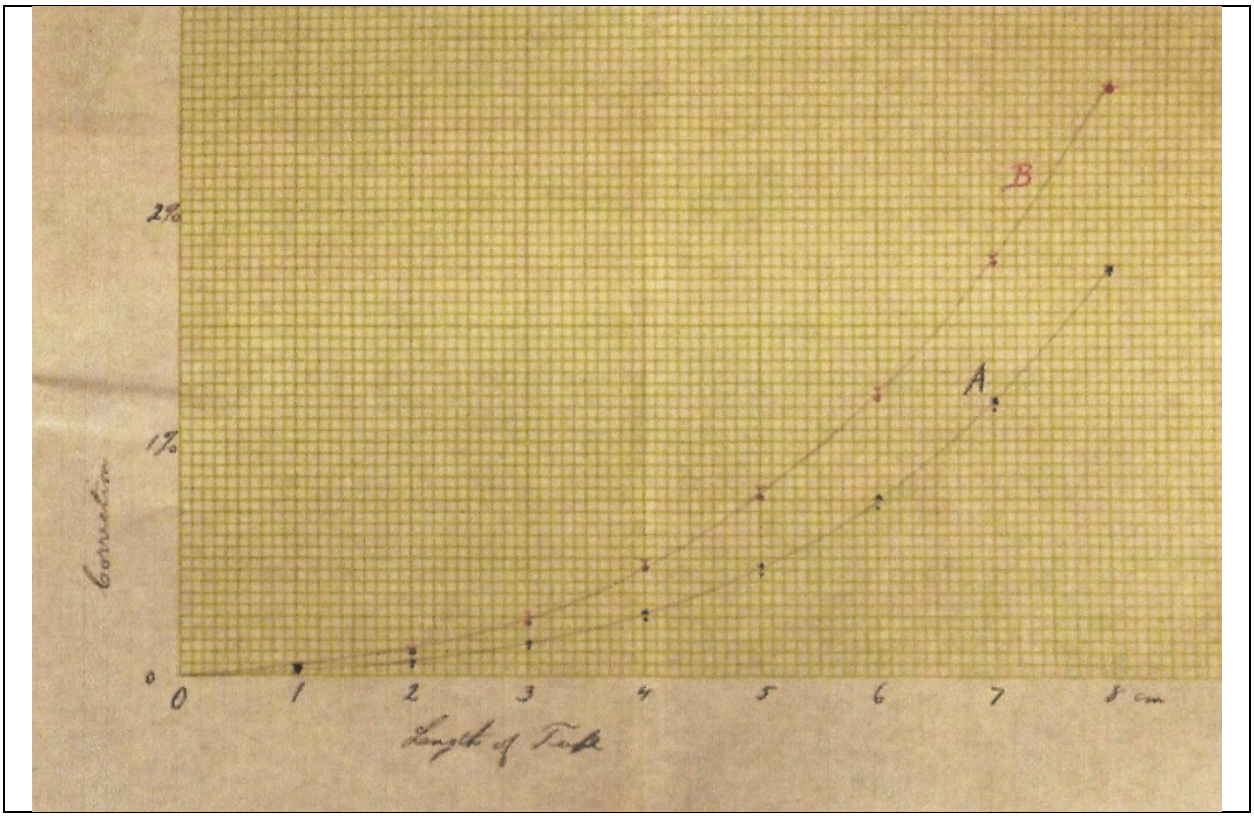
The measurements were made with the cubical electroscope made of 1 /8 inch lead; and the radiations were filtered through a large plate of 0.5 inch lead placed in close to the electroscope. The tube was placed to the side of the electroscope in a carriage that can be rotated about a vertical axis. By reversing the carriage any lack of symmetry in the line of sight was eliminated.

Trusting the that the tube may arrive safely, I remain,

Yours truly,

N. Ernest Dorsey

Enc: Graphs



This is the first of Dorsey's correspondence where he has a secretary (PFD) type the letter on official Department of Commerce letterhead. Dorsey made some hand corrections to the corrections listed on page 2. He struck through and altered the corrections (0.81% to 0.75%, and 1.32% to 1.21%) for the curves A & B. There was not a certificate for this calibration of the Yale standard. The correction for the attenuation in the brass tube is an average of five values. On the typescript on the copy in the archives it is difficult to distinguish between 2 and 3. There must have been a transcription error in that the mean value cannot be 2.00% or 3.00%.

To accommodate the smaller mass of the Yale specimen, Dorsey made measurements against Standard No. 6 (15.44 mg Radium) and a smaller NBS standard of 1.052 mg. The latter must have been the one from Standard Chemical Company which they had called 1.04 mg. Standard No. 6 is 22 mm in length. The length of the smaller (1.052 mg) NBS standard is not given on the SCC certificate.

In his letter of December 15, 1914, Boltwood indicates that his "standard" is approximately 1.3 mg of radium. Boltwood must have been pleased with the Bureau's assigned value of 1.305 mg. Dorsey did not include an error budget with this informal calibration of the Yale standard. The length of the tube may not have been the major source of uncertainty as the Yale standard described in Boltwood's December letter was "an impure salt". The source self-absorption would depend on the mass and chemical form (sulfate, bromide or chloride).

Finally, Dorsey's letter indicates that **graphs** are enclosed. But only the one shown here is in the archives. Lastly, the length correction for a line source should diminish with distance from the detector. Maybe it wasn't practical to back off 20 cm or so from the electroscope.

Two-page typed letter of April 12, 1915 from Dorsey

Department of Commerce
Bureau of Standards
Washington

NED

PFD

Address reply to

Bureau of Standards April 12, 1915.

Prof. B. B. Boltwood,
Yale University,
New Haven, Conn.

My dear professor Boltwood:

We have just received from the American Radium Pharmaceutical Co. (formerly American Radio Thor X Co.) of Chicago a request for the B.S. to test their products. This circular appears to have all the ear marks of a fraud, but it is considered desirable to let them send the stuff.

For most of their products they claim the presence of radium and mesothorium. Can you suggest a workable method for quantitatively determining small quantities of mesothorium? I expect that the actual amount of radium to be dealt with will be of the order of 10^{-9}gm, and less mesothorium. Would an alpha ray measurement using thin films (McCoy), combined with an emanation determination of the radium be of any value?

Would you please give me the names of firms in this country from whom mesothorium can be obtained? What

-2-

is the present price? I have been wishing to get some to play with but the money has not been available. It is barely possible money enough may be available in the next few months, and I wish to be ready to take advantage of it when the time comes.

Would it be possible to arrange for an informal comparison of our standard radium solution with yours? It appears to me that such a comparison would be very interesting.

Kindest regards to Bumstead and Uhler. I was very glad to meet Johnson the other day, please give him my regards.

Very sincerely,

N. Ernest Dorsey

Dorsey's letter in the Spring of 1915 is interesting for a number of reasons. WWI has been underway for nearly a year, but it is not clear if that is the cause of the shortage of funds. Dorsey mentions that he would like to compare the NBS radium solution standards with those prepared at Yale by Boltwood. He may be looking for some validation that his serial dilutions have been done correctly.

He also intends to calibrate radium sources at the nanogram level. He asks about using the emanation technique. That suggests that he has an electroscope that can be set up for comparative measurements of radon from different sources. And he is reading the current literature on alpha-particle measurements with thin films. He refers to McCoy's measurements of thin films (e.g., McCoy and Ashman, 1908).

Typed letter of April 13, 1915 from Boltwood

April 13, 1915

Dr. N. Ernest Dorsey
Bureau of Standards
Washington, D.C.

My dear Dr. Dorsey:

I have just received your letter of April 12th in regard to the testing of certain materials for mesothorium and radium.

I expect to be in Washington for the Meeting of the National Academy of Sciences, April 18-22 and think I can accomplish more by talking the matter over with you than by attempting to discuss it in a written communication. The program of the Academy Meeting is a rather full one and I do not expect to have much free time but if you could come around to the National Museum on Monday, Tuesday or Wednesday afternoon or Tuesday morning, you would be pretty sure to find me there and I could talk the matter over with you.

The only firm that I know of that sells mesothorium is Dr. O. Knoefler & Co., Plotzensee b. Berlin, and the last price quotation I saw was at the rate of fifty dollars per milligram radium bromide equivalent (measured by the gamma rays).

I will try to arrange to bring down some of my radium standard solution so that you can compare it with the solution that you have. I think it will be very interesting to find the relative strengths of these two solutions.

Hoping to see you in Washington, and with kind regards, I am

Yours sincerely,

It seems the US Postal Service was incredibly efficient. A letter mailed in Washington on April 12, 1915, is replied to the following day from New Haven. This is the first instance where Boltwood addresses him as "Doctor Dorsey". The National Museum refers to the Smithsonian Institution. (Rutherford had been there to give a lecture the previous April.) Interesting that Boltwood refers Dorsey to a mesothorium supplier in Berlin. There is a war going on and Rutherford's student James Chadwick had taken a fellowship with Hans Geiger at the PTR in Berlin (Brown, 1997). Chadwick was interned as a prisoner of war in Berlin for the duration of the war. Boltwood had a certain affinity for the Germans, and this caused some tension between

him and Rutherford during the war (Badash, 1969). There is no further information about the plan to compare the solutions from NBS and Yale.

Typed letter of May 20, 1915 from Dorsey

Department of Commerce
Bureau of Standards
Washington

NED

PFD

Address reply to
Bureau of Standards May 20, 1915.

Prof. B. B. Boltwood,
Yale University,
New Haven, Conn.

Dear Prof. Boltwood:

Can you tell me anything regarding the ratio of the old Curie and Vienna standards to the International Radium Standard? These ratios must have been known when the International Standard was adopted but I have found nothing bearing on the subject. So far as I have been able to ascertain the only old standard for which the value on the present basis has been published is Rutherford's. He once stated that the old standards differed among themselves by about 20%, but their relative values, so far as I know, have never been given.

The Chicago therapeutical preparations have not yet been sent to us, so the thorium question is still in abeyance. There being a lot of other work on hand just now I've not written Mr. Miner. Shall, however, do it during the summer.

Very truly,
N. Ernest Dorsey

Enc: Addressed envelope.

It is interesting that Dorsey is asking about the ratio of the old Curie and Vienna standards. Curie had reported that the 21.99 mg Paris standard and the 31.17 mg Vienna standard agreed to within 0.2 % (Curie, 1912). Meyer and von Schweidler (1916) showed good agreement between the values from the Curie Institute and Institute for Radium Research. Dorsey and Boltwood may not have had all this information in May of 1915. Boltwood replied to Dorsey on May 22, 1915, but asked that his reply be kept confidential. That letter is not included in the archives. He probably made some more unkind remarks about Marie Curie's mass measurements and did not want those on the record. One cannot believe that the original standards differed by 20 %. Dorsey may have been referring to verbal remarks he had heard from Rutherford, rather than his published papers. There is no written record that Dorsey met Rutherford, but it is most likely that he talked to him during Rutherford's visits to the Bureau and to APS meetings in Washington in 1914 and 1917.

3.9. First Electroscopes at NBS

Dorsey mentioned to Boltwood in 1914 the electroscopes that he was using but did not publish any details until he provided this description in his 1921 textbook for medical physicists (Dorsey, 1921a).

> "The simple electroscope consists of a metal case within which, and near its center, is supported in a vertical position a well-insulated metal strip to the top of which is attached a narrow strip of thin foil, preferably of gold leaf. This strip of foil is usually spoken of as the leaf. The strip of metal and the leaf constitute the insulated system of the electroscope. When the insulated system is electrically charged by a suitable switch passing through the wall of the case, the leaf is repelled by the strip, and is deflected from its normal, vertical position. In opposite sides of the case are windows through which the position of the leaf can be observed. Such observation is usually made by means of a microscope having in its eyepiece a ruled scale.
>
> When intended for gamma ray measurements, the electroscope should be carefully screened on all sides except one with lead at least one inch thick, so that the air in the electroscope will be protected from scattered radiations that would otherwise enter it. For the same reason, the windows should be as small as is conveniently possible. The ionizing radiation of which the intensity is to be measured enter the electroscope through the unscreened side. In order to minimize the effect of the absorption of the radiation by the wall of the container and by the salt itself, the measurements should be based upon the hard, penetrating radiation emitted by radium-C. For this reason it is desirable that the radiations entering the electroscope be filtered through lead at least 15 mm thick so that very little of the soft gamma radiation from radium-B enters the electroscope.
>
> When everything is ready, all radium preparations are placed at such distances and so screened that they produce as small an ionization in the electroscope as possible – the preparations to be compared must be so placed that they produce only a negligible ionization. The insulated system is then charged and insulated, and the time required for the image of the leaf to move over a few divisions near the middle of the scale in the microscope is determined. From this, the rate of drift of the leaf, in divisions per second, when there is no radium near the electroscope, is determined. This is called the natural, or the blank, drift. It results from imperfect insulation and slight residual ionization of the air.
>
> Then the tube under test is placed in a suitable position, and the time required for the leaf to drift over a certain portion of the scale is determined. If the blank drift is subtracted from the rate of drift observed when the tube under test is in position, the difference will be the rate of drift due to the radiation from the tube; this is known as the corrected drift.
>
> The tube under test is now removed to its former position where it does not affect the electroscope, and the standard tube is placed in exactly the same position previously occupied by the tube under test. Its corrected drift is determined in exactly the same way as was that for the tube under test.
>
> The ratio of the two corrected drifts is equal to the ratio of the intensities of the two radiations; which, in turn, is equal to the ratio of the amounts of radium-C that are contained in the two tubes, provided that the absorption of the radiations by the walls of

> the two tubes is the same in both cases. Knowing the amount of radium-C in the standard, the amount in the tube under test can now be computed at once."

Although there are no records in the archives of the electroscopes that Dorsey purchased or constructed, there is a photograph from a newspaper in 1924 that shows Mary Brower Harrington (at NBS from 1921-1929; 1943-1954) using a gold-leaf electroscope to measure gamma rays from a radium source. The next version of the radium calibration range included a projection electroscope to provide more separation between the observer and the gamma-ray source.

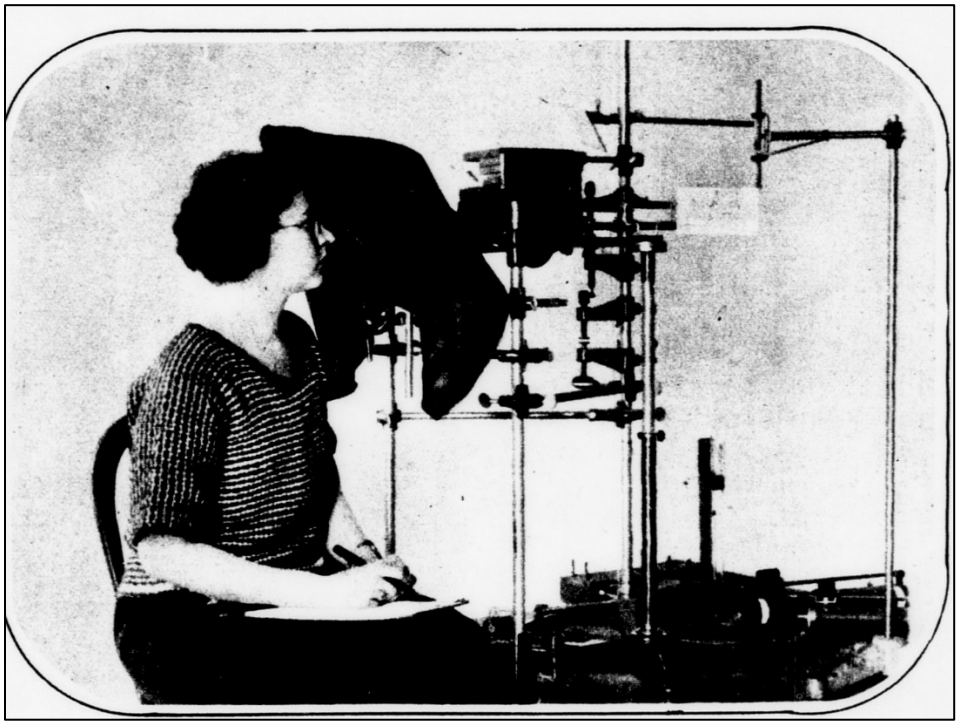


Figure 7. "Miss Mary Brower of the bureau of standards, an expert who measured four million dollars worth of radium recently. The photograph shows he method of measurement". Washington Evening Star, Jun 8, 1924.

There is also nothing in the NBS/NIST archives or publications that describes the apparatus used for the emanation (radon) measurements, although such measurements were being made in 1914 as noted in the Director's Annual report of June 30, 1914 (Director's Annual report, 1914). In 2020, during the clean-out of old storage areas in the Radiation Physics Building at the NIST site in Gaithersburg, Maryland, the author noticed an interesting piece of equipment that turned out to be a Lind emanation electroscope (See Figure 8.) (Lind, 1915).

Figure 8. Lind electroscope at NIST (credit NIST)

This instrument was referred to 9 years later by Elizabeth Hughes who reported that she had used it under the direction of Dorsey when she was in the NBS Radium Section in 1919 (Hughes, 1928).[10] In addition to Lind's original paper, there are good descriptions of the different versions of Lind's electroscope collected by Paul Frame in exhibits of the Oak Ridge Museum of Radiation and Radioactivity (ORAU, 2023).
https://www.orau.org/health-physics-museum/collection/electroscopes/radioactivity/lind.html

3.10. Radium at the UK National Physical Laboratory (NPL)

In the United Kingdom the National Physical Laboratory was founded in 1900 a year before the NBS in 1901. The British program in radiation metrology began at about the same time as Dorsey's work at NBS. Hönigschmid prepared the UK standard – International Secondary Standard No. 3 – in October of 1912. It was received at the NPL from Vienna in December 1912 and then sent to Paris for Marie Curie to measure relative to the Paris Standard. Curie returned it to NPL in June 1913 (Ariouet, 2021) with a certified value of 21.13 mg radium chloride (based on an average of the Vienna measurement of 21.10 mg and the Paris measurement of 21.16 mg.), equivalent to 16.04 mg radium element with an uncertainty of 0.3% (Smith, 1975).

[10] "The reverence attached to objects of history is a thing men feel. One could even say what endows any thing with significance is solely the history in which it has participated," Cormac McCarthy, The Crossing.

Dr. G.W.C. Kaye, who had worked with x rays with J.J. Thompson at the Cavendish Laboratory assembled the instruments and began testing in December 1913. Preliminary work at NBS started in late September 1913 with the two standards from Pittsburgh.

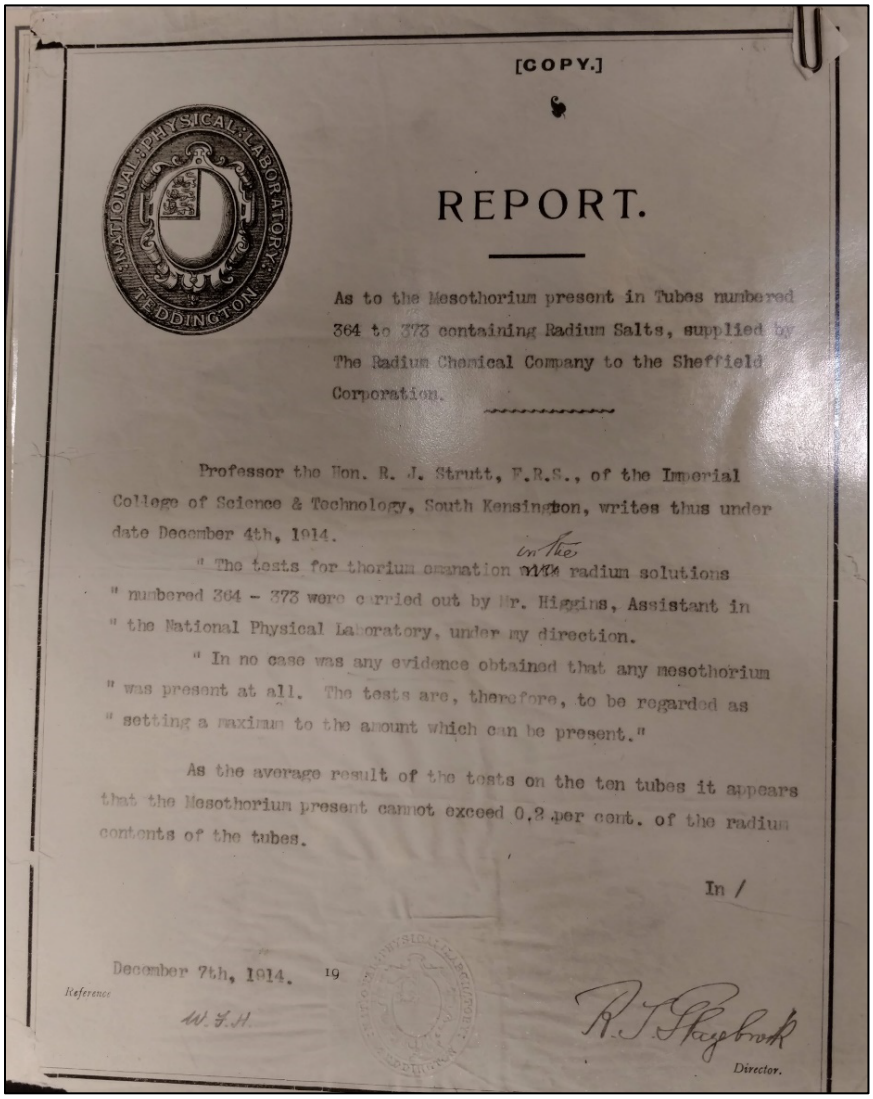
According to Smith's short summary of the NPL radium work (Smith, 1975):

> "It was reported that three different types of apparatus had been installed, each based on ionization measurement and incorporating at least 5 mm lead filtration."
>
> "These three types of apparatus were not novel, even at that date. The first, which was referred to as Rutherford's direct method, consisted of a combined gold-leaf electroscope and ionization chamber, the radium standard and test specimen being placed in turn in a carrier mounted on an optical bench, the radium contents being compared by timing the fall of the gold leaf. The second was the balance method of Rutherford and Chadwick, in which the distance of the standard or specimen was adjusted until the ionization current in one chamber balanced that produced by a film of uranium oxide in another. The relative radium contents of the standards and test specimen were determined by applying the inverse square law. The third method was that of Mme Curie, which consisted of a sandwich of a 60 cm diameter aluminium plate and two thick lead plates of similar size separated by air gaps of about 2 mm. The two lead plates were connected to a high potential and the aluminium plate to an electrometer. The standards and specimen were placed on the centre of the upper plate and compared by means of the ionization produced."

Smith reported that the first NPL test report was a Certificate of Examination for a radium bromide source dated December 23, 1913 (5.2 mg of Ra in equilibrium).

The NPL radium work proceeded very slowly during WWI and didn't pick up until Kaye returned to NPL following the war. The NPL report by Smith mentioned two other activities related to radioactivity during the war. They received requests to certify radium in luminous paints. They purchased a radium bromide source and used this to certify the activity in luminous sources to about 5 %.

The next intersection of the NPL radioactivity work with that of the US came later in 1914. NPL reported on measurements in September of 1914 of "mesothorium" (radium-228) in a commercial shipment of radium bromide. This was the radium-226 material that Glenn Kammer had brought to the UK from the Standard Chemical Company in Pittsburgh (Lubenau and Landa, 2017). Kammer left Pittsburgh for the UK with 4 grams of radium in a lead-lined valise intended for several hospitals in the country (Silverman, 1950). At Manchester he met with Ernest Marsden (Rutherford was away in Australia at the time.) and they found good agreement (better than 0.1%) between their assays. Part of the shipment was then accepted by the Royal Infirmary in Manchester. But other hospitals, Sheffield, Greenwich, Northampton, Swansea, could not accept the company's assayed values for the radium without a certification from the NPL that the materials were free of mesothorium (Silverman, 1950). NPL prepared solution samples from the submitted materials and measured for mesothorium by an emanation method under the "supervision" of Professor R.J. Strutt at the Imperial College of Science and Technology to demonstrate the absence of radium-228 (Smith, 1975). A copy of Strutt's report from December 7, 1914, was found in the NIST archives. It is not known whether this copy came from the NPL or from the Radium Chemical Company (probably the latter).



[COPY.]

NATIONAL PHYSICAL LABORATORY. TEDDINGTON.

REPORT.

As to the Mesothorium present in Tubes numbered 364 to 373 containing Radium Salts, supplied by The Radium Chemical Company to the Sheffield Corporation.

Professor the Hon. R. J. Strutt, F.R.S., of the Imperial College of Science & Technology, South Kensington, writes thus under date December 4th, 1914.

" The tests for thorium emanation in the radium solutions " numbered 364 - 373 were carried out by Mr. Higgins, Assistant in " the National Physical Laboratory, under my direction.

" In no case was any evidence obtained that any mesothorium " was present at all. The tests are, therefore, to be regarded as " setting a maximum to the amount which can be present."

As the average result of the tests on the ten tubes it appears that the Mesothorium present cannot exceed 0.2 per cent. of the radium contents of the tubes.

In /

December 7th, 1914. 19

Reference

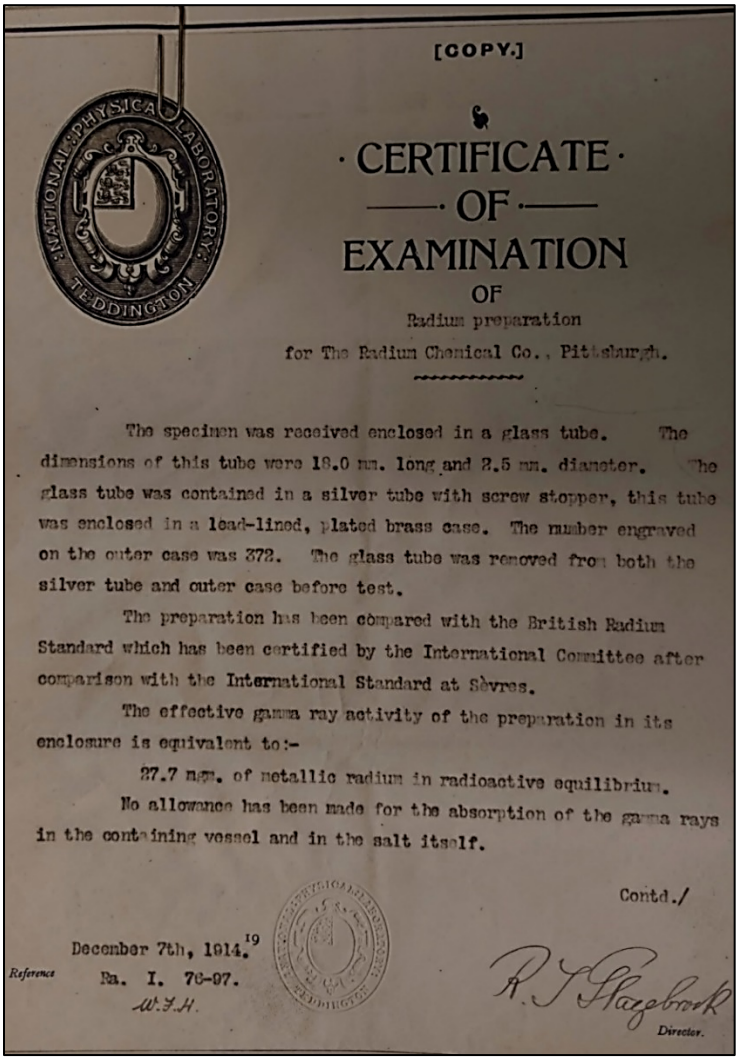
W.J.H.

R. T. Glazebrook

Director.

Figure 9. Report from the National Physical Laboratory of December 7, 1914, describing measurements at the Imperial College of Science and Technology indicating the absence of radium-228 in samples of radium-226 from the Radium Chemical Company.

On recommendations from NPL, the hospitals accepted the SCC radium.[11] On the same date in December of 1914 the NPL signed a Certificate of Examination of a sample from the Radium Chemical Company.



[COPY.]

NATIONAL PHYSICAL LABORATORY TEDDINGTON

· CERTIFICATE ·
—· OF ·—
EXAMINATION
OF
Radium preparation
for The Radium Chemical Co., Pittsburgh.

The specimen was received enclosed in a glass tube. The dimensions of this tube were 18.0 mm. long and 2.5 mm. diameter. The glass tube was contained in a silver tube with screw stopper, this tube was enclosed in a lead-lined, plated brass case. The number engraved on the outer case was 372. The glass tube was removed from both the silver tube and outer case before test.

The preparation has been compared with the British Radium Standard which has been certified by the International Committee after comparison with the International Standard at Sèvres.

The effective gamma ray activity of the preparation in its enclosure is equivalent to:-

27.7 mgm. of metallic radium in radioactive equilibrium.

No allowance has been made for the absorption of the gamma rays in the containing vessel and in the salt itself.

Contd./

December 7th, 1914.[19]

Reference Ra. I. 76-97.
W.J.H.

R. T. Glazebrook
Director.

Figure 10. Copy of a National Physical Laboratory Certificate of Examination of a source submitted by the Radium Chemical Company (NIST archives (2023).

During 1914 – 1915 NPL also reported exchanging sources with the SCC and generally found agreement to within 0.5%. It is not clear whether future SCC shipments to UK hospitals still required certifications at the NPL. The war soon disrupted the program at NPL, as Kaye was called to military service, and there was no mention of activities of the Radium Section in 1915 – 1916 (Smith, 1975).

[11] The carnotite ores from the Western US contained very little mesothorium (less than 0.2%), but NBS was unable to make that assessment in 1914. Six years later Director Stratton sent a letter to the Editor of the JAMA advising customers on the experience at NBS and how to assure themselves that radium purchases were free of mesothorium (Stratton, 1920).

3.11. Radiation Metrology at NBS and the NPL (1912 – 1915)

It is interesting to compare the radiation metrology at NBS and NPL in this period from 1912 – 1915. Both programs began with little understanding of radioactivity but with strong expertise in fundamental electrical measurements. They received specimens of the international radium standards at nearly the same time and began to develop similar infrastructure for both electroscopic gamma-ray measurements and emanation techniques. With the outbreak of WWI, both institutes had to focus activities on work to support the war effort, but this happened more quickly in the UK. The NBS programs were driven by requirements of the radium industry and the US Bureau of Mines, which led Dorsey to concentrate first on the radium work. He noted that the x-ray work had to be put aside (NBS Director's Reports). The NPL placed a higher priority on the x-ray work to support the widespread use of x-ray diagnostics for imaging wounded soldiers in the European theater. Responding to the needs of the defense departments, both NBS and NPL developed capabilities to calibrate self-luminous paints used for military applications (to be described later). But the priorities of the programs at NBS and NPL seem clear from the best-selling textbooks from Dorsey (NBS) and Kaye (NPL) published after the war: *Physics of Radioactivity* (Dorsey, 1921a) and *The Practical Application of X-Rays* (Kaye, 1922).

3.12. Rosa report to Journal of the Franklin Institute

Edward Bennett Rosa at NBS had many responsibilities as the Chief of the Electricity Division and Chief Physicist. He left the radium work to Dorsey, but he did maintain supervision of the Radium Section and included their work in his annual submission to the Journal of the Franklin Institute. His report in 1915 is the only early NBS publication that gives an overview of the radioactivity programs (Rosa, 1915).

> "One of the most recent lines of work taken up by the Bureau is the investigation and measurement of radium and radio-active substances, including radium emanation, radium minerals, and radio-active waters. The Bureau has a standard specimen of radium which is standardized in terms of the international standard preserved at Paris, and makes a large number of measurements of sealed radium specimens to determine by means of their gamma radiation the quantity of radium contained. Various radium products and preparations for therapeutic use are also submitted to the Bureau for test. It is important that physicians using radium shall know definitely the quantity contained in their specimens in order to be able to correlate the results obtained with the strength of radiation employed. Also, when radium sells at the rate of three and one-half million dollars an ounce, it is important that the purchaser of a specimen should know how much radium is actually contained in the brown powder, some of which may be sand which without measurement may be passed as radium. It is also important to subject so-called therapeutic preparations of radium and radium salts to a test of actual measurements in order to discourage fakes and frauds. The Bureau has made a very careful study of methods of measurement, and has a considerable instrumental equipment for such measurements."

4. Standards: 1916 -1920

4.1. Introduction

During the early years of WWI, the radium calibrations of the US and the production capabilities of the SCC and NRI were greatly expanded. During this period the SCC in Pittsburgh produced the bulk of radium for the world market. All this material for medical uses and for the Army and Navy was calibrated at NBS before being shipped to end users.

4.2. Annual Report of the Radium Section 13 of the Electricity Division

The Director's Annual reports during these years show the constant pressure on the Radium Section for what were essentially routine calibrations of submitted radium. There are very few records in the NIST archives on the research carried out during this period. But there is one extensive report in Dorsey's file at NIST on the activities of his Section 13 of the NBS Division I for the fiscal year ending June 30, 1920. At this time the radium section was identified as the Radioactivity and X-Ray Measurement Section. The photocopy has been transcribed and is included here in its entirety (NIST archives, 2023):

Bureau of Standards

Division I – Section 13

Fourteenth Report of the Progress of the Work of the

Radioactivity and X-Ray Measurement Section

July 1, 1919 to June 30,1920

The year covered by this report has been the most strenuous in the history of the section. The volume of gamma ray testing has increased so greatly and so rapidly that everything else has had to give way to it. X-ray and luminous material work have of necessity been discontinued except for a few tests that have come to us without encouragement; and those few have had to wait, often for months, until a lull occurred in the gamma ray work. During the year just closed over 25 ½ grams of radium and mesothorium were received for tests, while during the preceding 5 ½ years a little less than 32 ½ grams were received. All indications point to a further increase in the demand for this work and it is hoped that the Division may be able to make such provision that it can be carried without unnecessary embarrassment.

Work in luminous materials and x-rays should be renewed as soon as the finances of the Bureau justify the appointment of a sufficient force to carry out the work with credit. Naturally, research work along all lines should be renewed at the earliest opportunity.

Personnel: As in the past there have been many changes in the personnel of the Section. The only member who has been continuously connected with the Section since its creation is its chief, N.E. Dorsey, and he, feeling the need for rest and desiring to take up

consulting work, tendered his resignation in April, effective June 30. After that date Mr. W.H. Wadleigh will look after the work of the Section. Dr. N.E. Dorsey has been appointed in the capacity of consulting physicist on a per diem basis, effective July 1.

The history of the personnel in the Section during the year is shown in the following table:

Name	Position	Appointed	Resigned
Dr. N.E. Dorsey	Physicist, Chief of Section	Mar. 1913	June 30, 1920
Miss N.N. Alderton	Lab. Assistant	Apr. 1918	Aug. 2, 1919
Mr. W.D. McCrea	Aid	Sept. 1917	July 31, 1919
Mr. S.F. Fegah	Aid	June 21, 1919	Apr. 8, 1920
Miss A.D. Berg	Lab. Assistant	Aug. 11, 1919	Dec. 31, 1919
Miss E.E. Damon	Lab. Assistant	Dec. 1, 1919	----------
Mr. L.R. Kleinschmidt	Lab. Assistant	Feb. 2, 1920	June 4, 1920
Mr. F.F. Chesnut	Aid	Mar. 1, 1920	July 31, 1920 (?)
Miss W. Kenyon	Lab. Assistant	April 7, 1920	July 31, 1920
Mr. W.H. Wadleigh	Assoc. Physicist	June 7, 1920	----------
Mr. W.W. Fuller	Aid	June 18,1920	Appointed for 3 mo.
Mr. C.H. Moseley	Lab. Assistant	June 21, 1920	Temporary appointment

The frequent changes in personnel are both undesirable in themselves and inimical to the efficient growth and functioning of the Section. They are, however, not surprising when all the conditions are considered. In the radium work it is desirable to employ Aids of more than usual ability. Such an aid regards his position at the Bureau as merely temporary, to be held while he is studying, acquiring experience, or looking for a position elsewhere. The same is true of most of the Laboratory Assistants of ability. Appointments of higher grade, excepting the Chief of Section, have been assigned to the Section only on temporary appointments, and both they and prospective appointees have in many cases been carefully advised by the Division that the appointment is not to be considered permanent or as extending beyond a specified date a few months in the future. Until conditions become such that the Section can be given several members that may be regarded fairly permanent a heavy turnover and relatively inefficient work is to be expected.

A greater stability will also be assured by assigning to the Section a sufficient force to enable it to do more that carry out routine testing. With a force sufficient to allow the constructive members of the Section to study and map out research problems relating to the work and to allow the observers opportunities to take part in real investigational work, the monotony of the routine would be broken, the interest and enthusiasm of the younger members would be maintained, and a longer tenure of office would undoubtably result.

Tests: During the year 1240 gamma ray measurements and 178 other tests have been performed; 23259 mg of radium and 2019 mg of mesothorium were submitted for test. This material has a market value of over $2,600,000.00.

Seventy-nine preparations containing a total of 1390 mg of radium were certified for export; it was distributed as shown in Table 1:

Table I: Radium Certified for Export During Fiscal Year ending June 30, 1920

Country	No. of Preparations 1919	No. of Preparations 1920	Total No. for Year	Radium (mg) 1919	Radium (mg) 1920	Total for Year
Japan	11	11	22	211.03	306.26	517.29
Philippines	11	_	11	344.60		344.60
Cuba	6	_	6	92.30		92.30
Brasil	2	7	9	7.63	85.35	92.98
Canada	1	5	6	24.47	48.02	72.49
Columbia	2	_	2	49.75		49.75
Italy	5	2	7	45.63	3.98	49.61
Australia	_	4	4		32.08	32.08
India	1	1	2	5.05	24.85	29.90
Spain	1	1	2	26.38	2.59	28.97
Panama	_	1	1		24.99	24.99
Porto Rico	1	1	2	10.19	12.41	22.60
South Africa	1	_	1	16.05		16.05
Peru	1	_	1	9.89		9.89
Holland	_	3	3		6.18	6.18
Totals	43	36	79	842.97	546.71	1389.68

Growth: The increase in the work handled by the Section during the 6 ½ years during which it has been doing routine testing is shown in Table II.

Table II: Showing Growth of Routine Testing Performed by the Radium and X-Ray Section

Interval	Gamma-ray	Others	Total tests	Radium (mg)	Fee
Jan. to June 1914	28		28	486	$210.64
July to Dec. 1914	37	8	45	437	309.84
Jan. to June 1915	45	8	53	1660	628.16
July to Dec. 1915	45	25	70	2164	758.49
Jan. to June 1916	71	36	107	2367	948.39

July to Dec. 1916	98	2	100	2760	875.50
Jan. to June 1917	125	67	192	3878	1175.98
July to Dec. 1917	108	281	389	2957	1159.35
Jan. to June 1918	69	790	859	2419	1413.73
July to Dec. 1918	139	447	586	4625	1627.11
Jan. to June 1919	341	141	482	8742	2373.12
July to Dec. 1919	457	97	554	10298	2820.52
Jan. to June 1920	783	76	859	14980	4967.00*
Total	2346	1978	4325	57773	$19267.83

*These data are only approximate, full data for June not available.

The growth of the gamma ray work is shown graphically in Fig. 1 (not available). This curve of monthly receipts is plotted from the values "smoothed" over 7 month intervals.

Cooperation: During the first half of the year the Section cooperated with the Ordnance Department of the Army in the study of luminous targets and of radium lamps for use in reading scales. It has also measured the brightness of a number of luminous gun sights both before and after field use. For other Sections of the Bureau radiographs of metal and of wood have been made.

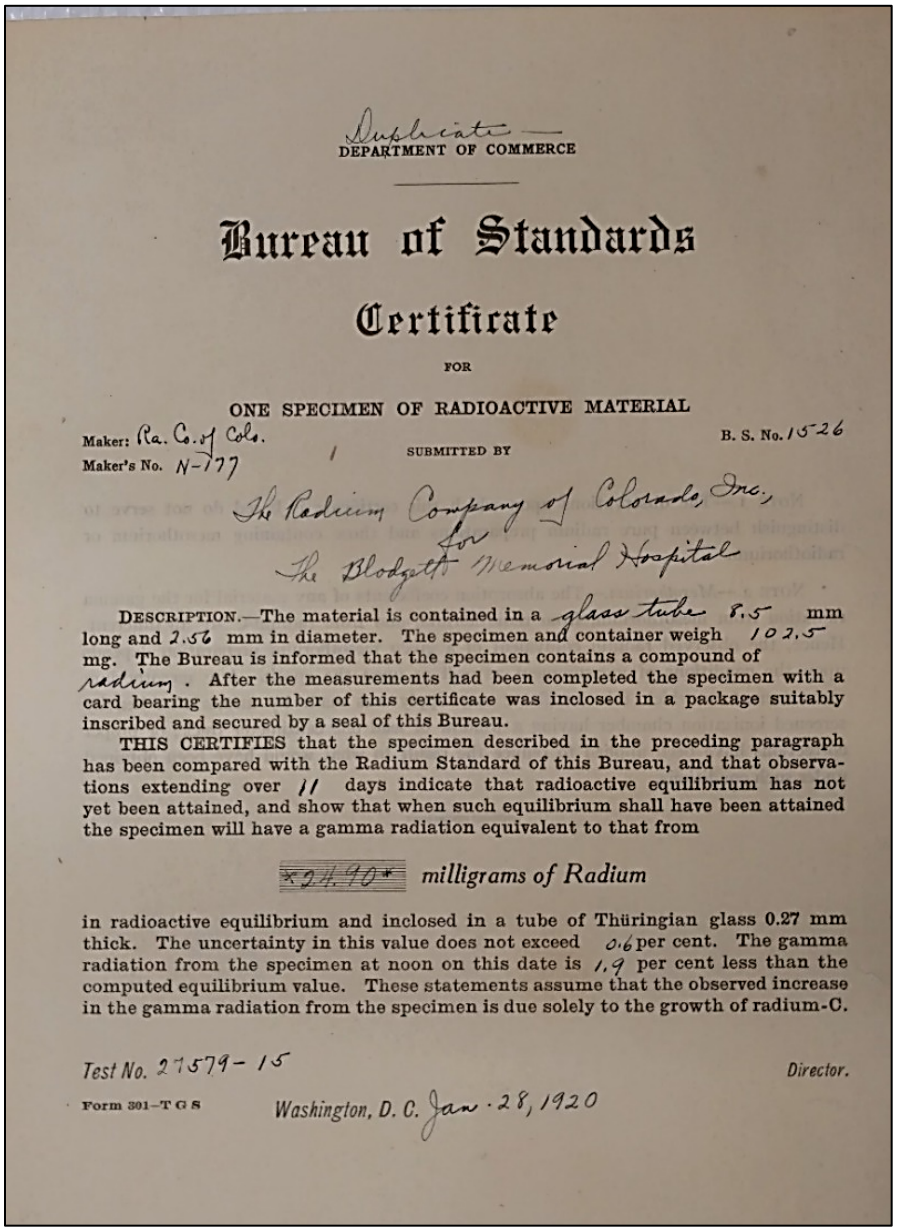
General: The Bureau has requested the Department of Justice to investigate Dr. A.E. Kay who appeared to be offering for sale as radium a product that is not radioactive.

What is particularly striking about this report is the staffing for the section in that they were essentially all part-time or short-term workers. Dorsey had two decades of experience, but the only other senior physicist listed was Hiram Wadleigh, who had recently transferred into the radium work. Wadleigh joined because Dorsey had announced his retirement the previous April, and Rosa needed someone to take the position of Section Chief. Another important member of the section was Elizabeth Damon who joined NBS in 1919 with a bachelor's degree from Vermont University (Coursey, 2021). She would later report that NBS was measuring all the radium supplied to doctors in the US. She is shown here in a photograph from 1920 in which she is providing what is said to be a gram of radium for use in New York State. Likely this material was intended for the Roswell Park Cancer Center in Buffalo, New York (Landa, 2023). The total value of materials calibrated during that fiscal year was an astounding $40B in 2023 USD.

Figure 11. “Elizabeth Damon of the bureau of standards handing a representative of the Radium Chemical Company a tube containing one gram of radium for New York State, valued at $120,000. This is the first shipment tested by the bureau. It will be used for social service purposes in the New York State Institute for Cancer Research.”

Damon (later Elizabeth Hughes) reported that she calibrated submitted radium sources with the gold-leaf electroscope under the direction of Dorsey (Yale archives, 2023). The only report of calibration from this period in the NIST archives is shown here. It is likely one of the last sources calibrated by Damon and Dorsey as they both left the Section in the Summer of 1920.

Certificates such as this were signed by the NBS Director:



Duplicate

DEPARTMENT OF COMMERCE

Bureau of Standards

Certificate

FOR

ONE SPECIMEN OF RADIOACTIVE MATERIAL

Maker: Ra. Co. of Colo. B. S. No. 1526

Maker's No. N-177

SUBMITTED BY

The Radium Company of Colorado, Inc.,
for
The Blodgett Memorial Hospital

DESCRIPTION.—The material is contained in a glass tube 8.5 mm long and 2.56 mm in diameter. The specimen and container weigh 102.5 mg. The Bureau is informed that the specimen contains a compound of radium. After the measurements had been completed the specimen with a card bearing the number of this certificate was inclosed in a package suitably inscribed and secured by a seal of this Bureau.

THIS CERTIFIES that the specimen described in the preceding paragraph has been compared with the Radium Standard of this Bureau, and that observations extending over 11 days indicate that radioactive equilibrium has not yet been attained, and show that when such equilibrium shall have been attained the specimen will have a gamma radiation equivalent to that from

~~24.70~~ *milligrams of Radium*

in radioactive equilibrium and inclosed in a tube of Thüringian glass 0.27 mm thick. The uncertainty in this value does not exceed 0.6 per cent. The gamma radiation from the specimen at noon on this date is 1.9 per cent less than the computed equilibrium value. These statements assume that the observed increase in the gamma radiation from the specimen is due solely to the growth of radium-C.

Test No. 27579-15 Director.

Form 301—T G S Washington, D. C. Jan. 28, 1920

Figure 12. Certificate from NBS of a radium sample submitted by the Radium Company of Colorado[12] intended for use at the Blodgett Memorial Hospital in Detroit, MI (NIST archives).

The format of the certificates changed very little over the next century although the statements of the uncertainty are continually refined.

[12] The Radium Company of Colorado was founded by Willy Schlesinger and Henry Koenig (who had left the SCC) from a collaboration they formed in Princeton. From 1914 they operated mining and radium extraction facilities in Colorado and in 1918 changed the name to the Radium Company of Colorado. They opened a subsidiary plant in Denver, the Cold Light Manufacturing Company, to produce luminous dials for the military (Hart and Twitty, 2008; Landa, 1982).

4.3. Mesothorium Calibrations at NBS

Dorsey's annual report of 1920 reported that NBS provided calibrations of mesothorium (radium-228) materials said to contain over 2 grams of mesothorium. The NIST archives examined to date do not include any reports of calibration of samples known to be radium-228. But a good description of the NBS mesothorium calibrations comes from Dorsey's textbook on radioactivity printed in 1921.

> "Mesothorium preparations are sold on the basis of their gamma-ray activity as compared with the activity of a known amount of radium. As filtering must be used in order to remove the beta rays, and as the penetration of the rays from mesothorium is different from that of those of radium, it is evident that the relative activity observed is dependent upon the filtration employed. It is customary to use a standard filtration of 1 cm of lead, and to designate as 1 mg of mesothorium that amount of mesothorium 30 days old that will produce, with this filtration, the same ionization of air as is produced by 1 mg of radium under like conditions. The specification that the preparation is to be 30 days old is to eliminate errors that might arise from the variation in the activity of the preparation during this period. Frequently in practice the age of the preparation is not specified; the measurement then indicates merely the apparent number of equivalent milligrams of mesothorium. The actual monetary value of one apparent milligram of mesothorium decreases as the age of the preparation at the time of measurement increases, until an age of seven or eight years is reached, after that it remains constant, is independent of the age at which measured. It is very evident that a preparation that contains one apparent milligram and is at its maximum, is worth much less than one that contains one equivalent milligram and is a month old (Dorsey, 1921a)".

With this description of "1 mg of mesothorium," Dorsey is actually defining a "mg radium equivalent". An actual mass ratio for equivalent activities of radium-226 and radium-228 would be based on the ratio of half lives, that is, 278.3. A separated sample of radium-228 rather quickly becomes a mixture of radium-228 ($T_{1/2}$ = 5.75 years) and a decay product thorium-228 ($T_{1/2}$ = 1.91 years). To explain this Dorsey provides the following figure.

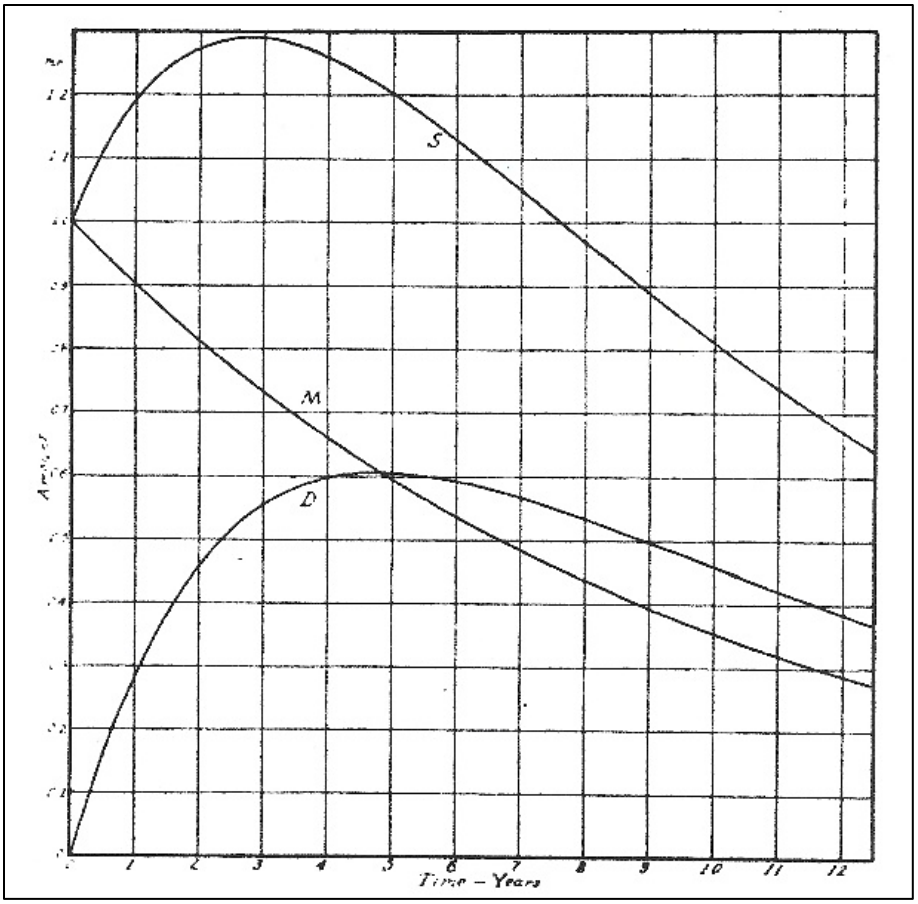


Figure 13. “Curves M, D and S show, respectively, the decay of mesothorium, the accumulation and decrease of Thorium-D, and the sum of the amounts of mesothorium and Thorium-D, each expressed in millirutherfords.”

A key to this graph is:

X- axis, time in years

Y- axis, activity in millirutherfords (1 rutherford equals = 10^6 becquerel)

M is the decay curve for mesothorium (radium-228); Dorsey uses $T_{1/2}$ of 6.7 years rather than the current value of 5.75 years.

D is the ingrowth curve for thorium-228; Dorsey assumes $T_{1/2}$ of 2 years rather than the current value of 1.91 years.

Radium-228 was being produced in large quantities from waste materials in the production of thorium at the Welsbach Gas Mantle Company in Berlin and in Gloucester, NJ. Professor Herman Schlundt from University of Missouri was working with the chemist Harlan S. Miner at the Gloucester plant to separate radium-228 from waste products (Gibbons, 2013). The radium-228 was shipped 90 miles north to the US Radium Corporation in Orange, NJ for use in fabrication of self-luminous materials (Coursey, 2021; Martinez et al., 2021). Marie Curie visited the Gloucester plant on May 24, 1921, during her US tour and received 50 mg of mesothorium (JIEC,1921; Lubenau, 2012). From the foregoing, we can assume that this was “50 mg radium

equivalent," as that mass of radium-228 would have had a gamma-ray exposure far exceeding the 1 gram of radium-226 that she took back to France. There are no records of whether this mesothorium sample was calibrated at NBS. Or how it was shielded for transport to Paris.

In addition to his consulting work with the Welsbach Company and the U.S. Radium Corporation, Schlundt had collaborated for several years with the USBM chemists Lind and Moore. In 1922 the USBM published a pamphlet on mesothorium with Schlundt as the sole author (Schlundt, 1922). Badash reported from private correspondence that Schlundt also shared samples of mesothorium with Dorsey at NBS (Badash, 1979). There are no such records in the NBS/NIST archives, but it seems likely that Schlundt provided Dorsey with the separated materials used to calibrate the electroscope at NBS. Schlundt credits the Germans, Otto Hahn and Lise Meitner, and Austrians, Stefan Meyer and Victor Hess, for their research on the "German Radium" and their methods of standardizing mixtures of radium-228 and thorium-228 against gamma-ray standards of radium-226 (Hahn,1908; Meyer and Hess, 1914; Meitner, 1918; Schlundt, 1922). The details of those papers are beyond the scope of this work. But suffice it to say that any assay value of mesothorium must include the date at which the radium-228 was separated from thorium isotopes and the filtration used for the gamma-ray comparison. The mesothorium was employed extensively in radiotherapy in Europe (Sgantzos et al. 2014, Robison, 2015), but in the US it was mainly limited to applications in luminous paints (to be described later).

Medical centers in the US were concerned about the cheaper mesothorium as a contaminant in their radium purchases for therapeutic applications. Dorsey had written to Boltwood in December 1913 asking how to make such determinations and Boltwood had suggested chemical separation procedures (Boltwood – Dorsey Letters, Yale, 2023). But NBS chose to assure themselves of the purity of submitted samples by measuring the gamma-ray emission rate as a function of time. In 1920 NBS Director Stratton sent a letter to the Editor of the Journal of the American Medical Association advising buyers about mesothorium in their radium purchases (Stratton, 1920). The article shown here was reprinted in the SCC journal **Radium**.

Measurement of Radium

To the Editor:

In the routine testing of hermetically sealed radium preparations, the ionization produced in a given ionization chamber by the penetrating gamma radiation proceeding from the preparation is compared with that produced under the same conditions by the similar radiation from a standard containing a known amount of radium. Mesothorium preparations also emit a penetrating gamma radiation, and consequently by a single comparison with the radium standard in the manner just indicated there is no means for distinguishing such a preparation from one containing only radium and its derivatives.

It is for this reason that the usual radium certificate issued by the National Bureau of Standards contains no statement concerning the actual amount of radium contained in the preparation, but merely a statement of its equivalent radium content. The primary object of the measurement of such preparations by the bureau is to insure the purchaser against any serious error in the radioactive measurement of the preparation.

The carnotites, from which the domestic radium is produced, are known to contain only a negligible amount of mesothorium. Tests made on radium produced from such ore gave no evidence of the presence of mesothorium, and were such as to indicate that the mesothorium present cannot exceed 0.2 per cent, of the radium content of the material. Consequently, it is quite safe to assume that the radium produced from these deposits will be practically free from mesothorium unless the latter product is deliberately added. This is a matter over which the producer has control and concerning which he can speak with confidence. It is customary for the domestic producers of radium to guarantee that their product is practically free from mesothorium, and such a guarantee might well be requested by the purchaser.

Although the examination of a hermetically sealed radium preparation for the presence of mesothorium forms no part of the routine measurement of such materials by the bureau, such examination will be made when requested under conditions that justify the work. These examinations are laborious, require the opening of the preparation and the removal of some of the salt, and involve the risk of a considerable loss of material. As in the case of all tests made by the bureau, the applicant must furnish the material used, assume the risk of loss, and pay a fee commensurate with the labor involved.

On the other hand, even without the examination the purchaser is not left entirely to the mercy of unscrupulous dealers. Repeated gamma ray comparisons, using radiations filtered through different thicknesses of lead, will in general furnish data from which it can be determined whether much of the radiation from the preparation is due to mesothorium. Such tests on sealed specimens are deliberately made from time to time, and similar but less complete data are incidentally obtained from many specimens in the routine course of the testing. In no case has such test revealed to us the presence of mesothorium in any preparation that has been submitted to this bureau as one free from mesothorium; but few imported preparations have been so tested.

Another check on the possible presence of a significant amount of mesothorium in a radium preparation is afforded by its measurement. If the preparation contains a significant amount of mesothorium, then a second measurement made several months after the first will reveal:

A growth in the intensity of its radiation if all radiothorium has been removed from the material shortly before the first measurement.

Little or no growth if the radiothorium was last removed two or three years before.

A decrease of the radiation if the radiothorium had not been removed for over three years.

It is evident that unless the two measurements are very specially related to the age of the contained mesothorium, they will reveal its presence.

In the course of its work, the bureau has to its knowledge remeasured forty-seven radium preparations after intervals varying from two weeks to four years. Preparations from three domestic producers are included in the list. Some of these preparations were resubmitted by their producers, others by their purchasers. For fifteen of the preparations, the interval between these measurements exceeded six months. In no case did the difference between the two measurements exceed the sum of the allowable uncertainties of the two measurements. Excepting a single case in which the initial determination was known to have an unusually low precision, a difference as great as 0.9 per cent, was found in only one instance. The

average difference was 0.34 per cent. The second measurement usually, but not always, exceeded the first. This probably results from the fact that in many cases the radium had not fully attained its equilibrium at the time of the first measurement. Even thirty days after the radium preparation is scaled, it is 0.45 per cent. short of its maximum gamma radiation.

Whence it is seen that as yet we have found no indication that any hermetically sealed preparation offered by a domestic producer as a radium preparation contains an appreciable amount of mesothorium. Very few such specimens offered by small dealers, jobbers or importers of radioactive material have been submit ted to the bureau. Consequently we are at present not prepared to express an opinion concerning the material obtained from such sources.

S. W. Stratton-. Ph. D., Washington. D. C.
Director, Bureau of Standards.

As mentioned previously, radium–228 was used extensively in luminous paints in the period 1916 – 1921 and was widely implicated in the radium poisoning of the radium dial painters (Coursey, 2021). A recent dosimetry retrospective by Professor Martinez at Clemson University and her colleagues compares the radiobiological effects of ingested radium-226 and radium-228 (Martinez et al., 2021). NBS did not develop radioactivity standards for radium-228 until the 1980s when low-activity solution standards were needed for environmental assays (Noyce, 1981; Noyce et al., 1983).

4.4. Luminescence Measurements

When small amounts of radium-226 or other alpha emitters are incorporated into mixtures of zinc sulfide phosphors, they can be used as self-luminous paints. Slaughter reports that William Hammer was the first to report using such paints in May of 1903 (Hammer, 1903; Slaughter, 2013). The applications of self-luminous paints in the new century were clear to the general public, military and industry; luminous dials, luminous numbers on watches, clocks and handles that could be seen in the dark, and many uses for the military such as cockpit dials for WWI military aircraft and deck markers for Naval ships (Fischer, 1998). About 10 companies were formed in the US to produce and market self-luminous products (Martinez et al., 2021). Three of the larger producers of self-luminous sources were the Radium Luminous Materials Corporation (later the US Radium Corporation) in Orange, NJ (Coursey, 2021), the Radium Dial Company in Peru, IL, which was a subsidiary of the Standard Chemical Company of Pittsburgh (Lubenau and Landa, 2019), and the Radium Company of Colorado, which marketed Marvelite through their subsidiary, The Cold Light Manufacturing Company in Denver (see Figure 14).

Figure 14. Two 1917 advertisements for luminous paints for military applications (Fischer, 1998).

By the time that World War I was under way in Europe – but before the US entered the war – manufacturers and the defense agencies were requesting that NBS develop methods of standardization of self-luminous products. This was first mentioned in the Director's Annual Report of June 30, 1916.

4.4.1. Director's Annual Report – June 30, 1916

> "In response to urgent requests, the Bureau is about to undertake the testing of luminous preparations containing radium. These preparations are not only used for the illumination of watch and clock dials, but are most valuable in the construction of many military and naval appliances designed for use at night. The Bureau is informed that one company in this country is making regular monthly shipments abroad of large amounts of this material."

The following year, the Director's Annual Report was able to provide a summary of the extensive work during that first year. The manufacturers and the military were cooperating with NBS by supplying materials and listing their specifications for different applications. Further examples of these products are given in the ORISE catalog (Buchholz and Cervera, 2008). The emphasis in the Director's report is clearly on the military importance of the luminous dials and markers and the sense of urgency placed on NBS by their defense counterparts. The US entered WWI on April 6, 1917; just prior to the preparation of this Director's report in June of 1917.

4.4.2. Director's Annual Report – June 30, 1917

> "Owing to the fact that the luminescence of radium luminous preparations persists for months or years and is not dependent upon a previous exposure to light, such preparations are of great value in the illumination of signs and dials that have to be read in the dark. They are largely used on instruments employed in aviation and for other military uses.
>
> The Bureau began the investigation of these preparations during the year, and now has under study 50 specimens obtained from various sources. A photometer for the measurement of the brightness of these preparations has been constructed and described, and apparatus for the measurement of the material after it has been applied and for the routine testing of prepared dials is being designed. The manufacturers and users of these materials are cooperating with the Bureau in this work. Conferences concerning the military use of these materials have been had with members of the Aviation Corps and of the French Scientific Commission. Preliminary reports of the work have been furnished on request to the National Advisory Council for Aeronautics and to the Signal Corps".

4.4.3. Rutherford Visit to Washington 1917

US combat involvement in WWI was from April 6, 1917, to November 11, 1918. The NBS staff doubled due to the war effort from 517 in 1917 to 1117 in 1918 (Behrman, 2022). In June of 1917, Rutherford led a UK-French scientific delegation to inform their new allies on their research on military-related activities; he wanted to avoid unnecessary duplication of efforts. The delegation visited military establishments in Canada and Eastern US cities and federal agencies in Washington, DC. NBS Director Stratton was their host for meetings in Washington. Rutherford visited NBS on June 4, 1917, and had lunch at the Cosmos Club with Stratton on June 5th (Eve, 1939; Coursey, 2018). Discussions at NBS probably included Rosa and Dorsey on matters related to radium and luminous materials calibrations. But the delegation was more concerned about SONAR technology for detection of German U-Boats (Eve, 1939). A year later the Director's Annual Report contained far more detail.

4.4.4. Director's Annual Report – June 30, 1918

> "Both the apparatus for and the technique of the measurement of the brightness of self-luminous materials have been much improved during the year. The fifty preparations under study in June, 1917, and a few others obtained since then have been measured at intervals. The conclusions drawn from these measurements have been made available to the military authorities interested in such materials.
>
> These materials are largely used for airplane instrument dials. Since little information regarding the brightness needed for satisfactory legibility of dials was available, the Bureau arranged to have a number of dials of various known brightnesses examined under service conditions of illumination by members of the air service of the United States and allied nations. A better idea of the brightness desired under different conditions was thus obtained. The variation of the legibility with the size and brightness of the characters has also been studied.

The conclusions thus reached have been used, and passed on to those interested, as rapidly as obtained and are now being collated in a series of recommended specifications for luminous articles.

Regarding the permanence of luminous material under service conditions little was known. It was believed that in general, an exposure to direct sunlight markedly decreased the brilliance of applied material. Observations made during the past winter have, however, shown no very marked decrease in brightness, due to an exposure either to the sun or to a mercury arc. The dials used in this test were of low brightness. Similar tests upon other dials will be begun at once.

Specifications for luminous markings upon airplane dials have been recommended to the Signal Corps, United States Army, the Bureau of Steam Engineering, United States Navy, and informally to other departments of the Government. These specifications have been in the main adopted by the Signal Corps.

Since September 1917, the brightness of a large number of dials has been measured for the Science and Research Division of the Signal Corps, United States Army), (recently transferred to the Bureau of Aircraft Production), as a basis for the awarding of contracts for the illumination of airplane instruments.

At the request of the Inspection Department of the Signal Corps, the Bureau has prepared and standardized equipment for factory inspection of dials, and has instructed inspectors in the details of factory testing. Since this equipment has been in use, brighter dials have been secured.

Since the first of January, 1918, a percentage of the luminous dials accepted by the Inspection Department has regularly been sent to the Bureau for careful measurement of brightness. Certain of the dials have been submitted to a life test, and a number of compass cards have been measured both dry and submerged in kerosene. The effect of continued immersion in kerosene also has been studied. The cause of some of the changes observed on immersion is still not clear; the study is being continued.

An investigation of methods for applying luminous materials to dials and other objects was undertaken late in November. Luminous materials were obtained from the different manufacturers and were applied to dials by means of various adhesives and methods. The various characteristics of the different adhesives and methods were noted and the brightness of the finished product was measured. The problem was studied from many angles and a method of application that is more efficient than any now in use was developed.

The following reports have been issued: " Self -Luminous Materials Containing Radioactive Excitants," "Notes and Suggestions Concerning Luminous Instruments," " Self -Luminous Materials — Brief Notes Covering a Few Points of Practical Importance." All of these were typewritten reports issued in editions of a few copies each and have been distributed to those Government departments that have appeared to be most interested.

Throughout this work the cooperation of the manufacturers and users of these materials has been most gratifying and of great assistance to the Bureau.”

4.4.5. 1918 -1919: NBS Program in Self-Luminous Materials, N.E. Dorsey

The NBS Director's Annual Reports from 1916 – 1921 provide increasing details on how NBS was responding to military and industry pleas for standards, instruments and quality assurance metrics for self-luminous materials, but they lack technical details about who was involved and how they carried out their studies. Characterization of self-luminous materials was an interdisciplinary project that required expertise in physical metrology (radioactivity and photometry) but also in materials science, chemistry, human vision perception, and quality assurance and quality control. With the increase in staff due to the war effort, NBS was able to assign multiple workers to these individual tasks. Dorsey in the Radioactivity and X-Ray Section 13 of the Electricity Division was the project leader. The photometer that was built for the project was probably coordinated with staff in the Colorimetry Section under the direction of Section chief Irwin Priest.

The Director's annual reports reference circulars that were prepared and distributed to the defense agencies, but these are not in NIST archives. However, there is an exhaustive report by Dorsey that was subsequently published by the National Advisory Committee for Aeronautics under the title, "On Self-Luminous Materials" (Dorsey, 1919a). It did not appear in print until 1919. Dorsey noted "The cumbrous title of the circular – "Notes and Recommendations regarding Specifications for the Illumination of Articles by Means of Self-Luminous Materials Containing Radioactive Substances" – indicates it purpose and scope." In April of 1919, he presented a paper at the American Physical Society "Studies of Self-Luminous Materials," with three co-authors (P.T. Weeks, T.B. Brown and E. Kung-Kwai[13]. There is no written record of the April APS talk, but it was probably a much-shortened account of the material in his report to the Aeronautics Advisory Committee.

Dorsey's comprehensive report for military users went far beyond basic metrology. It was intended to give the federal agencies all they needed to know about the nature of self-luminous paints and products. His team addressed six different aspects of the design, testing and use of the luminous artifacts:

- How self-luminous paints work.
- Which radionuclides are used.
- Deterioration of paints over time.
- Appropriate unit of brightness.
- Consideration of human eye perception for self-luminous artifacts.
- Quality assurance/quality control for federal agencies.

[13] One of the earliest known Asian American women to work at NBS, Elizabeth Yung-Kwai was the daughter of a Chinese American diplomatic family. In 1917 the Wellesley College student began work in Section 13 investigating self-luminous materials. She was included on the April 1919 paper, "Studies of Radium Luminous Materials," N.E. Dorsey, P.T. Weeks, T.B. Brown, and Elizabeth Yung-Kwai, that was presented at a meeting of the American Physical Society. After raising a family during the 1920s and 1930s, she returned to government service during World War II as a member of the Women's Army Corp. She died in 1984 in California at age 86 (NIST archives, 2023).

Dorsey reported that the light from the self-luminous sources (yellow-green to yellow in the visible spectrum (550 nm – 590 nm)) was the result of alpha particles incident on the zinc sulfide (ZnS) phosphor. About 0.000005 to 0.000010 radium to ZnS was required to produce a visible marker (Dawson, 1952). The relationship between the source radioactivity and the light output is more straightforward for a single alpha-particle emitting radionuclide such as polonium-210. Alpha particles travelling in the plane of a thin source will deposit energy to excite the phosphor. The situation is more complicated for the other nuclides such as radium-226 and radium-228. Decay of these nuclides leads to several alpha particles from the progeny, but some of the progeny nuclides can diffuse away from the surface prior to decay. And, depending on the decay schemes and the half-lives of the progenies the painted sources will "ripen" at different times depending on the radiochemical composition at the time the paint is applied to the product. Dorsey went to great lengths to explain how this works for polonium-210, lead-210-polonium-210, radium-228-thorium-228, thorium-228 alone and radium-226. But rather than including equations for the user, he provided qualitative information on how the brightness of a paint changes with time due to the radioactivity. The most important nuclides used in the paints were radium-226 and radium-228. For the radium-226 it was necessary to wait about one month for the radon-222 and short-lived progenies to come to an equilibrium condition. The initial increase in brightness of the "mesothorium," that is radium-228 that has been separated from thorium ores (thorium-232), is critically dependent on the degree of ingrowth of the 1.92 year $T_{½}$ thorium-228. In the absence of equations, Dorsey's description of the process is less helpful, although he does provide a figure showing that the alpha-particle intensity of a separated radium-228 source will attain a maximum value in about 5 years. It was well-reported that the mesothorium paints would "ripen" and increase in brightness with the ingrowth of the progenies.

The commercial paints were prepared by suspending the ZnS and radium in proprietary solvents. As noted in the Director's reports, NBS reverse-engineered the luminescent materials and prepared some of their own paints. These specimens and many of the fifty samples mentioned in the Director's Report were subjected to physical and chemical exposures to measure degradation with time. Increased temperature and reduced pressure were shown to decrease the emanation (radon-220 and radon-222), with a concordant decrease in alpha-particle scintillations from the surface. Actions of light, air, humidity and alpha-ray bombardment on the base material usually caused a decrease in responsiveness (Dorsey, 1919a). The brightness of a marker with time was thus a trade-off between the ingrowth of alpha-particle-emitting progenies and the degradation of the base paint by various means.

4.4.6. NBS Unit of Brightness

An important step in the metrology was Dorsey's selection of the unit of brightness:

> "By the brightness of a surface is meant the luminous intensity per unit area of the surface as projected on a plane perpendicular to the line of sight, it being assumed that the linear dimensions of the portion of the surface considered is negligibly small as compared with the distance at which it is viewed. It is evident that the brightness as thus defined is independent of the area of the luminous surface.
>
> As in measuring a length we choose for our unit a length that is exactly defined (foot, meter, etc.), so in measuring a brightness we choose as our unit the brightness of an exactly defined bright surface.

> For the measurement of the brightness of self-luminous materials a suitable unit is the brightness of a perfectly diffusing and completely reflecting surface when illuminated by a source of unit candlepower placed at a distance of 10 meters. This unit of brightness is called a *microlambert* because it is the one-millionth part of the lambert; the latter is a well-known and accepted unit of brightness. The microlambert is equal to 0.000 93 "equivalent foot- candles" and to 0.000 000 318 candles per square centimeter (3.18 millicandles per square meter)."

His report did not, however, include some other critical details about the photometric standard, the design of the photometer, and how the self-luminous sources were presented to the photometer. These missing details are given in papers published decades later by Dorsey's successor at NBS, Leon Francis Curtiss who published two papers on brightness meters in the 1930s (Curtiss, 1934, 1935) and critical reports from Dawson in the Photometry Branch, Optics Division at the Naval Research Laboratory in the 1950s (Dawson, 1952, 1955). These papers suggest that the primary standard of unit candlepower was provided by a tungsten lamp at 2360 K. The Consultative Committee on photometry changed the standard of luminance to 2042 K in 1950, but the higher temperature lamp was used in the earlier measurements of self-luminous sources (Dawson, 1955). Dawson reported that samples of self-luminous materials were submitted to NBS for certification. "The Bureau prepared its fundamental standard by illuminating a glass diffusion screen from behind a tungsten lamp of known candlepower and color temperature. An optical glass filter of known transmission, the color of which matched the radiation from the radioactive self-luminous material was interposed between the tungsten source and the diffusing screen (Dawson, 1955)". Samples calibrated at NBS in this manner served as secondary standards for the Navy and other users. In the later papers from Curtiss, he reports on measurements with glass tubes containing luminous samples, the brightness of which can be measured between 2 and 75 microlamberts (Curtiss, 1934). Dorsey's photometer had a narrower range but seemed adequate to report reference values to customers in the range of 10s of microlamberts.

Dorsey went on to distinguish the *brightness* of an object and the *total light*. "It is necessary to distinguish between the brightness of a luminous surface and the total amount of light the eye receives from it. The former is determined by the luminous material, while the latter depends also upon the area of the luminous surface. When the luminous area is small as compared with its distance from the eye it is the total light that gives the impression of brilliance and determines the ease with which the luminous area can be seen."

However, from the standpoint of perception with the human eye, the factor of major importance to the pilot trying to view dim self-luminous markings on an airplane cockpit dial was the brightness of the markings. Dorsey broke this into a series of problems and described these in the following manner (Dorsey, 1919a):

a) *Position marks. -Ma*rks that have to be seen, but of which the shapes do not have to be distinguished, require only a low brightness if they are to be viewed against an extended black background. Lines 1 millimeter wide and 4 millimeters long upon a black metal surface in moonlight are not readily seen in all positions at a distance of 75 centimeters (30 inches) if they are no brighter than 2 microlamberts. If the surface and the observer's eyes are shaded from the moon then such lines are seen without difficulty.

b) *Legible character.* -The luminous markings (numerals, letters, etc.) are of such a character that their shapes have to be distinguished, the brightness must be greater than that required for the mere seeing of position marks. Complex characters require a higher brightness than simpler ones of the same size. Numerals 8 millimeters high with a line width of 1 millimeter can be readily read in the moonlight at a distance of 75 centimeters (30 inches) if they have a brightness of 5 microlamberts.

c) *Relation of brightness to* size. -

There are at least four distinct phenomena that limit the distance at which a luminous character can be read:

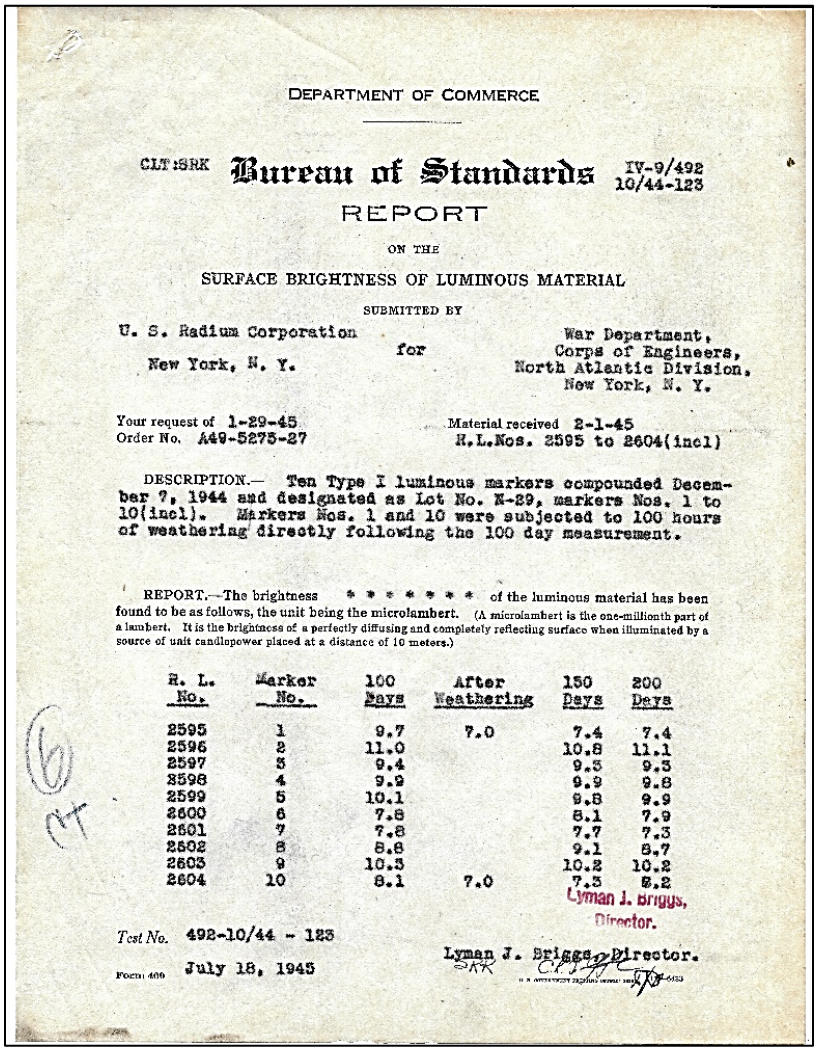
First. The resolving power of the eye. Unless the angle the character subtends at the eye exceeds a certain value, the shape cannot be recognized, however bright the character may be. This limiting angle varies greatly from eye to eye.

Second. The sensitivity of the eye. This also varies over a wide range and depends upon both the past and the present history of the eye. An eye that has recently been exposed to light is far less sensitive than if it had been kept in total darkness for some time; also if the eye is illuminated by an extraneous source of light its sensitivity is different from what it is when the eye is illuminated by the luminous character alone.

Third. Contrast. It is evident that the character cannot be distinguished if there is no contrast between it and the region bounding it. The percentage contrast in brightness that can be just distinguished increases as the brightness is reduced. For the range of brightness covered by articles illuminated by self-luminous materials this limiting contrast varies approximately from 5 to 20 per cent. That is, very faint markings will not be seen unless they are at least 20 per cent brighter than the surrounding surface. In most practical cases the lines are illuminated by the same light as the surface on which they are painted; their effective brightness is therefore enhanced. This, however, may not offset the ill effect of the light from the main surface of the article. For example, a white dial illuminated directly by the full moon might have a brightness of 20 microlamberts. If the marks upon it had a brightness of 1 microlambert and a coefficient of reflection as great as that of the dial their effective brightness would be but 21 microlamberts. This is but 5 per cent greater than the brightness of the main surface of the dial, and the marks would be almost if not entirely indistinguishable.

Fourth. A fourth limit is set by the total amount of light entering the eye from the character. Assuming that the conditions are such that this fourth limit is the only one operative, then for the same surface brightness a given character can be distinguished twice as far if all its dimensions are doubled; if its size is kept the same its brightness must be multiplied by 4 if it is to be distinguished at twice its former distance. Expressed otherwise, this fourth limit states that the distance at which a given character can be distinguished is proportional to its linear dimensions multiplied by the square root of its brightness. As the determining factors are the brightness and the angle which the object subtends at the eye, the effect of increased size can be secured by the use of a magnifying lens. (Dorsey, 1919a)”

NBS would continue to refine the metrological techniques for self-luminous materials for decades working in concert with the manufacturers and the military. The support that NBS provided to the federal agencies during WWI was a critical contribution to the war effort. As the Director reported in 1919, NBS provided services to the Bureau of Aircraft Production the Ordnance Department, the Engineer Corps, and the Inventions Section, General Staff, Army, the Bureau of Steam Engineering, the New York Navy Yard, and the Bureau of Lighthouses. After the war there were reorganizations at NBS that facilitated this work as the radium and radioactivity work and the photometry were incorporated into a new group in the Optics Division on Atomic Physics, Radioactivity and X Rays (Cochrane, 1956). The certification of surface brightness of luminous materials continued throughout World War II as shown by the report given here for samples submitted by the Army Corps of Engineers in April of 1945. The materials for test had been prepared for the Army by the U.S. Radium Corporation. The calibrations were performed by Constance Torrey in the Radioactivity Group (Coursey, 2021) and the certificates signed by NBS Director Lyman Briggs. The nominal brightness of the samples is of the order of 10 microlamberts and the results of time and weathering on the samples are noted in the report.



DEPARTMENT OF COMMERCE

CLT:SRK **Bureau of Standards** IV-9/492
10/44-123

REPORT

ON THE

SURFACE BRIGHTNESS OF LUMINOUS MATERIAL

SUBMITTED BY

U. S. Radium Corporation
New York, N. Y.

for

War Department,
Corps of Engineers,
North Atlantic Division,
New York, N. Y.

Your request of 1-29-45
Order No. A49-5275-27

Material received 2-1-45
R.L.Nos. 2595 to 2604(incl)

DESCRIPTION.— Ten Type I luminous markers compounded December 7, 1944 and designated as Lot No. N-29, markers Nos. 1 to 10(incl). Markers Nos. 1 and 10 were subjected to 100 hours of weathering directly following the 100 day measurement.

REPORT.—The brightness * * * * * * * of the luminous material has been found to be as follows, the unit being the microlambert. (A microlambert is the one-millionth part of a lambert. It is the brightness of a perfectly diffusing and completely reflecting surface when illuminated by a source of unit candlepower placed at a distance of 10 meters.)

R. L. No.	Marker No.	100 Days	After Weathering	150 Days	200 Days
2595	1	9.7	7.0	7.4	7.4
2596	2	11.0		10.8	11.1
2597	3	9.4		9.3	9.3
2598	4	9.2		9.9	9.8
2599	5	10.1		9.8	9.9
2600	6	7.8		8.1	7.9
2601	7	7.8		7.7	7.3
2602	8	8.8		9.1	8.7
2603	9	10.3		10.2	10.2
2604	10	8.1	7.0	7.3	8.2

Lyman J. Briggs,
Director.

Test No. 492-10/44 - 123

Form) 409 July 18, 1945

Lyman J. Briggs, Director.

Figure 15. Sample report by NBS on ten markers submitted for measurement of surface brightness.

The luminous dial paints also appear prominently in the tragedy of the radium dial painters (Mullner, 1999). The NBS work with the manufacturers and public health agencies on the metrology and health aspects of the radium dials has been described elsewhere (Coursey, 2021).

A final summary of the NBS studies on self-luminous materials was prepared after WWI in a special publication on War Work of the Bureau of Standards (Bureau of Standards, 1921). There is a good deal of overlap between this summary and the Director's Annual Reports from 1916 to 1921, but it is an excellent overview of the rationale for the NBS work in the different categories. The technical details are better provided in Dorsey's report to the Aeronautics Advisory Committee (Dorsey, 1919a).

4.5. X-Ray work at NBS

Dorsey mentioned in his June 4, 1913, letter to Boltwood his meeting with the x-ray scientists in New York and that he saw potential for future work along those lines (Yale, 2023). But the NBS entry into x-ray measurements was long delayed due to the priority of the radium measurements. The contrast in priorities is striking between the US (NBS) and Europe. In France, Marie Curie and her daughter Irene turned their attention to helping the French government field a large contingent of mobile x-ray systems for the war effort (Pasachoff, 2000). Curie was named Director of the Red Cross Radiology Service and trained nurses, radiological technicians, and US servicemen in the use of x rays. She was unable to return to radium research until the war's end. In the UK, Dorsey's counterpart at the NPL, G.W. C. Kaye was called to military duty and spent the war years leading British efforts on x-ray developments for the military (Kaye, 1921). In his 1921 textbook on x rays Kaye made this statement concerning the impacts of x-ray radiography. "The late war brought this home in unexampled fashion, and no man can over-estimate the services which radiology rendered in the great war tragedy. While human endeavor reached its maximum in almost every phase of life, a word may be spared in recognition of the way British radiologists and British x-ray manufacturers flung themselves into the gigantic task of expansion (Kaye, 1921)." One can contrast this with Dorsey's annual report in June of 1920 noting that all the requests for x-ray work were delayed because of the push for routine (but lucrative) radium calibrations.

4.5.1. Director's Annual Report, June 30, 1918

> "The X-ray equipment purchased in the spring of 1917 was installed in July. The apparatus has been studied in some detail, subsidiary equipment has been constructed and purchased, the routine testing of protective materials has been established, and miscellaneous radiographs have been taken for the purpose of obtaining information regarding the internal structure of materials and articles.
>
> In its endeavor to secure the domestic manufacture of improved protective materials, the Bureau met with splendid cooperation on the part of the manufacturers. As a result, one can now obtain commercial material which gives nearly twice as much protection per unit thickness as was given by material obtainable a year ago. (Improvement in glass reaches 80 per cent, in rubber 100 per cent.) A note on this work has been published in the American Journal of Roentgenology, and a more detailed paper has been prepared for the same journal.

> A study of the technique for the radiographic detection of flaws in aluminum and in steel is still in progress."

The x-ray equipment mentioned was a Waite-Bartlett transformer and high-voltage mechanical rectifier (Taylor, 1981) and the publication referred to was in the American Journal of Roentgenology (Gorton, 1918). W.S. Gorton and others were working in Dorsey's Section at the time and Dorsey presented some of their work at an American Physical Society meeting in 1919[14]. L.S. Taylor's comprehensive review of US standards for x rays 1913 – 1964 has a short description of the work during WWI and the details are as useful as the 8-page paper by Gorton.

> "The first problem was to determine the effectiveness of lead glass used in fluoroscopes and *protective screens. At that time, glass was purchased on specifications which merely required that it be "adequate" or "sufficient" for protection under unspecified conditions.*
>
> A system was set up for comparing the lead glass with sheet lead of various thicknesses and judging the lead equivalence of the glass by the blackening of a photographic plate. Some of the samples submitted were found to be plain window glass, but the better grades of material showed a lead equivalence in the range of 1/2 mm of lead. For the study, the Bureau acquired its first x-ray equipment in 1917, a Waite and Bartlett transformer and a high-voltage mechanical rectifier. Dorsey described the equipment as having a "voltage ranging from 3" to 9" spark gap." He was referring, presumably, to a so-called needle gap, in which case the range would have been in the order of 65- to 135-kV peak. When reporting the results of this work at a meeting of the Western Roentgen Society in Chicago, the Bureau made its first public request for the assistance of the radiological societies to set up its x-ray programs. At that time (1918 or 1919), the Western Roentgen Society was just forming. In fact, the Society was the forerunner of the Radiological Society of North America (Taylor, 1981)".

Gorton did not report the peak kilovoltage achieved with their system although Taylor surmises that the "3 inch to 9 inch spark gap" would have produced peak x rays of the order of 65 to 135 kV[15]. They set about using the new system to solve some practical problems for the emerging radiology market, that is, to measure protective properties of leaded glass and leaded rubber shielding materials. They compared the attenuation of the x rays with a lead step wedge. The scales had steps 1 cm wide and thicknesses of one step to the next was 0.1 mm for a total thickness of 0.5 mm. The exposure was captured on a photographic plate with the material under test alongside the lead step wedge. A piece of material was said to be equivalent in "protective power" to a thickness of lead giving the same density on the photographic film. This thickness was called the "lead equivalent" (Gorton, 1918). This procedure worked satisfactorily for lead-impregnated rubber samples, but leaded glasses presented a problem as scattered radiation from other components in the glass also exposed the film. This was easily corrected by inserting filters

[14] "Nina M. Alderton (Moore) had undergraduate degrees in physics and mathematics from Mount Holyoke College and a master's degree in mathematics from Columbia University when she arrived at NBS to study x-ray protective materials. After the war she earned her Ph. D. in mathematics at U.C. Berkeley and taught at various colleges before returning to NBS from 1945-1954, working in the Temperature Measurements group."

[15] The Waite Bartlett x-ray system was developed by Dr. Henry E. Waite whose company manufactured medical equipment and equipped the first U.S. Hospital ship the US Solace in 1914, and in 1916 developed and manufactured a US Army medical bedside x-ray unit (US Army, 2023). By the end of the war the US Army Medical Corps had a large presence in military radiology (Amis, 1996) but there is no evidence that they reached out to NBS for assistance.

to remove the low-energy radiations. As noted in the Director's reports, the NBS assistance to industry quickly led to great improvements in the protective materials.

This new capability at NBS could also be used for industrial radiography as well as assistance in materials developments. Wadleigh prepared a summary of the physics of x rays and radium for a medical journal in 1921 but it was at the introductory physics level and added nothing to NBS metrology (Wadleigh, 1921). It is striking to compare the NBS facilities to those at the same time at the UK National Physical Laboratory. Kaye reported that the NPL had a battery of x-ray tubes with different anodes so that a variety of homogeneous rays could be obtained (Kaye, 1921). They routinely measured various x-ray spectra using different anodes and filters of molybdenum and other high Z elements (metallic salts as well as sheets). The medical physicists at the NPL were collaborating with their peers and advancing radiology and radiation protection in the UK. NBS would not make such investments for another decade (Taylor, 1981).

4.6. Dorsey resignation

By the Spring of 1920 Dorsey decided for several reasons that he could no longer continue with the radium work and submitted his resignation in a letter to Rosa on April 26, 1920 (shown below in full). The chief reason was that he was completely burned out – mentally and physically – from work with radium. He also wanted to establish a private consulting practice and he saw that as a clear conflict with his government service. He did not mention that he intended to write a textbook on radioactivity for medical professionals which was published the following year (Dorsey, 1921a). As Slaughter has observed, Dorsey's book was the first complete treatise on radioactivity that included cautions on the hazards of radiations, and likely initiated improvements in radiation health physics for the nation over the next decade (Slaughter, 2013).

The full text of Dorsey's letter is given here.

Dr. E.B. Rosa, Chief Physicist,
Chief, Division I,
Bureau of Standards,
Washington, D.C.

April 26, 1920.

Dear Dr. Rosa:

I hearby tender my resignation as Chief of Section 13 of Division I and as Physicist in the Bureau of Standards, my resignation to take effect at your discretion on or before the close of work on June 30, 1920.

Among the various causes that have contributed to my decision to take this step, two only need be mentioned.

First, the continued strain of work under high pressure and over long hours that has been necessitated by the conditions existing during and since the war has taken a heavy toll from my physical vitality until now I am running dangerously near a breakdown. In addition to this, perhaps on account of this condition, the effects characteristic of continued exposure to

radium radiations have developed rather rapidly during the past winter. As a result, a relief from the strain, a rest, and removal from exposure to radium radiations are almost imperative and should not be postponed longer than is absolutely necessary.

Second, I have for some time desired the opportunity to carry on certain lines of personal work; to go into business for myself. This can not be done, our even tried out satisfactorily until I am relieved from my duties at the Bureau. One can not serve two masters.

While I shall in many ways regret the severance of my connection with the Bureau, the step I am now taking has been considered carefully and I believe it is justified by the totality of the existing conditions. I shall always have a keen interest in the welfare of the Bureau and shall remember with pleasure the many enjoyable associations that bind me to it.

Respectfully,

N. Ernest Dorsey
Chief of Section 13.

5. Standards 1921

5.1. Introduction

1921 was the most significant year of the new century for the radium researchers at NBS. Marie Curie had been persuaded by an enterprising American journalist, Marie Brown Meloney, to make her first visit to the US on a publicity tour where she would receive a gram of radium paid for by donations from the Women of America. On their first meeting, Marie Curie told Meloney that the US had 50 grams of radium while she had only one gram. She reported that Baltimore had four grams, Denver had six grams, New York had seven grams, and went on to name the location of every gram (Eve Curie, 1938 p. 323). Slaughter lists five US centers in 1919 that had gram quantities of radium: the Kelly Hospital in Baltimore, Huntington (Harvard) Hospital in Boston, Memorial Hospital in New York, Simpson Radium Institute in Chicago, and Mayo Clinic in Rochester, Minnesota. Badash reported that in 1921 seven US centers had more than ½ gram radium (Badash, 1979 p. 134). All of these were operating radon emanation plants for cancer therapy (Slaughter, 2013).

5.2. Radon Emanation Plants

It was recognized early that radon-222 gas, separated from its parent radium-226 in solution form, provided a very effective gamma-ray source. The emanation was particularly useful in therapy as the separated gas could be sealed in small sources and used directly in interstitial and intracavitary applications. Boltwood had made a small generator for Howard Kelly and Lantsberry made a second generator for the Baltimore clinic when he came in 1914 (Kelly, 1918). But the major credit for the emanation plants goes to the Curie laboratory where the chemist André Debierne developed a radon extraction facility for the Curies, and the physical chemist/physicist William Duane developed the “radium cow” which was widely copied in Europe and the US (Brucer, 1993). In fact, Marie Curie reported to Meloney that the *one gram* at her institute in 1920 was devoted to cancer research (Curie, 1938).

William Duane studied physical chemistry with Walther Nernst in Göttingen in Germany and then received his Ph.D. in physics in 1897 from the University of Berlin under the direction of Max Planck (Brucer, 1993). He was a professor of physics at the University of Colorado and in a sabbatical year 1904 – 1905 he spent time with the Curies in Paris and with J.J. Thompson at the Cavendish Laboratory. In 1905, Pierre Curie requested a grant from Andrew Carnegie to allow Duane to work at their laboratory. Following Pierre's death in 1906, Marie Curie received the grant and Duane returned to the Curie laboratory for another six years. One of his major accomplishments during this period was his implementation of the "radium cow," as Marie Curie trusted the physical chemist Duane to handle the radon from the relatively large amount of radium at her institute (Duane, 1915; Duane and Greenough, 1918; Brucer, 1993)[16]. Slaughter reports that Duane was generous in crediting Rutherford and the chemists Debierne, Ramsay and Soddy for their contributions (Slaughter, 2013). When Duane returned to the US he designed and constructed an emanation plant of 920 mg of radium element for the Huntington Memorial Hospital in Boston (Brucer, 1993). Duane recommended that the Memorial Hospital in New York hire a young physicist, Gioacchino Failla in 1915 (Marinelli, 1962). The generator that Failla constructed in New York contained 3.56 grams of radium (Harrington, 1944; Brucer, 1993).

A radon generator containing one gram radium in solution, assuming an 85 % efficiency of sweeping and purifying the gas, would yield about 31.4 GBq (850 mCi) of radon-222 (Harrington, 1944). After about 4 days of radon ingrowth the generator would have of the order of 18.5 GBq (500 mCi) of radon. Thus, assuming the same 85 % collection efficiency, the generator could produce about 15.7 GBq (425 mCi) of radon every 4 days. This could be subdivided to prepare sources for therapy of 74 MBq to 1.85 GBq (2 mCi to 50 mCi). About ½ gram radium in solution was needed for a typical emanation plant (Slaughter, 2013). Among Failla's many other accomplishments at Memorial Hospital he designed gold seeds to contain the radon (Robison, 2015). The glass ampoules previously used could not stop the short-range beta particles that led to radiation damage immediately adjacent to the source. A gold tube, typically 0.15 mm in inner diameter with a wall thickness of 0.30 mm, could be attached to the gas manifold. The tube was preformed into sections allowing multiple sources to be prepared by crimping the seeds along the tube (Harrington, 1944). The length of the seed could be adjusted to give higher activity sources.

From the perspective of radium metrology, all the therapeutic irradiations were traceable to calibrations at NBS against the 1913 International Secondary Standard No. 6 (Figure 5) and a subset of in-house standards that NBS had intercompared with Standard No. 6. The gram quantity sources used in the emanation plants were shipped from the manufacturers to NBS and calibrated by Elizabeth Damon, Mary Brower and Constance Torrey prior to shipment to the hospitals. (Figures 7 and 11). Burnam mentions that the 750 mg of radium received at the Kelly Clinic in Baltimore was packaged in 50 mg glass tubes which were calibrated by Dorsey at NBS (Burnam, 1936). This calibration was probably by electroscope comparison with the 49 mg secondary standard that NBS had from the US Bureau of Mines[17]. For larger quantities of

[16] When Marie Curie visited the US in 1921, she was asked what she wanted to see in the US. She replied: Niagara Falls, the Grand Canyon and William Duane.

[17] On May 28, 1921, Samuel Lind, at the time at the US Bureau of Mines in Reno, NV, wrote to his colleague in Washington with more information about the two sources that the USBM had provided. The smaller was identified as 48.7 mg radium element and the larger as 91.8 mg radium element. He reported that one of the sources had

radium, in 100 mg lots, NBS could use the 91 mg secondary standard which had also been provided by the USBM. Dorsey preferred that source calibrations be carried out by comparison with a standard that was within a factor of two in activity of the sample under test. The accuracy of the quantity of radium received from the manufacturer was important because of the high cost of the material, of the order of $100,000 per gram.

But the accuracy in patient treatment depended on an assay of the source used in the therapeutic procedure. The radon seeds prepared from the emanation plants were calibrated by the hospitals by gamma-ray comparisons with in-house secondary radium-226 sources that were traceable to the NBS standard source (Harrington, 1944). Attenuation corrections had to be applied for the appropriate encapsulation, gold wall for the radon seeds and platinum tube for the radium needles. Decades later the US National Cancer Institute would order hundreds of radium needles with directions that they be calibrated at NBS and delivered to hospitals all over the US (NIST archives, 2023).

5.3. Preparation for Marie Curie visit to NBS

After the Armistice in 1918 Marie Curie was able to return full time to her radium work. Over the following two years almost all the radium in Europe was provided by the US suppliers, mainly the Standard Chemical Company in Pittsburgh. Buyers in England and France would routinely have the radium mass assays verified by the NPL or the Curie Laboratory. On one or two occasions Curie contacted NBS to question some of the assays. On February 6, 1919, Dorsey wrote to Marie Curie to answer her query about the accuracy of some radium sources that had been shipped to France. He pointed out that NBS had used the standard that she had certified in 1913 (Standard No. 6) and asked a series of questions about the sources received at the Curie laboratory. Were the sources under the seal of NBS? Did they appear to have been opened? Did the weights and dimensions of the sources match the information on the certificate provided by NBS? As it happened, the sources had been opened and re-packaged by clerks at the Banque du Radium and the loss of material in the transfer accounted for the discrepancy with NBS values (NIST Archives; Coursey et al. 2002).

Marie Curie was scheduled to arrive in Washington on May 21, 1921. In the weeks before her arrival she contacted NBS again with a question on some of the assays. Rosa at NBS contacted Ernest Rutherford, the president of the International Radium Standards Commission on the matter by letter on April 4, 1921. Rutherford replied on April 19, 1921. His letter is given here.

One-page typed letter of April 18, 1921 from E. Rutherford

Cavendish Laboratory,
Cambridge,

Dear Rosa,

I have just received your letter of April 4th re Radium measurements, and also copy of the letter you have sent to Madam Curie. In the absence of a National Laboratory in France, I

previously developed a leak and that Dorsey had sealed it within another tube. He did not know which source developed a leak, but suspected it was the larger one.

understand that the measurements are made by Madam Curie who must take the responsibility of their accuracy. Apart from the N.P.L. which has the official standards, I do not feel that the measurements of any other Institution are to be taken seriously. For example, I refer to the London Radium Institute; I am not aware that they have either the staff or apparatus for accurate measurement. I am of the opinion that the relative measurements should not have a greater error than ±1%. The result of the comparison of the earlier standards showed on the average a much greater accuracy than this.

I note what you say about the relative measurements of the Bureau and Madam Curie. I have no doubt that she will have looked into the question and given a reply. I think it would be unlikely that the standards in the different countries are seriously in error and the difference of the magnitude you mention must be ascribe either to carelessness of measurement or some other extraneous cause.

If you feel any serious doubt about the possible error in standards, say in the Bureau and Paris, it would not be a difficult matter to interchange a secondary standard for comparison with the official standards. Such a comparison would be of interest in throwing light on the accuracy of measurement both possible and probable. This however is a matter to be dealt with between the National Laboratories.

I shall be at your service if you wish to discuss the matter further.

Yours sincerely,
(signed) E. Rutherford

He reassured Rosa that comparisons between the national metrology institutes should be in agreement assuming an uncertainty no greater than ±1% on a given value. It is interesting that he expressed little confidence in measurements by the London Radium Institute (Ramsay and Soddy were on their Board of Directors). He may not have been aware that the Institute for Radium Research in Vienna had sent one of the 1912 secondary standards to Dr. Alton at the London Radium Institute. He suggested an exchange of a calibrated source between Washington and Paris if there were still any doubts about the agreement between the laboratories. But he seemed to indicate that this was a dispute the two laboratories should settle. Rutherford's reply seems consistent with his role as the president of the commission whose laboratory was not directly involved in standards measurements. About one month later May 18, 1921, NBS Director Stratton replied to Rutherford and explained that another opportunity for a comparison had indeed come up and the results showed good agreement between NBS and the Curie laboratory.

His reply is given here in full (NIST archives, 2023).

Typed two-page letter of May 18, 1021 from NBS Director

ECC:MBJ
I-O May 18, 1921

Sir Ernest Rutherford,
Cavendish Laboratory,
Cambridge, England.

Subject: Radium Measurements in Different Countries

Dear Sir Ernest:

We have received your letter of April 18, in reply to ours of April 4, regarding measurements of radium in various laboratories. We were not acquainted with the London Radium Institute, and did not know how much weight to give the results reported by that institution.

Since our letter was written we have received a report from Madame Curie regarding measurements of one more preparation. In this case special care was taken in the measurement made in the Curie Laboratory and the results agreed with ours within a small fraction of one per cent.

Exceptional opportunity for comparison with Madame Curie will be afforded by measurements of the gram of radium which is to be presented to her during her visit to America. This material is to be sent to our laboratory within a few weeks. We understand that it will be made up in ten separate preparations, and there will therefore be opportunity to find very exactly the relation between average results here and in Madame Curie's laboratory. The only difficulty will be that the preparations will not have attained equilibrium when we measure them, and that consequently values will have to be extrapolated somewhat.

In general the information we have been able to obtain confirms your opinion that the relative measurements should not have a greater error than one per cent. It is only in the case of one preparation that the difference between Madame Curie's measurements and our own has been larger than this. We have, however, been able to get definite figures on only a few samples and consider it

May 18, 1921

S.E.R. -2.

significant that the differences between the Curie measurements and our own appear to be always in the same direction. We shall be glad to write you further as soon as there is available more definite information regarding comparative results.

Very sincerely yours,

(Signed S.W. Stratton)
Director
(initials ECC)

The timing and sense of urgency in this correspondence is interesting. Curie was scheduled to arrive in Washington on May 20, 1921, to visit the White House, NBS and the USBM and to receive a gram of radium calibrated at NBS. NBS Director Stratton and the Chief Physicist Rosa wanted to make sure everything was in order before her visit. Dorsey had taken a leave-of-absence the previous Summer and his replacement Walter Hiram Wadleigh did not have the experience of his predecessor.

5.4. Marie Curie's visit - A gram of Radium

The Marie Curie Fund intended to raise $100,000 to purchase a gram of radium for the Curie Institute (about $1.7M in 2023 USD). The Fund placed the order with the Radium Chemical Company and the responsibility for preparation of the material was assigned to the chemist Glenn Kammer (Lubenau and Landa, 2019). Kammer started with 2 grams of radium to make sure that he had sufficient material and subdivided Curie's gram into ten 100 mg aliquots in glass tubes. Photographs of him "tubing" the radium at the time show the lack of proper attention to radiation safety at the time, which in fact led to his death at age 39 from aplastic anemia due to excessive radiation exposure (Silverman, 1927). Only 3 days before the visit was to occur, Stratton had written to Rutherford that the gram of radium was *expected within a few weeks*. In fact, the Radium Chemical Company was keen to have the material delivered to Washington, that is to NBS, in time for the White House ceremony. The gram of radium remained at NBS while a replica of the storage container and the individual needles were used in the ceremony (Lubenau, 2012).

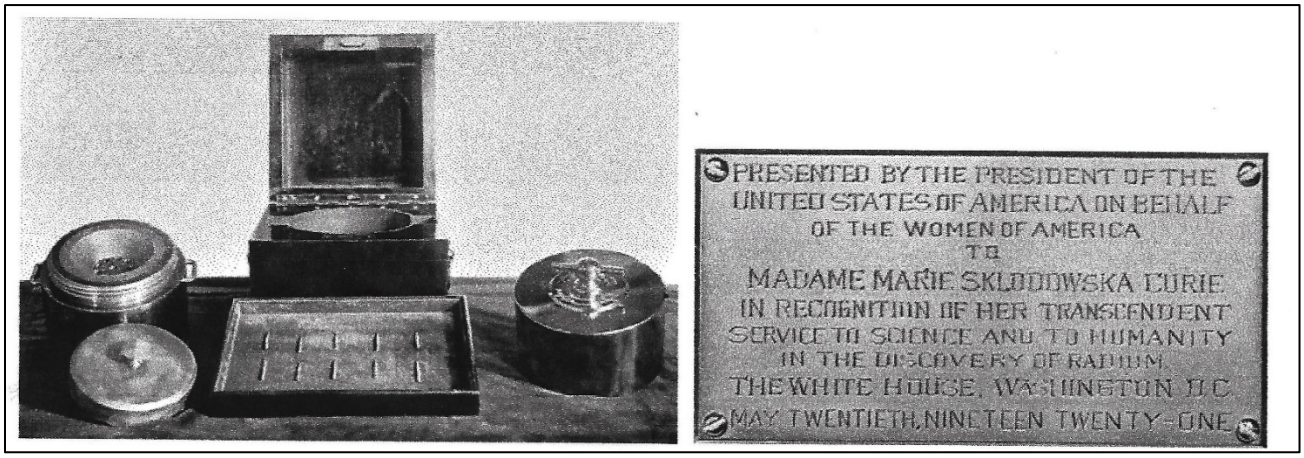


Figure 16. The shipping container for the 10 glass ampoules each containing approx. 101 mg of radium and the engraved plaque (photo from Lubenau and Landa, 2019).

The White House prepared a fact sheet for visitors and press for the event. A draft of the fact sheet was probably prepared by NBS and provided to the White House press office for review and approval. The fact sheet is reproduced here in full.

Facts about the Curie Radium

Early in the year 1921 an association of American Women known as the Marie Curie Radium Fund was formed in this country under the Chairmanship of Mrs. Wm. B. Meloney, and with headquarters in New York City. The purpose of this organization was to solicit subscriptions to a $100,000 fund with which to buy a gram of radium for presentation to Mme. Marie Sklodowska Curie, the eminent Polish-French scientist and codiscoverer of radium on her coming to this country.

On opening the competitive bids the contract for furnishing this gram of radium was finally let to the Radium Chemical Co, Pittsburgh, Pa. The material was put up in ten hermetically sealed glass tubes, each about 3.8 mm. in diameter and 38. mm. long, and containing an average of 101. milligrams of anhydrous radium barium bromide. These were each enclosed in two other glass tubes bearing designating numbers and data concerning the preparation, and all enclosed in a lead cylinder one and one-half inches thick, the whole being encased in a beautifully polished mahogany case on the cover of which is a gold plate 3 x 5 inches bearing the inscription.-

Presented by Warren G. Harding, President of the United States of America on Behalf of the Women of America to Mme. Marie Sklodowska Curie in Recognition of her Transcendent Service to Science and Humanity.

At the White House, Washington, D.C.

May 20, 1921.

Following the presentation to Mme. Curie at the White House at 4:00 o'clock on the afternoon of May 20, this case and contents were delivered at 6:15 P.M. to W. H. Wadleigh, Associate Physicist and Acting Chief of the Radium Section of the United States Bureau of Standards, for measurement and certification, and receipted for by him. Here it underwent thorough and repeated measurements. With the aid of Secretary H.D. Hubbard of this Bureau a special certificate was designed by Mr. Wadleigh and engrossed by Mr. C.A. Wickerly of the U.S. Geological Survey, on Strathmore grained white cardboard tinted to a light buff with air brush and shaded with lamp black, the result being an artistic and beautiful piece of work. On June 24 the radium and case with this certificate were taken to New York by W.H. Wadleigh, and delivered to Purser C.P. Lancaster of the R.M.S. Olympic by whom it was carefully stowed away in the special room of the liner for safe keeping during the voyage to France.

The special certificate designed for the occasion is shown here. The nominal mass values of radium element in each ampoule were about 101 mg. The White House fact sheet is misleading in that the 101 mg in the glass tubes are said to be anhydrous radium barium bromide. In fact, as the certificate shows they contained 101 mg radium element. The Radium Chemical Company knew the total mass of the mixed alkali metal halide in each ampoule from Kammer's measurements, but NBS did not include this information on the certificate. On receipt of the material at NBS the staff in the radium section, probably Constance Torrey under the supervision of Wadleigh, used the gold-leaf electroscope to compare them with either the original 15.44 mg radium standard or possibly the 91.8 mg radium standard that they had acquired from the USBM earlier. The values on the certificate are the NBS values. Since Kammer had only recently filled

the 10 ampoules from the raw material, corrections had to be applied because the radon was not in equilibrium at the times of the measurements in Pittsburgh and Washington.

UNITED STATES OF AMERICA
Department of Commerce
Bureau of Standards - S. W. Stratton, Director

CERTIFICATE
FOR
RADIOACTIVE MATERIAL

Submitted for measurement and certification.

By the Radium Chemical Company, Pittsburgh, Pa. Distributers

For the Standard Chemical Company, Pittsburgh, Pa. Makers

And presented by
THE PRESIDENT OF THE UNITED STATES OF AMERICA
ON BEHALF OF THE WOMEN OF AMERICA
to
Madame Marie Sklodowska Curie,
Paris, France.
May 20, 1921.

This certifies that each specimen of the above mentioned radioactive material, as described below, has been compared with the Radium Standard of the National Bureau of Standards, and that from the data so obtained the equilibrium intensity of the measurable gamma radiation has been computed to be equivalent to that from a quantity of radium whose mass is specified in the right hand column of the following table of values.

Designating numbers of tube*			External dimensions of tube (millimeters)		Combined weight of tube and contents (milligrams)	Computed radium element equivalent° (milligrams)
Assigned by makers	Assigned by the Bureau of Standards: Serial	Assigned by the Bureau of Standards: Test	Length	Diameter		
1	4082	Tex.31302-40	41.9	3.77	554.7	100.7
2	4083	" " 41	35.0	3.66	465.3	101.3
3	4084	" " 42	38.5	3.91	541.9	100.9
4	4085	" " 43	40.5	3.82	552.6	101.0
5	4086	" " 44	38.5	3.76	517.7	100.9
6	4087	" " 45	37.7	3.92	537.5	100.5
7	4088	" " 46	36.0	3.62	479.3	101.2
8	4089	" " 47	36.8	3.81	505.6	100.9
9	4090	" " 48	39.4	3.84	546.9	101.1
10	4091	" " 49	33.6	3.72	462.8	101.6

° *The computed radium element equivalent specified in the right-hand column represents a quantity of radium in radioactive equilibrium with its first four derivatives, being hermetically sealed in a tube of Thüringian glass having walls 0.27 millimeters thick.*

The residual uncertainty in the numerical value of the radium element equivalent of these several specimens does not exceed 0.7 of one percent.

Observations extending over 25 days indicate that the radium contained in these specimens is now in radioactive equilibrium.

* *The radioactive material herein certified is divided into ten lots each sealed in a separate glass tube and designated by the respective numbers shown in the first three columns of the above table. The Bureau is advised by the makers that the material in each of these preparations is anhydrous radium barium bromide. When the measurements had been completed, each specimen, with a label giving the number assigned by the makers, the serial and test numbers assigned by this Bureau, and the computed radium element equivalent, was enclosed in a larger glass tube. Each tube so enclosed was then placed in the cavity designed for it in the lead lined, cylindrical steel case provided by the makers, and the whole secured by a seal of this Bureau.*

Washington, D. C., June 22, 1921.

S. W. Stratton
Director

Figure 17. Certificate for one gram of radium-226 presented to Marie Curie in 1921 (NIST archives, 2023).

Marie Curie's arrival in Washington a century ago was a major social event. The Washington Sun Star (1921) listed the 8 scientists and 18 prominent local women who were part of the local organizing committee for her visit. The Director of NBS – Samuel Wesley Stratton – was a member of the scientific committee, along with Charles Parsons and Richard Moore, chemists from the USBM. The Executive Committee of women included Mrs. Herbert Hoover – future first lady – and Alice Roosevelt Longworth – first daughter of President Teddy Roosevelt and grand dame of Washington Society for most of the 20th century. These Washington women were also members of the Executive Committee of Women for the Marie Curie Fund.

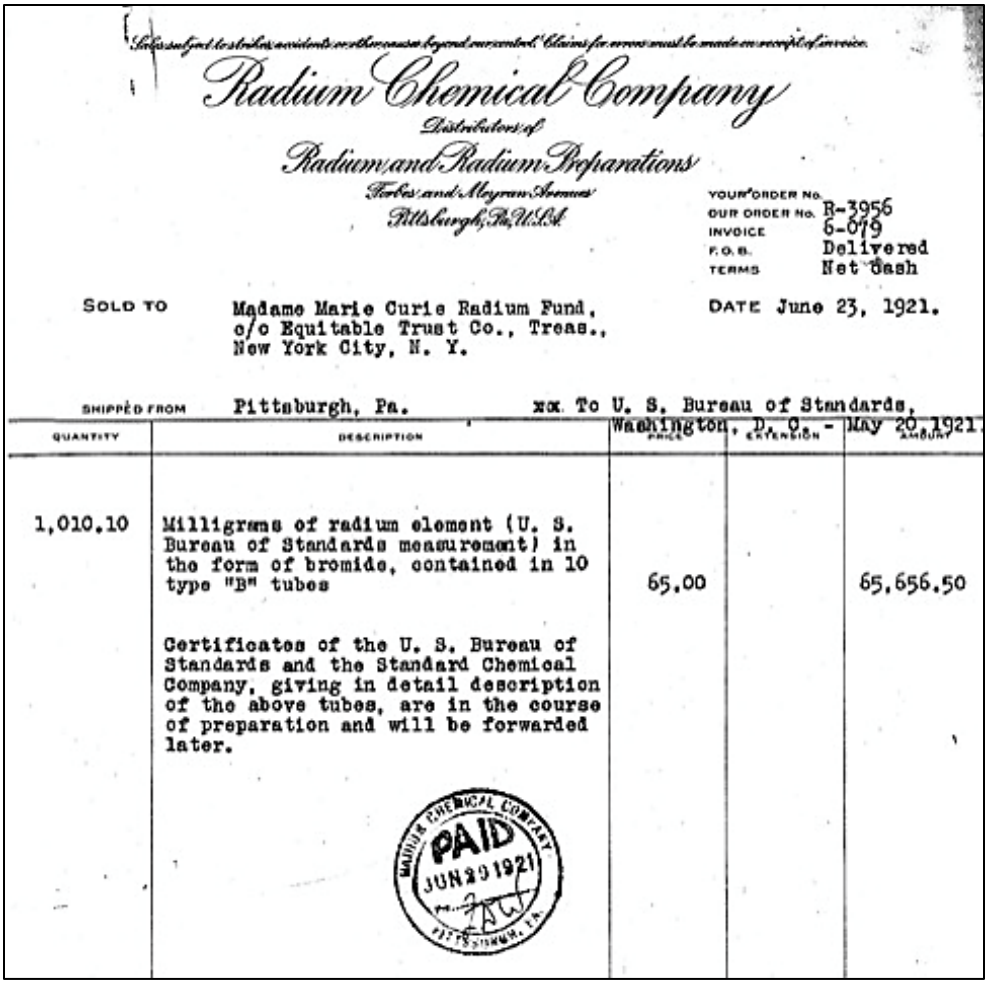
Curie arrived on a sleeper train from New York at 6:30 AM Friday morning on May 20th. She had a little time to rest at the home of a local diplomat and then attended the White House ceremony in the afternoon. That evening there was a large reception in her honor at the National Museum (soon renamed the Smithsonian Institution). Saturday, she planned to attend two scientific institutions, the USBM and NBS, and then take a sailing trip on a Navy Launch down the Potomac River to Mt. Vernon, followed by dinner at the French Embassy. The only event scheduled for Sunday was a dinner at the Polish Legation.

Wadleigh is identified as Acting Chief of the Radium Section. This was a time of reorganization at NBS, and the radium work was soon incorporated into a new Atomic Physics and Radium Section in the Optics Division under Paul Foote. Years later Foote recalled Marie Curie's visit to NBS. "We used to have a great many visitors… I remember Madame Curie calling one Sunday morning to visit the spectroscopic and atomic physics laboratories. To our surprise she was accompanied by her younger daughter Irene who became much interested in spectroscopy. With such an excellent beginning, it is only natural that years later Irene became a Nobel laureate like her mother (Foote, 1966)."

This note from the NBS archives is important because it shows that Marie Curie visited NBS on a Sunday morning rather than the scheduled Saturday visit. On Saturday she made the planned visit to USBM, where she participated in commissioning a new low-temperature laboratory for the production of liquid helium. The Washington Herald (1921) reported on her visit to NBS the following day that was hosted by NBS Secretary H.D. Hubbard. (Director Stratton was away at the time.) Marie and Irene met with W.H. Wadleigh, E.C. Crittenton, W.F. Meggers, W.W. Coblentz and P.D. Foote to discuss photometry, radiometry and atomic physics as well as the radium work. She later remarked "I had the great pleasure of meeting in their laboratories (also referencing the USBM tour) several very important American scientific men. The hours I spent in their company are among the best of my travel" (Curie, 1923).[18]

The invoice that the Radium Chemical Company submitted to the Marie Curie Radium Fund is shown in Figure 18. The material was supplied with a certificate from the "Bureau of Standards." The company had won the bid for $100,000 (about $1.7M in 2023 USD), but in the end, they only billed the Marie Curie Fund $65,000 for their costs of production.

[18] There is no mention of Marie Curie meeting N.E. Dorsey, who was a consultant at the time of that unscheduled Sunday morning visit. However, it seems likely that Dorsey saw her given the physical description he provides in the paean he published in a national news weekly: "Marie Curie, the most famous woman in the world." (Dorsey, 1921c).



Radium Chemical Company

Distributors of

Radium and Radium Preparations

Forbes and Meyran Avenues

Pittsburgh, Pa., U.S.A.

YOUR ORDER No.
OUR ORDER No. R-3956
INVOICE 6-679
F.O.B. Delivered
TERMS Net Cash

SOLD TO Madame Marie Curie Radium Fund,
c/o Equitable Trust Co., Treas.,
New York City, N. Y.

DATE June 23, 1921.

SHIPPED FROM Pittsburgh, Pa. xx To U. S. Bureau of Standards, Washington, D. C. - May 20 1921.

QUANTITY	DESCRIPTION	PRICE	EXTENSION	AMOUNT
1,010.10	Milligrams of radium element (U. S. Bureau of Standards measurement) in the form of bromide, contained in 10 type "B" tubes	65.00		65,656.50
	Certificates of the U. S. Bureau of Standards and the Standard Chemical Company, giving in detail description of the above tubes, are in the course of preparation and will be forwarded later.			

MADAME CHEMICAL COMPANY PAID JUN 29 1921 PITTSBURGH, PA.

Figure 18. Invoice to the Marie Curie Radium Fund from the Radium Chemical Company Pittsburgh for a gram of radium held at the U.S. Bureau of Standards (NBS). The invoice was dated June 23, 1921, for the material delivered to NBS on May 20, 1921. The invoice was paid on June 29, 1921.

The final responsibility of NBS was to deliver the gram of radium to the Olympic sailing ship in New York for the return to France. A good description of this transport appeared in a Scientific American article a few weeks after her visit (Lescarboura, 1921). Wadleigh had the unenviable job of transporting the 60 kg (130 pound) box containing radium valued at $100,000, equivalent to about $1.7 million in 2023.

5.5. Death of Edward Bennett Rosa

Tragically, Marie Curie did not have an opportunity to meet the NBS chief physicist, Edward Bennett Rosa when she visited. Rosa died at his desk at NBS on Tuesday May 17th, 1921, just three days before the events in Washington. As the NBS chief physicist he was clearly planning to play a role in Curie's visit to NBS. Although Rosa left the technical management of the radium work to Dorsey and Wadleigh he was engaged in high-level metrology matters such as agreements with other national metrology institutes. Rutherford had written to him only a month before about the discrepancy in measurements with the Curie laboratory. But the reply to

Rutherford on May 18th came from Director Stratton who must have been distraught with the death of his friend and colleague the previous day. During Curie's visit to Washington, Drs. Burgess and Vinal were accompanying Rosa's body to a funeral service in Wellsville, New York (Wellsville, 1923; Coblentz, 1934; NIST archives, 2023).

5.6. Postscript to Marie Curie's visit

There are a few other important pieces of correspondence in the NBS archives from 1921 that were not directly related to Curie's visit but were possibly tangentially related. These three are listed here and described in the following pages. First in January, Dorsey made a proposal that the community consider a new unit called the *rutherford* (rd). Second, on May 28, 1921, Samuel Lind sent a letter from the USBM to his colleague in Washington inquiring about the status of the two radium sources that USBM had previously supplied to NBS. And third, on July 14, 1921, the Director of NBS wrote to the Director of the National Physical Laboratory, Sir Joseph Petavel, regarding intercomparisons of radium sources between NBS and the NPL.

5.6.1. Proposal of the rutherford unit of radioactivity

Dorsey and others at NBS had met Rutherford several times over the past 15 years and were not satisfied with the *curie* unit of radioactivity. Dorsey proposed by letter to Boltwood and in a submission to the journal Science that the *rutherford* (rd) be an accepted unit of activity. The exchange of letters with Boltwood is given here.

Typed letter of January 12, 1921 from Dorsey

Radium
Luminous Materials
Problems Pertaining to
Medical Arts and Sciences

Telephone North 3215

Consultant to
National Bureau of Standard

N. Ernest Dorsey, Ph. D.
Consulting Physicist
1519 Connecticut Avenue
Suite 4
Washington, D.C.

January 12, 1921

Prof. B. B. Boltwood,
Yale University
New Haven, Conn.

Dear Prof. Boltwood:-

Will you be so kind as to look over the enclosed note and offer suggestions and criticisms. Am expecting to take the matter up with Dr. Stratton tomorrow, and hope to have it published in some form as a contribution from the Bureau, unless it appears to you that there is some good reason for not doing so.

I have just heard of Bumstead's death on the first; the news was a great shock as I had seen him apparently well just a short time before Christmas. I have heard none of the details. Please give my regards to Uhler and other friends at Yale.

Very sincerely,

N. Ernest Dorsey

This is the first recorded correspondence between Dorsey and Boltwood in nearly six years. Perhaps there are other letters that are not included in the Yale archives. The two must have met several times at APS and Academy of Sciences meetings during that period. This letter of 1921 is written on Dorsey's stationery for his consulting business. He took a leave of absence from NBS starting July 1, 1920; but he said at the time that he would serve as a consultant to NBS. From his letterhead it seems he was consulting clients in radiological physics (for device calibrations) and industry (for the luminous materials). But he maintained his interest in national and international standards. His "enclosed note" was a draft in which he proposed the "Rutherford" as a unit of measurement of radioactivity. Dorsey doesn't mention if he cleared this note with Rosa. He did meet with Director Stratton who permitted him to submit the note to **Science**, with his affiliation given as NBS (Dorsey, 1921b).

Dorsey extends his condolences on the death at age 50 of Dr. Henry Bumstead the late chair of the Physics Department at Yale. Bumstead was about the same age as Dorsey and had done a sabbatical with J.J. Thompson at the Cavendish Laboratory in Cambridge in 1904.

Boltwood replied to Dorsey by letter on January 20,1921.

January 20, 1921

Dr. N. Ernest Dorsey,
1519 Connecticut Avenue,
Washington, D.C.

My dear Dorsey:-

Your letter of January 12th with the enclosed paper on "A Unit of Radioactivity" was duly received. I am sorry that I have not had an earlier opportunity to answer it.

I am frankly interested in your proposal of the "rutherford" and I am not at all sure that I approve of it. It involves a long story but the brief outline is this:- at the last Radiological Congress, which was held in Brussels in 1910, I was appointed the U.S. member of an International Committee on Radioactive Standards. It was this Committee which arranged for and secured, through Marie Curie, the present International Radium Standard. At our meetings an attempt was made to establish a practical standard for working purposes and after a session that lasted nearly all night we decided to adopt the curie "as the quantity of radium emanation in equilibrium with 10^{-8} grams of radium". Marie Curie was a member of the committee and agreed in this decision BUT – at an unearthly early hour the next morning, she arrived at the hotel where Rutherford and I were stopping and informed us that after thinking the matter over she felt that the use of the name "curie" for so infinitesimal small

quantity of anything was altogether inappropriate, being (or let us perhaps say merely as she was) a woman, arguments at the eleventh hour and fifty-ninth minute were of no avail and in order to get the matter before the next session of the Congress, we compromised by letting her have her own way and adopted an experimental standard which was 10^8 times larger than anyone wanted; result – foreseen and duly anticipated – the "curie" of emanation is now hardly ever mentioned and instead we have constant references to micromillicuries.

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At the time of the Congress it was the informally expressed opinion of the members of the Standards Committee, other than the Madam, that the term "rutherford" should be reserved and at some later time adopted as the name for a unit representing the amount of any radioactive product in equilibrium with one gram of either the parent elements, uranium or thorium.

So much by way of explanation. My objection to your proposal of the "rutherford" is therefore based on the fact that it seems to me to be millions of times too large a unit for all ordinary purposes. The disturbances due to the war have postponed indefinitely any further meetings of the Congress on Radiology and there have been no meetings of the International Committee on Standards since 1910. I do not think however that any of the more recent developments in radioactivity have altered appreciably the situation that existed in 1910 and I do not think that the members of the Standards Committee would at all favor the adoption of the unit you have suggested. Also I think that you appreciate that a suggestion which would not meet with general approval would rather hinder than help things. I think that if you reduce your unit to one thousandth of the present suggested value, namely, to the basis of one milligram of radium, it might fulfill a useful purpose and would certainly be more generally desirable than a unit a thousand times as large.

Bumstead's death was a great shock to all of us. He died peacefully in his berth at night in a sleeping car between Chicago and Washington. Vernon Kellogg was with him which was a very fortunate circumstance. We are only just beginning to appreciate the magnitude of the loss and it will be a long time before we are able to fully realize it.

With regards and best wishes,
Yours sincerely,

BBB/C

This is an important letter for the history of radioactivity standards in the US. Boltwood points out that he was the US representative to the International Committee on Radioactive Standards, and that they had only met once in 1910. The official name for the organization was the *Commission Internationale des étalons de Radium*. Officially it was a commission focused on radium. Perhaps the "committee on radioactive standards" transitioned into a "commission on radium standards". But it may be that Boltwood didn't think this title was particularly important since they did not meet again after the war. (He had missed the 1912 meeting in Paris to compare the Paris and Vienna standards.)

His statement on the ideas that they had considered in 1910 for the "rutherford unit" are baffling. How could a standard be the amount of a radioactive product in equilibrium with a gram of uranium or thorium? His suggestion doesn't seem promising as a standard unit. At least the mass of radium was something they could all agree on. He did not totally discourage Dorsey on the suggestion of the "rutherford" unit but suggests a possible improvement.

The reference to Vernon Kellogg who was with Bumstead when he died on the train trip is to the Stanford University entomologist who was soon to become the first permanent secretary of the US National Research Council. He must have been travelling with Professor Bumstead from a meeting in Chicago back to Washington. After the death of Professor Bumstead, Boltwood became increasingly involved in administrative duties on the Yale faculty including the chores of building a new chemistry building on the Yale campus.

Dorsey replied to Boltwood by letter on January 25, 1921.

Three-page typed letter of January 25, 1921 from Dorsey

Radium
Luminous Materials

Problems pertaining to
Medical Arts and Sciences

Telephone North 3215

Consultant to
National Bureau of Standard

N. Ernest Dorsey, Ph. D.
Consulting Physicist
1519 Connecticut Avenue
Suite 4
Washington, D.C.

January 25, 1921

Prof. B. B. Boltwood,
Yale University
New Haven, Conn.

Dear Prof. Boltwood:-

I greatly appreciate your letter of the twentieth instant in relation to my suggestion regarding the rutherford, and am glad to note that there occurs to you but two objections to the suggestion: the size of the unit, and the fact that the name proposed had been informally considered by the International Committee, but in another connection. May I be allowed to state somewhat more fully my views of the subject, having in regard your suggestions?

As regards the size of the unit, there are two distinct problems: 1. The fixing of the fundamental unit; and 2. The fixing of one or more subsidiary units designed to facilitate the expression of numerical results. The first unit forms the integral part of a system of mensuration embracing the entire field of closely related measurements. It should be fixed in such a manner as to be consistent with related existing units, to articulate perfectly with them, and to lead to a homogeneous and simple system of nomenclature. The second, the subsidiary units, should in the decimal system be integral decimal multiples (or submultiples) of the

fundamental unit. If the units are given special names, names other than those formed from the name of the fundamental unit in the manner characteristic of the metric system, then such names, -- e.g., micron, -- however convenient they may be, are in reality a species of scientific jargon, or slang, and are a blot upon the system. The use of such words, like the use of slang, often results in great economy, and, in my opinion both are fully justified in communications between those belonging to the guild to which the jargon pertains, but in communications intended for others the unit properly derived from the fundamental unit should be used, or the significance of the special term should be specifically explained.

The unit I had in mind is a fundamental unit; a unit that is not even limited to a single family of radio-elements, but is applicable to all. Hence it should be coordinate with the fundamental units already internationally established in the field of radioactivity, should articulate perfectly

-2-

with them, and should form with them a homogeneous and simple system of nomenclature. These conditions are fulfilled by the rutherford as defined by me in the note I sent you. The rutherford is strictly coordinate with the gram of radium, and with the curie, the two internationally recognized units. Likewise the milligram, the millicurie, and the millirutherord are strictly coordinate. The gram, the curie, and the rutherford form a consistent, closely articulated, homogeneous, and simply related system of units.

The size of the rutherford, and also of the curie, was actually fixed by the International Committee when they accepted the gram as the unit in which to measure radium. The only thing left open was the name.

Actually a gram of radium nowadays is not such a very large amount, a single contract may call for over 2 grams, the Bureau measured over 25 grams during the fiscal year ending in June last, several tenths of a curie are pumped off daily in each of several institutions, the Bureau in one shipment received for measurement over a half a curie of tubed emanation, the Memorial Hospital reports the use of active deposit applicators containing the deposit from 0.1 to 0.2 curie. For these reasons I cannot regard as inordinately large a unit that coordinates with the gram.

In experimental research into radioactive materials, in the study of rocks, and ores, and to 10^{-9} curie (a millimicrocurie) is required, and it may be desirable to give a special name to the fact that the International Committee informally considered reserving the name rutherford for another purpose makes one hesitate to propose this name for the unit I suggested. But a more careful consideration removes this hesitancy. The use of this name, as informally suggested by the International Committee, to designate the amount of a radio-element that is in equilibrium with a gram of uranium (or of thorium, as the case may be) relegates it to a strictly subsidiary unit that is limited to one family of elements, and that is out of harmony with the fundamental units already adopted. (If it is applied to the uranium family it will be related to a gram of radium by an experimentally determined factor of about 3.23×10^{-7}.) If the name is used as I suggested it will denote one of the most fundamental units in radioactivity, a unit that

-3-

is applicable to all radio-elements independent of their relation one to another, a unit that is in exact harmony with the Internationally accepted units. Of the two proposals mine conveys the more honor and consequently should be given precedence.

To my mind the main objection to using the name rutherford is its length; it is in many ways desirable that names of the fundamental units be words of one or two syllables,

Very sincerely,

N. Ernest Dorsey

This is the last of the eighteen letters between Dorsey and Boltwood over an eight-year period. There is no indication that Boltwood ever responded. Their roles in the US measurement system had evolved. Dorsey was clearly suggesting that NBS was invested in establishing quantities and units for radioactivity measurements that would be consistent with the other units in the national measurement system of 1921. Boltwood had little time to argue the point as he was burdened with his duties as chair of the chemistry department and the plans for new chemistry laboratories at Yale. Since Dorsey had the support of NBS Director Stratton he did go ahead with his publication in **Science** promoting the rutherford unit (Dorsey, 1921). Although the *rutherford* (rf) was not internationally accepted, it was often used in reports from NBS (e.g., in Figure 13) (NIST archives, 2023). One of the radium artifacts at the Smithsonian Institution is certified in units of *rutherfords*.

It is interesting to look at the letter in the context of what was happening in 1920 -1921. Dorsey had taken a leave of absence from July 1, 1920. His letterhead and the article in **Science** both list his continuing affiliation with NBS. He listed specifics on NBS radium calibrations in the past fiscal year when he was chief of Section 13. Dorsey indicates here that sources containing hundreds of millicuries were often calibrated at NBS by comparison with the appropriate standard radium sources.

Finally, it is amusing that Dorsey refers to a small amount of radioactivity as a millimicrocurie which would now be referred to as a nanocurie or 37 *becquerel*. The international system of prefixes for subunits was not in use in 1921.

5.6.2. A question from the US Bureau of Mines

Only one week after Marie Curie's visit to NBS the chemist Samuel Lind from the USBM wrote to Dr. L.I. Shaw at the USBM in Washington with some questions about the two radium sources that USBM had provided to NBS in 1916. It is the author's suggestion that these sources were no longer needed at the USBM after completion of the contract with the National Radium Institute. The agreed upon 7 grams of radium had been delivered to Baltimore and New York. Lind's letter of May 28th is given here.

DEPARTMENT OF THE INTERIOR

Bureau of Mines
Rare and Precious Metals Experiment
Station

Reno, Nevada.
May 28, 1921.

Dr. L.I. Shaw,
U.S. Bureau of Mines,
Washington, D.C.

Subject: Radium Tube W_4.

Dear Doctor Shaw:

Replying to your letter on this subject.

Radium tube W_4, delivered to the Bureau of Standards in 1916, contains 0.1372 gram of anhydrous radium bromide. Their first measurement reported 48.7 milligrams of radium element. I am puzzled at its abnormal rate of decrease of activity. Has Dr. Wadleigh examined it for a crack or possible leak of emanation around the insealed platinum wire, which might amount for such loss? In case he is interested in the original data on the other tube T_3, delivered to them in August, 1916, it contains 0.2861 gram of anhydrous radium bromide, and was determined by them to contain originally 91.8 milligrams of radium element. One of these tubes did at one time develop a crack and was sealed by Dr. Dorsey inside another tube, but I do not know which one of them it was. I am inclined to think, however, it T_3.

Very truly yours,

(Signed) S.C. Lind.

The timing of Lind's letter to USBM in Washington is interesting. Since he posted the letter from Nevada on May 28, it seems that he was not able to attend the ceremony in Washington the previous Friday. Had he been in Washington he would have had an opportunity to discuss this matter directly with NBS. The masses he gives for the anhydrous radium bromide do not accord with the NBS assays of the radium element (if one uses 58.6 % as the mass of radium in anhydrous $RaBr_2$). The USBM sources may have contained residual barium ($BaBr_2$) from incomplete separation in the final crystallization step. Lind may have been questioning how Wadleigh at NBS carried out the standardization of the 100 mg ampoules that made up the Curie gram source. He did not direct these questions to Wadleigh, but instead referred to previous correspondence with Dorsey. The fate of the two large sources from the USBM is not found in the NIST archives. The international standard from 1913 and the two from 1934 are the only radium salts in glass ampoules still at NIST in 2023.

5.6.3. Proposal for International Comparisons

It is possible that Marie Curie's visit to NBS and to the Standard Chemical Company prompted the first steps towards formal establishment of radionuclide metrology. The Director of NBS wrote to the Director of the National Physical Laboratory with a rather detailed proposal to exchange calibrated radium sources. His letter of July 14, 1921, came two months after Curie's visit, prompted by a letter a month earlier from the NPL enquiring about calibrated materials from Pittsburgh. His letter is given here in full.

July 14, 1921

Sir Joseph E. Petavel, Director
National Physical Laboratory
Teddington, Middlesex,
England

Subject: Radium measurements in different countries

Dear Sir Joseph:

We have your letter of June 13, (your reference GWCK/MKP), regarding radium measurements, and the enclosed tabulation of results of measurements made from 1914 to 1916 and in April, 1921, on various preparations of the Radium Chemical Company. We have examined our records but unfortunately find no measurements made on any of the preparations which are included in your list. We appreciate highly the trouble you have taken to compile this information. And we shall be very glad to follow your suggestion that a comparison of a sample of radium salt be undertaken by the two laboratories.

We have in course of preparation several tubes containing approximately 15 and 25 milligrams of radium element, and after we have had opportunity to measure these we can probably arrange to send one of each size to you for comparison. When this is done it may be worth while also to send the same tubes to the Curie Laboratory and possibly others in order to have a direct check by measurements on the same preparations in all laboratories. At present we cannot say just when these tubes will be ready, but when they are we will communicate further with you.

Very Sincerely yours,
Director

Signed for the Director
F.C. Brown

The letter was signed by NBS Assistant Director F.C. Brown. The NIST archives do not include the incoming letter from NPL, but it is interesting to note that the incoming letter references GWCK at the NPL, who was probably G.W.C. Kaye the NPL physicist referenced previously (page 52). It is also noted that the NPL list of sources from the Radium Chemical Company included some from 1914, probably the same sources in the 4 gram lot that Glenn Kammer had trouble with during his trip in 1914 to deliver material to UK hospitals. The archives do not show whether the NBS radium sources were sent to NPL (or other laboratories), but such a comparison would have been of great value in demonstrating agreement between such important measurement institutes. The data shown in Figure 6 on the sources exchanged between these same laboratories suggest results should have agreed, at least to within a few percent. Formal comparisons of radium sources did not begin in earnest until those organized by Wilfrid Mann at NBS in the 1950s (Davenport et al., 1954; Loftus et al., 1957).

This letter of July 1921 concludes the main correspondence from NBS staff regarding radium and x-ray measurements and standards during the period 1901-1921.

6. Epilogue

After 1921 NBS contributions to radiation metrology slowed considerably for several years. With Rosa's death in 1921, and Stratton's move to the presidency of the Massachusetts Institute of Technology in 1923, the work associated with radiation moved to a new group, Atomic Physics, Radium and X Ray in the Optics Division. Fortunately, for the study of the history of NBS, the institute published the Journal of Research beginning in 1928 (https://www.nist.gov/nist-research-library/journal-research-nist/about-journal). The NBS researchers in the following decades published in that journal often providing more details than in papers they submitted to **Science** and other journals in physics. The next major stages in metrology for radioactivity and x rays came after 1926 with the arrival of two physicists, both with doctorates from Cornell University. Leon Francis Curtiss (1895 – 1983) and Lauriston Sale Taylor (1902 – 2004) were to become the leading voices for radiation metrology for the US for the next four decades. In 1951, Taylor recruited Wilfrid Basil Mann (1908 – 2001) to replace Curtiss as Chief of the Radioactivity Section. Perhaps the two most significant accounts of radionuclide metrology and x-ray metrology in the mid 20th century are the 1985 edition of NCRP Report 58, "A Handbook of Radioactivity Measurements" (NCRP, 1985), and Taylor's NBS Special Publication 625, "X-Ray Measurements and Protection: 1913 -1964" (NBS, 1981).

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