

Rediscovery of the Elements

The Rare Earths—The Confusing Years

A gallery of rare earth scientists and a timeline of their research



Figure 1. Important scientists dealing with rare earths through the nineteenth century. Johan Gadolin (1760–1852)^{1a}—discovered yttrium (1794). Jöns Jacob Berzelius (1779–1848) and Martin Heinrich Klaproth (1743–1817)^{1d}—discovered cerium (1803). Carl Gustaf Mosander (1787–1858)^{1p}—discovered lanthanum (1839), didymium (1840), terbium, and erbium (1843). Jean-Charles deGalissard Marignac (1817–1894)^{1o}—discovered ytterbium (1878) and gadolinium (1880). Per Teodor Cleve (1840–1905)¹ⁿ—discovered holmium and thulium (1879). Lars Fredrik Nilson (1840–1899)^{1m}—discovered scandium (1879). Paul-Émile Lecoq de Boisbaudran (1838–1912)—discovered samarium (1879) and dysprosium (1886).^{1b} Carl Auer von Welsbach (1858–1929)^{1c}—discovered praseodymium and neodymium (1885); co-discovered lutetium (1907). Eugène-Anatole Demarçay (1852–1903)—discovered europium (1901).^{1e} William Crookes (1832–1919)^{1m}—spectral techniques; proposed “meta-elements.” Marc Delafontaine (1837–1911)—co-discovered holmium (1879). Charles James (1880–1928)^{1k}—co-discovered lutetium (1907). Georges Urbain (1872–1938)^{1k}—co-discovered lutetium (1907). Bohuslav Brauner (1855–1935)—predicted element 61.



James L. Marshall, Beta Eta 1971, and Virginia R. Marshall, Beta Eta 2003, Department of Chemistry, University of North Texas, Denton, TX 76203-5070, jimm@unt.edu

The rare earths after Mosander. In the previous *HEXAGON* “Rediscovery” article,^{1p} we were introduced to the 17 rare earths, found in the f-block and the Group III chemical family of the Periodic Table. Because of a common valence electron configuration, the rare earths have similar chemical properties, and their chemical separation from one another can be difficult. From preparations of the first two rare earth elements—yttrium and cerium—the Swedish chemist Carl Gustaf Mosander (Figure 1, 2) was able to separate four additional elements during 1839–1842: lanthanum, didymium, erbium, and terbium.^{1p}

Mosander’s discoveries signaled the possibility of yet more elements hidden in the parent yttrium and cerium. However, it was three and a half decades before the next rare earth was isolated. Mosander’s successes had been relatively “easy pickings,” because abundant lanthanum (see Table 1) could be quickly and quantitatively separated as the soluble trivalent salt (La⁺³) from insoluble tetravalent cerium (Ce⁺⁴); and the vivid colors of didymium (amethyst), erbium (orange), and terbium (rose) allowed visual tracking of their separation during repeated recrystallizations. By contrast, the remaining rare earth oxides all generally exhibit

Table 1: Crustal abundances of rare earths, ppm [ref 2]

Sc	Y	La	Ce	Pr*	Nd*	Pm	Sm	Eu	Gd	Tb**	Dy	Ho	Er**	Tm	Yb	Lu
22	33	30	60	8.2	28	trace	6	1.2	5.4	0.9	3.0	1.2	2.2	0.5	3.0	0.5

*Mosander’s didymium was a mixture of Pr and Nd.

**Tb and Er are now reversed from the original assignments of Mosander.

the same valence state (+3), are colorless, and exist in low concentrations in nature.

The advent of spectroscopy. During 1860–1861 Robert Wilhelm Bunsen (1811–1899) and Gustav Robert Kirchhoff (1824–1887) of the University of Heidelberg discovered the elements cesium and rubidium (1860–1861) in Dürkheim Spa mineral waters with their newly invented emission spectroscopy.^{1b} This spectral tool was immediately adopted by William Crookes^{1m} in London to discover thallium (1861) from Harz Mountain mines of Germany; and then by Hieronymus Theodor Richter (1824–1898) and Ferdinand Reich (1799–1882) of the Freiberg Mining School in Saxony to discover indium (1863) from the neighboring Himmelfürst Mine.^{1a,h} Spectral analysis was quickly recognized as a possible solution to untangle the confusing rare earth mixtures. With the simultaneous development of more sophisticated chemical separation techniques, rare earth research experienced a surge during the last quarter of the nineteenth century.

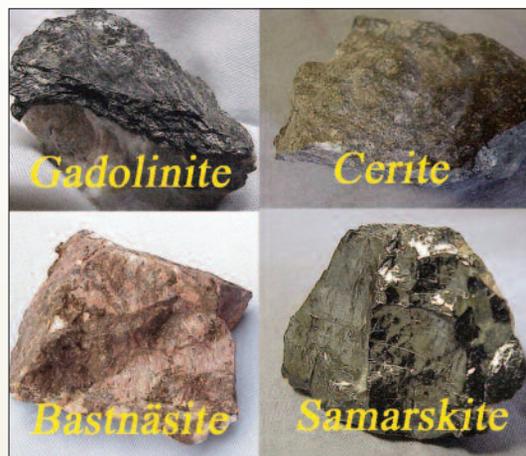
Spectral analysis—the situation becomes complicated.

One might expect that the new method of emission spectroscopy might prompt immediate discoveries of rare earths. Ironically, spectral analysis initially complicated the situation, because thousands of spectral lines now had to be separated and identified. These lines arose not only from mixtures of rare earths themselves, but also from many other elemental impurities such as iron, strontium, barium, etc., with which rare earths were chemically bound in the original minerals. Further complicating the situation, different source minerals would have different relative compositions of the rare earths (Figure 3), rendering corroboration difficult by others—it was even possible that a mineral specimen would be lacking one of the rare earths which would be present in abundance in a sample from a different geological site. Furthermore, different researchers would use different recrystallization conditions (concentration, temperature, etc.), resulting in subtle composition differences and different spectral appearances for the “same” element preparations. Announcements soon appeared of philipium, decipium, mosandrium, rogerium, glaucodymium, russium,^{3a} carolinium, berzelium,^{3c} celtium,^{3c} denebium, dubhium, welsium,^{3d} terbium-II and terbium-III, neoholmium and neo-erbium,^{3d} thulium-II and thulium-III,^{3c} etc., etc. Known rare earths with confusing manifold spectra led hasty chemists, eager to win the glory of new discoveries, to announce even more complex mixtures, e.g., X α , X β , X γ , X δ , X ϵ , X ζ , X η , in “element X” from



Figure 2. On the left is the building housing the Royal Swedish Academy of Sciences (second site), Wallingatan 2 (N59° 20.26 E18° 03.52), where Mosander lived and worked. Here he prepared lanthanum, didymium, erbium, and terbium.^{1p} The building is now used for general offices. The church at the end of the street is Adolf Fredriks Kyrka [Church], inaugurated in 1774. The view looked the same in Mosander's time. “Father Moses,” as Mosander was affectionately called by his friends, proudly told Berzelius (co-discoverer of cerium) that in this building was prepared the only pure sample of ceric oxide in the world — “white, slightly yellowish” (1842).⁴

Figure 3. Historically important rare earth minerals. Gadolinite, (Y,RE)₂FeBeSi₂O₁₀, from the Ytterby Mine, Sweden; source of the first rare earth discovered, yttrium, and named for the element's discoverer, Johan Gadolin; analyzes (%) for Y/Tb/Tb/Dy/Tm/Yb 16/2/2/2/5/3. Cerite, (Ce,RE,Ca)₁₀Fe(SiO₄)₆(SiO₃OH)(OH)₃, from the Bastnäs Mine; source of the first lanthanide; by Klaproth, Hisinger, and Klaproth, analyzes (%) for La/Ce/Pr/Nd/Sm 12/26/3/11/2. Bastnäsite, (RE)CO₃F, the main source of rare earths both in the U.S. and China. This sample from Mountain Pass, CA, analyzes (%) for La/Ce/Pr/Nd/Sm 33/49/4/12/1. Samarskite (RE,U,Th,Fe)(Nb,Ta,Ti)₅O₁₆, resembling “black obsidian,” has wildly variable compositions; this very radioactive sample from Jefferson County, CO, analyzes for (%) U/Th/Dy/Er/Yb/Lu 10/10/3/3/6/1.



All mineral photographs in these figures are taken of specimens in the private collection of the authors; all specimens possessed lesser (<1%) quantities of the remaining rare earths which are not listed.

samarskite (Figure 3); Er α and Er β in erbium; Tm α and Tm β in thulium; Sm α and Sm β in samarium; and Di α , Di β , Di γ , Di δ , Di ϵ , Di ζ , Di η , Di θ , and Di ι in didymium!^{13a} In fact, during the half century after Bunsen and Kirchhoff's

invention of the spectroscope, no less than ninety-four (94) spurious claims were made for new rare earths!¹³

Sometimes, in the hands of two independent researchers, different but “similar” ele-



Figure 4. Two unusual minerals illustrating that rare earth distributions can vary widely in nature. Left: Kuliokite (purple crystal), $(Y,RE)_4Al(SiO_4)_2(OH)_2F_5$, from the Kuliok River, Kola peninsula, Russia. The mineral has unusually high amounts of the “rarer” rare earths—(%) Y/Gd/Dy/Ho/Er/Tm/Yb/Lu 56/0.4/1.0/0.2/2.0/0.1/3.0/0.1. Right: Schuilingite (bluish coat), $PbCu(Nd,RE)(CO_3)_3 \cdot 1.5H_2O$, from Kasompi, Shaba, Zaire, a copper-rich district in Africa. The concentration of europium is incredibly high (europium is the red phosphor in color television screens)—(%) Y/Nd/Sm/Eu/Gd 14/12/8/7/13. Obviously, the mineral did not form by the ordinary volcanic or ion-adsorption geological mechanisms¹⁹ but instead by an unusual secondary hydrothermal process.



Figure 5. Monazite, $(RE,Th)PO_4$, was used extensively by Welsbach to develop his rare earth enterprises. He utilized the newly discovered monazite sands of Brazil; this crystalline sample from Minas Gerais, Brazil, illustrates the appearance before erosion to sand. “Misch metal,” a crude mixture of the lighter rare earths (with admixed iron to impart hardness), is pyrophoric and serves as the “flint” in cigarette lighters. The rare earth composition (%) of this sample is La/Ce/Pr/Nd/Sm 28/52/4/13/2.

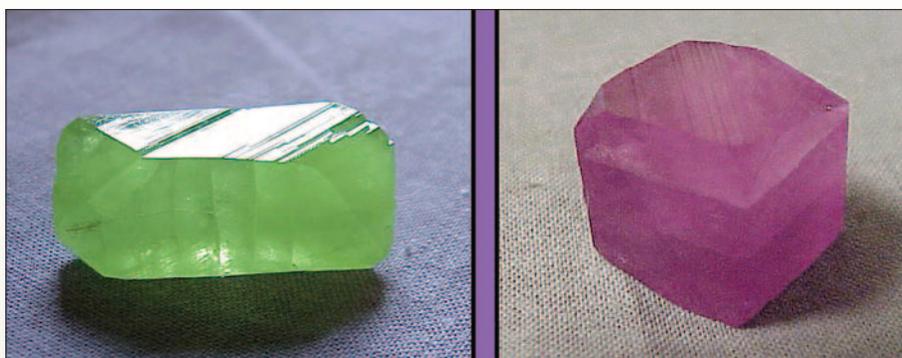


Figure 6. Crystals of praseodymium (green) and neodymium (magenta) sulfates. Neodymium has often been in the news lately, being an important ingredient in small permanent magnets which are critical in modern electronic devices. These mixed elements were considered a single element, called “didymium” by Mosander, before they were separated by Welsbach half a century later. Separating the two photographs in this figure is an amethyst-colored separator, the color of the didymium crystals collected by Mosander.¹⁹ Didymium safety glasses, with this same amethyst hue, are still being used by glassblowers and jewelers to filter out the blinding yellow glare of molten glass.

ments were recognized as being identical. An example was holmium,⁴ which Delafontaine observed spectroscopically^{5a} before Cleve isolated it.⁶

Although Delafontaine is not generally recognized as the primary discoverer of a rare earth, his imaginative analytical schemes were recognized as important in clarifying the situation. Before it was realized that the rare earth composition of different mineral specimens could be wildly diverse (Figure 4), there was a dispute regarding the nature, if not the existence itself, of Mosander’s original erbium and terbium. Delafontaine produced definitive work that confirmed the existence of both, and to avoid confusion in the literature, he suggested (1877) a formal reversal of the original assignments of erbium and terbium by Mosander, which the scientific public accepted.^{5a}

Advanced separation techniques. Realizing that the classical recrystallization techniques were inadequate for the job of separating the rare earths, investigators sought more advanced solvent/salt systems. One technique that proved to be particularly successful was the double salt method, whose first major success was the separation of the components of Mosander’s didymium. It had been suspected that didymium was actually two elements, because samples of didymium from different geological sites might exhibit the same spectral lines, but with different intensity patterns—the prime example of this was seen with samarskite, notorious for its variable composition (Figure 3), studied by Delafontaine and Boisbaudran.^{5a} The person to resolve this question was Welsbach, the inventor of the gas mantle and the cigarette lighter.^{1c}

Welsbach procured monazite (Figure 5), originally used as a ballast for sailing ships, to obtain thorium. He impregnated lantern mantles with thorium oxide which glowed bright white when heated by a flame (Coleman gas lantern mantles purchased in the U.S. 15 years ago used thorium oxide, but now use yttrium oxide).

The remaining rare earths he called “Misch metal,” a pyrophoric mixture for flints in cigarette lighters (which he also invented) (Figure 5). From Misch metal he isolated didymium and in 1885 was successful in separating its components—two elements which he called praseodymium (“green twin”) and neodymium (“new twin”) (Figure 6). For his solvent system, he used ammonium double nitrates $(RE)(NO_3)_3 \bullet 2NH_4NO_3$ in concentrated nitric acid^{5b} (“RE” = rare earth element).

Other examples of new efficient separation schemes included those of Urbain, who used a

TOP RIGHT: Figure 7. Crookes moved to this house in 1880 at 7 Kensington Park Gardens in Notting Hill (N51° 30.69 W00° 12.16.) where he spent the remainder of his life. Crookes was never formally associated with any university; instead, he worked in his home and at the editor's office of his *The Chemical News*.

Figure 8. The laboratory inside Crookes home. It was here where terrestrial helium was first spectroscopically verified.¹⁰ At the end of the laboratory is his spiral Periodic Table, prepared to show his theory of an electrical oscillation phenomenon that created the elements out of a primordial material. Crookes' imagination took him to many places, including spiritualism, which he seriously explored for a large portion of his life.

combination of double magnesium nitrates (RE)(NO₃)₃ • 3Mg(NO₃)₂ and isomorphous bismuth nitrates. In this elaborate procedure, he separated the rare earth mixture into light and heavy fractions. Bismuth nitrate was extremely useful here, as its solubility lay between those of samarium and europium. The bismuth could be removed with hydrogen sulfide, and then the remaining two fractions could be further separated and purified by employing double magnesium nitrate salts. By this method, Urbain was able to refute many of the spurious rare earth discoveries of others.⁷

Charles James, professor at the University of New Hampshire and member of AXΣ's *Mu* Chapter,⁸ is credited⁴ with developing the best and most efficient overall scheme for separating the rare earths. In his complex flow chart, he used a large variety of rare earth salts, including oxalates, bromates, sulfates, ethylsulfates, and double nitrate salts of the rare earth and magnesium, ammonium, sodium, bismuth, and nickel—each specific step carefully chosen after trying out all possible systems. Even with his grand achievement, he admitted that it was not perfect, warning the reader that there is simply "no quantitative method of separation of any of the rare earths."⁸

Crookes, phosphorescence spectroscopy, and the meta-elements—anticipating the concept of isotopes? The complexity of the rare earths and confusion in their separation could engender unusual ideas. The classic example was the hypothesis of "meta-elements," spawned in the imaginative mind of William Crookes during his spectroscopic study of yttrium.

Crookes is perhaps best known for his invention of the "Crookes tube" in 1875, the original cathode ray tube which eventually led to the discovery of electrons in 1897 by Joseph John Thomson (1856–1940). Crookes also



invented the radiometer, a popular scientific toy to this day. Another creation of his was the spintharoscope, which was popularized in the 1940s as the “Atomic Bomb Ring” displayed proudly by schoolboys who ordered them from breakfast cereal box advertisements.^{1m}

Crookes split his activities between his home laboratory in east London (Figures 7–9) and the offices of *The Chemical News*. He founded and served as the editor of this “bumptious, gossipy”⁹ journal in which he recounted the daily chemical discoveries (including his own, of course) and other happenings in the industrial world and the professional societies. His research was well respected, and he was elected a member of the Royal Society, of which he was president 1913–1915.

Another invention of Crookes was “phosphorescence spectroscopy,” achieved by irradiating samples in his Crookes tube.¹⁰ These spectra were more complex than the usual emission spectra, and he was able to detect variations with samples which otherwise showed identical spectra in ordinary emission spectroscopy.⁷ He announced that “there were probably eight constituents into which yttrium might be split,”¹¹ and he reported similar behavior with samarium and gadolinium. Thus, he claimed, he had evidence for chemically identical elements which differed in their physical properties. He called these “meta-elements.” And what would cause a difference in physical properties? Obviously, their atomic weights, he concluded.

This idea of meta-elements held appeal for some, particularly those in Great Britain, since the idea paralleled Prout’s hypothesis (William Prout, 1785–1850) that all elements were built up of multiples of hydrogen.⁷ Perhaps even a better candidate for the universal simple substance—called *protyle*⁷—might be helium, first spectroscopically observed in the sun in 1868,¹ⁿ whose single spectral line (only one was known at that time) signaled its simple nature. By contrast, heavier elements, such as iron, exhibited a complex spectrum, consistent with a wide distribution of meta-elements.⁷ Even some organic chemists weighed in with their endorsement; Jean-Baptist Andre Dumas (1800–1884)^{1f} suggested that elements might be unusually stable radicals, just as an organic molecule is built up of smaller organic radicals.⁷

Crookes expanded his hypothesis to include genesis of the elements themselves: a cooling process of the *protyle*⁷ in stars—he proposed the elements condensed into a statistical distribution of weights but with identical chemical properties. Thus, while the measured atomic mass of calcium was 40, actually there might be some 39, 38, and 41, 42—or perhaps 39.9, 39.8 and 40.1, 40.2, and so on.¹²



Figure 9. The chemical portion of Crookes’ laboratory. Crookes applied his chemical and spectroscopic skills to the study of the rare earths. After radioactivity was discovered in 1896, he studied the radioactive elements.

Others had doubts as to the usefulness of the phosphorescence spectroscopy method. Boisbaudran^{1b} stated that variations would likely to be simply due to impurities; and in fact he reported that in an ultra pure sample of yttrium, he could not observe a phosphorescence spectrum at all, but instead the usual emission spectrum.⁷ Marignac^{1o} argued that since there was no experiment to confirm the authenticity of meta-elements, the concept was useless.⁷

Clarification of the phosphorescence spectra phenomenon came two decades later. In 1919 Urbain completed a thorough study showing that trace amounts of impurities could drastically alter the phosphorescence spectra (anticipating the use of dopants in modern phosphors). Urbain could duplicate Crookes data by artificially prepared mixtures of the rare earths in the proper ratios.¹³ Urbain also worked out procedures for the fractionation of double salts of the rare earths (RE/Mg) to prepare pure samples of the rare earths—and he extended Boisbaudran’s work for yttrium to report that for all utterly pure samples of the rare earths, the anomalous phosphorescent spectra ceased to exist.¹³

Utterly unfazed by Urbain’s criticisms, Crookes took advantage of his position as president of the Royal Society to announce in his 1914 President Address to the Royal Society¹⁴ that he had anticipated Soddy’s discovery of isotopes¹ⁱ which had just been announced: “[Soddy’s] ‘isotopic’ elements occupy the same place in the Periodic Table. He has thus arrived, by a totally different path from the one I trav-

elled, at the conception of an element having atoms of different weight though chemically identical.”

Although it is frequently mentioned that Crookes “was the first to suggest the existence of isotopes,”⁹ it must be remembered that any similarity between Crookes’ meta-elements (based on a false interpretation of spectral data) and Soddy’s isotopes (securely founded on a wealth of radioactive and transmutation experimentation) is fortuitous—although great credit must be given to Crookes’ spectral skill as well as his vivid imagination. At his Nobel address in 1921—“The Origin of the Conception of Isotopes,”¹⁵ Soddy did not mention Crookes’ work nor his meta-elements, but instead concentrated on the research on radioactive elements that provided the evidence for the concept of isotopes.¹⁶

The last of the natural rare earths. The last naturally occurring rare earth discovered was lutetium, in 1907. In *The HEXAGON* we have previously visited the convoluted story of this discovery.^{1k} Today the discovery of lutetium is credited to three chemists: the opportunistic Urbain who published first (and prematurely), Welsbach who presented evidence that he had discovered the element earlier, and James who had the only pure sample of lutetium existing at that time. Today all three scientists are considered co-discoverers of lutetium.

At the time of the discovery of lutetium, Urbain was claiming evidence of another element beyond lutetium (atomic number 71),

which he called “celtium.”^{1k} However, his pre-conceived notion that the next element would be a rare earth was leading him astray (the next element, atomic number 72, was actually hafnium^{1k}, which was not found in rare earth minerals but instead in zircon^{1k}). Whereas Urbain was claiming he was obtaining purer and purer samples of “celtium” (which he followed by magnetic susceptibility measurements¹⁰), he was actually witnessing purer samples of lutetium, which others had already prepared.

Atomic numbers. By the turn of the century, the major question was: Just how many rare earths *did* exist? Even at this late stage, spurious rare earths were being reported; Crookes was announcing “ionium” and “incognitum” (1906) in his rare earth mixtures;^{3f} Urbain was stubborn with his “celtium” (1907),^{3c} and there was a smattering of other elements from Demarçay (1900; Σ, Γ, Δ, Ω, Θ), Brauner (1900; thorium-α and thorium-β), and Welsbach (1911; thulium I, II, III), as well as a few lesser-knowns.^{3f}

Brauner, professor of chemistry at the University of Prague (today’s Czech Republic), on the basis of the solubility trends of salts of the known rare earths, as well as the relatively large difference in the atomic masses of neodymium and samarium, in 1902 predicted an element between the two.^{3b} Then in 1914 Henry Gwyn Jeffreys Moseley (1887–1915), with his X-ray studies, experimentally developed the concept of atomic numbers.¹ⁱ Moseley showed that celtium was spurious and that a void lay between neodymium (element 60) and samarium (element 62), just as Brauner had predicted. One would surmise that this would quickly clarify the situation, because scientists would know where to look—in crude preparations of neodymium and samarium—but instead it led to more confusion and contention, and element 61 was not found until three decades later. ☉

In the next issue of *The HEXAGON*, the search for element 61.

Acknowledgment.

The silent editor of these “Rediscovery” articles—Gerard R. Dobson, 1990 Kuebler Awardee, who has been invaluable for countless corrections and suggestions over the past 15 years—has in his possession, and on display in his home, one of the large crystallization dishes that Charles James used in his rare earth studies.

“REDISCOVERY” ARTICLES ARE NOW ON-LINE

All *HEXAGON* issues that include “Rediscovery” articles—a series which began in 2000—are now on-line at: <http://digital.library.unt.edu/explore/collections/HEXA/>

These *HEXAGON* issues, as a group, are fully searchable and thus are amenable to scholarly research. One can search either for words, Boolean “OR” combinations, or for full phrases (by placing in quotation marks). Not only the original “Rediscovery” articles may be accessed, but also cover photographs by the authors and other auxiliary articles connected with the “Rediscovery” project.

Additionally, the UNT Digital Library has separated out all these individual articles and placed them in the “Scholarly Works” section. These articles may be located and perused at: <http://digital.library.unt.edu>. At the top of the webpage, search for “James L. Marshall” as “creator” and for convenience, “sort” by “Date Created (Oldest).” The “Scholarly Works” articles are not searchable as a group, but only within each individual article.

References.

- J. L. and V. R. Marshall, *The Hexagon of Alpha Chi Sigma*, (a) **2001**, 92(2), 20–22; (b) **2002**, 93(4), 78–81; (c) **2002**, 93(1), 9–11; (d) **2003**, 94(1), 3–8; (e) **2003**, 94(2), 19–21; (f) **2007**, 98(1), 3–7; (g) **2008**, 99(1), 8–11; (h) **2008**, 99(3), 42–46; (i) **2010**, 101(3), 42–47; (j) **2010**, 101(4), 68–71, 74; (k) **2011**, 102(3), 36–41; (m) **2011**, 102(4), 62–67; (n) **2012**, 103(2), 20–24; (o) **2014**, 104(3), 46–51; (p) **2016**, 106(3), 40–45.
- J. L. and V. R. Marshall, “Walking Tour of the Elements,” in *Rediscovery of the Elements*, JMC Services, Denton, TX, ISBN 978-0-615-30795.
- M. Fontani, M. Costa, M. V. Orna, *The Lost Elements. The Periodic Table’s Shadow Side*, **2015**, Oxford University Press; (a) 119–127; (b) 175; (c) 192; (d) 202–215; (e) 235; (f) 488–490.
- M. E. Weeks and H. M. Leicester, *Discovery of the Elements*, 7th ed., **1968**, 675–684.
- C. H. Evans, ed., *Episodes from the History of the Rare Earth Elements*, Kluwer Academic Publishers, **1996**, (a) F. Szabadvary and C. Evans, “The 50 years following Mosander,” 55–65; (b) E. Baumgartner, “Welsbach,” 113–128.
- Delfontaine’s collaborator was Jacques-Louis Soret (1827–1890) of Geneva, Switzerland, who is also known for his determination of the composition of ozone (ref 5a).
- R. K. DeKosky, “Spectroscopy and the Elements in the Late Nineteenth Century: the Work of Sir William Crookes,” *B. Journ. Hist. Sci.*, **1973**, 6(4), 400–423.
- C. James, *Chem. News*, **1908**, 97, 205–209.
- S. Kean, *The Disappearing Spoon*, **2010**, Little, Brown, and Company, 255–259.
- J. F. Spencer, *The Metals of the Rare Earths*, **1919**, Longmans, Green and Co.
- W. Crookes, Report of the British Association for the Advancement of Science, 1886, (London, 1887).
- W. Crookes, *Chem. News*, **1891**, 63 (6 March), 114.
- G. Urbain, *Comptes rendus*, **1908**, 147, 1472–1474.
- W. Crookes, *Chem. News*, **1914**, 110, (11 Dec), 290; *Proc. Roy. Soc.*, **1915**, 91(625), 106–119.
- F. Soddy, “The Origin of the Concept of Isotopes,” Nobel Lecture, December 12, **1922**, 371–399, available at http://www.nobelprize.org/nobel_prizes/chemistry/laureates/1921/soddy-lecture.html.
- However, Soddy did mention elsewhere that Crookes was the first to conceive of the idea that all atoms of an element might not be identical in all respects: F. Soddy, *J. Chem. Soc., Trans.*, **1919**, 115, 1–26.