ISOTOPES: IDENTIFYING THE BREAKTHROUGH PUBLICATION (1)

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Abstract

Selection of the isotope concept for a Citation for Chemical Breakthrough award in 2013 presented both a dilemma of identifying the most appropriate publication to honor and an opportunity for reflection on the nature of this discovery in particular and of scientific discovery more generally. Several findings in the early years of the twentieth century led Frederick Soddy to introduce the term isotope (a word suggested by classics scholar Margaret Todd) for varieties of the same element that have different atomic masses. The public birthday of the term is well established: it was first published in the December 4, 1913, issue of *Nature* (2, 3). The public debut of the concept, however, is much more difficult to date. Five plausible candidates are reviewed here, from the recognition of distinct but chemically inseparable "radioelements," to the elucidation of the pathways of radioactive decay collectively organized under the laws of radioactive displacement, to the adoption of atomic number rather than atomic weight as the organizing principle of the periodic table. There happens to be no paper in which a proposal of the isotope concept is either the headline or bottom line result.

Introduction

For just over ten years, the ACS Division of the History of Chemistry's Citation for Chemical Breakthrough (CCB) Award has been recognizing publications and patents "that have been revolutionary in concept, broad in scope, and long-term in impact" (4). On several occasions, questions have arisen over just which publication to honor in connection with a well-defined discovery or invention selected for recognition. On such occasions, the non-voting Committee Secretary and general impresario of the CCB Award, Jeffrey Seeman, has engaged consultants to make recommendations on the most appropriate publication to recognize. On the one hand, such ambiguity is not surprising, given the incremental nature of the construction of scientific knowledge. On the other, the exercise of attempting to select "the" breakthrough publication has led to thoughtful considerations and interesting discussions of the development of particular inventions and discoveries and on the nature of scientific discovery more generally, some of which have been published in earlier volumes of this journal (5). Relevant issues have included both internal matters of technical content (identifying which of a series of publications included a crucial advance) and external considerations such as the impact and readership of a publication.

The list of nominations circulated to the 2013 CCB award committee included one said to be the "First proposal of isotopes by Soddy" (6). The paper put forward was a 1911 article on mesothorium (7), one of a plethora of radioelements (8) discovered over the preceding decade and a half. In the supporting information section, the nominator had pulled out the following key quotation (pp 81-82):

> It appears that chemistry has to consider cases, in direct opposition to the principle of the Periodic Law, of complete chemical identity between elements presumably of different atomic weight, and no doubt some profound general law underlies these new relationships.

I was not surprised when Seeman informed me that isotopes had been selected and asked me to look into the matter of identifying the breakthrough paper. The publication I recommended in the end was the *Nature* paper of December 1913 (2).

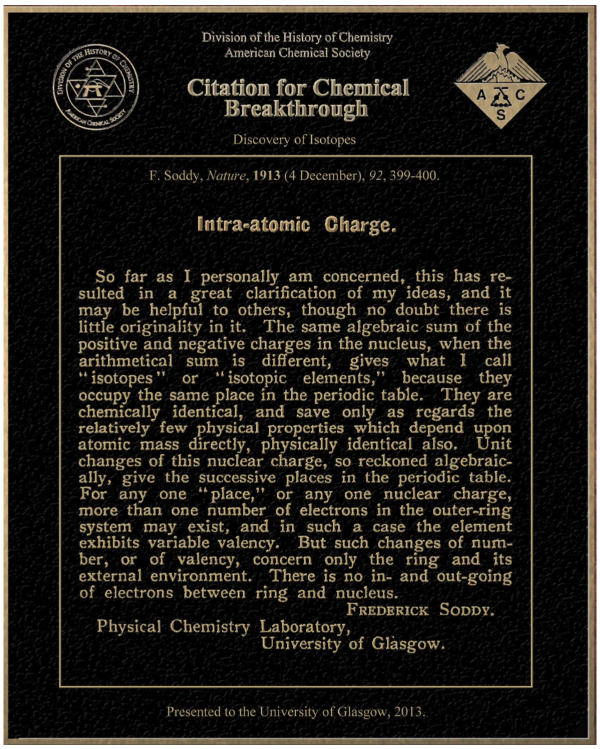

***Figure 1**. Plaque of the 2013 Citation for Chemical Breakthrough award honoring the discovery of isotopes.*

Methodology

Identifying a breakthrough paper is a rather artificial sort of historical exercise. It is akin to the very human impulse found in most awards programs to celebrate achievement and to apportion credit. Still, the notion that a threshold event can be identified before which an important concept did not exist or was not established and after which it does exist or is established is not a standard historiographical outlook.

Accounts of the establishment of the isotope concept have tended to be integrative, describing the contributions of a variety of investigators addressing a diversity of problems from a plurality of perspectives using a multiplicity of tools. Soddy himself engaged in this sort of historical treatment of the development of this very concept in his lecture upon receiving the 1921 Nobel Prize in chemistry in part for isotopes (9). Max Wolfsberg, W. Alexander Van Hook, and Piotr Paneth rely largely on Soddy's account in the central portion of the lengthy historical chapter that introduces their 2009 monograph on isotopes (10). In between, on the occasion of the centenary of Soddy's birth, came a symposium volume on his life and work, including, of course, the discovery of isotopes (11). And the physicist and historian of radioactivity, Alfred Romer, published a collection of key papers on radiochemistry and isotopes, accompanied by an extensive historical essay (3).

What I did for the CCB program was not integrative but differential. I examined five papers, combing each for how it contributes to the isotope concept. What I found is summarized below, including reasons for recommending the short letter in *Nature* that introduced the term isotope (2) as the paper to be recognized for the award.

In more or less chronological order, the papers under consideration were:

- a review article on radioactivity Soddy wrote for the Chemical Society of London's *Annual Reports on the Progress of Chemistry* for 1910 (12)
- the paper on the chemistry of mesothorium published by the Chemical Society in 1911 (7) and actually nominated for recognition
- an article titled "The Radio-elements and the Periodic Law," written and published in February 1913 in the *Chemical News* (13)
- a letter taking up just under a full page of type in the December 4, 1913, issue of *Nature* (2)
- a review article on radioactivity Soddy wrote for the Chemical Society's *Annual Reports on the Progress of Chemistry* for 1913 (14)

So much for what I was looking *at*. What was I looking *for*? What is at the core of the isotope concept? Isotopes are different forms of the same element. Upon reflection, this formulation appears to be robust and historically appropriate, for it uses terms and concepts that were current at the time under examination. "Same element" implies applying criteria by which elements can be compared and distinguished, but does not specify those criteria or fix them in time. Similarly "different forms" requires observable difference, implicitly recognizing that what is observable changes with time and technology.

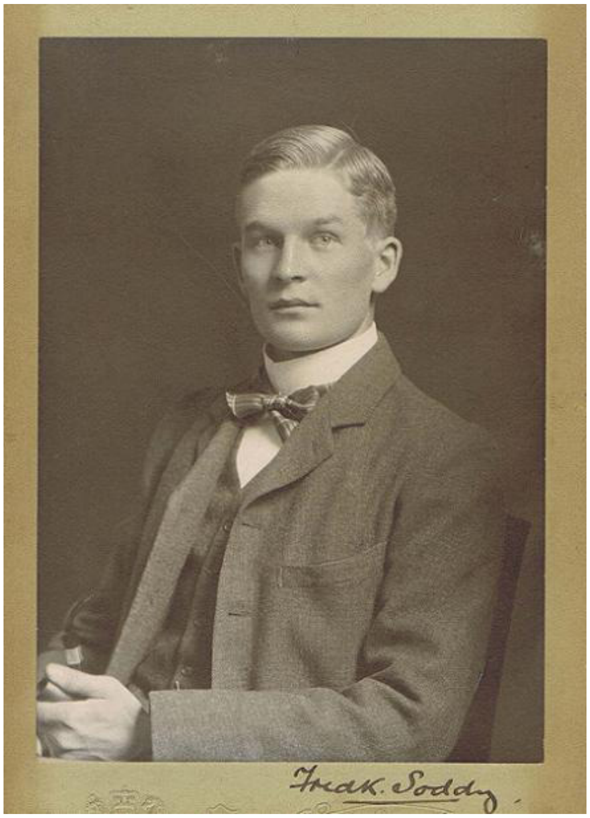

Figure 2. *Portrait of Frederick Soddy (1877-1956) from the earliest years of the twentieth century. With permission of the Frederick Soddy Trust.*

The Chemistry of Mesothorium, 1911 (7)

The paper that was actually nominated describes experiments conducted on the mineral thorianite, which, as one would expect from the name, contains thorium. As Soddy wrote,

> Thorianite is, from the radioactive point of view, the most complex material it is possible to work with, as it contains every one of the thirty or more radioactive elements known, in important quantity.

By this time, most of the details of radioactive decay chains had been worked out. It was known that the decay sequences of thorium and radium were independent of each other, and it was at least strongly suspected that that of actinium was also independent. Thorianite contained decay products from all of these chains.

Based on his experiments, Soddy concluded that "mesothorium-1, radium, and thorium-X appear to form a trio of chemically non-separable elements." That certainly sounds a lot like isotopes. Today we refer to those radioelements as radium-228, radium-226, and radium-224 respectively. Soddy was able to detect each of the radioelements based on its radioactive decay properties (decay times in particular), but he could not separate them or even enrich or deplete them by techniques of wet analytical chemistry, such as selective precipitation or fractional crystallization. Later in the paper, Soddy notes, "there is clear evidence also that thorium-X is always separated in any chemical operation in the same proportion as mesothorium and radium."

Soddy goes on to mention an attempt by Strömholm and Svedberg in 1909 to place some chemically similar ("isomorphic") radioelements in the periodic table (15). They had noted no chemical differences among the group radium, thorium-X, and actinium-X (known to us as radium-223) or among the group of three radioactive "emanations" from thorium, radium, and actinium (known to us as radon-222, -220, and -219, respectively). Strömholm and Svedberg had some inconsistent results for mesothorium—inconsistent between their own initial and subsequent experiments and inconsistent with what Soddy was reporting in this paper. Those investigators had placed mesothorium (which we know to be an isotope of radium) with thorium and radiothorium (which we recognize as thorium-232 and -228 respectively), and believed them to be analogous to the rare earths. Soddy writes, "The elements radiothorium, mesothorium, thorium suggest anything rather than the rare-earth group lanthanum to ytterbium."

The next words of the paper are the ones cited by the nominator and quoted above. Here Soddy asserts chemical identity among these "elements," even as he seems to despair of reconciling this phenomenon to the periodic law. Soddy refers to "elements *presumably* [my emphasis] of different atomic weight" because those atomic weights were at this time nearly all inferred rather than measured. The radioelements discovered over the previous 15 years were usually isolated in insufficient quantity or purity to enable measurement of their atomic weight. However, the decay sequences were sufficiently well known along with the masses of α (and β (16)) particles to infer atomic weights. For example, when thorium (atomic weight 232) emits an α particle, its daughter (mesothorium) must have an atomic weight of 228. Radium (17) and the so-called emanation of radium (18) were the only radioelements whose atomic weights had been experimentally determined by this date.

Soddy goes on to list other examples of inseparable elements that seem to have the same chemical behavior: the pair radiolead (now known as lead-210) and "lead" (now known to be a mixture mainly of isotopes 208, 207, and 206); and the trio thorium, radiothorium, and ionium (thorium-230). The chemical similarity in these cases was even greater than that among the rare earths, Soddy notes, and he was particularly impressed by the inability of Auer von Welsbach, an expert in rare earth chemistry, to separate thorium and ionium. Here Soddy adds a prescient speculation:

> The question naturally arises whether some of the common elements may not, in reality, be mixtures of chemically non-separable elements in constant proportions, differing step-wise by whole units in atomic weight. This would certainly account for the lack of regular relationships between the numerical values of the atomic weights.

Clearly much of the isotope concept as we know it is present in this article, in particular moieties whose chemical behavior is identical (not just similar) even though their atomic weight is different. Romer, an expert on the history of radioactivity and radiochemistry active 50 years ago, went even further: "In this paper, completed at the close of 1910, he [Soddy] proposed on somewhat less than adequate evidence a fully realized hypothesis of isotopes" (3).

I see this paper a bit differently, though, as lacking a key feature of the isotope concept, namely the conviction that the entities in question were the same element. What are the appropriate criteria for deciding whether or not two distinct entities are the same element? This is not an issue Soddy addresses. Experimental chemical behavior, including separability by wet chemical operations, would have been one reasonable criterion at the time. Classification in the periodic table according to the periodic law would have been another. An orthodoxy about elements from the time of Dalton that "the ultimate particles of all homogeneous bodies are perfectly alike in weight, figure, &c.," (19) may have been yet another criterion. Soddy failed to see how the state of knowledge about radioelements could be reconciled with the periodic law, but he had, apparently, found the absolute identity of all atoms of an element to be unnecessary nearly a decade earlier (20).

Radioactivity, 1910 Review Article (12)

If the nominated paper from 1911 is not quite the breakthrough paper, then it seems unlikely that a review article of 1910 could be in the running. One should note, however, that it is not clear whether this paper or the one just discussed was written first. The review article contains several references to work published in early 1911, including the paper just discussed (and not limited to Soddy's own work).

The relevant portions of this article and the 1911 paper are very similar. Both refer to Strömholm and Svedberg's work in trying to classify the radioelements (15). Indeed, Soddy places their work as the starting point of the relevant section of the review article. He rehearses several examples of apparent chemical identity among radioelements, concluding,

> Indeed, when it is considered what a powerful means radioactive methods of measurement afford for detecting the least change in the concentration of a pair of active substances, and the completeness and persistence of some of the attempts at separation which have been made, the conclusion is scarcely to be resisted that we have in these examples no mere chemical analogues, but chemical identities.

This review article also contains the speculation that the phenomenon extends beyond the realm of radioactivity:

> The recognition that elements of different atomic weight may possess identical chemical properties seems destined to have its most important application in the region of inactive elements, where the absence of a second radioactive nature, totally unconnected with the chemical nature, makes it impossible for chemical identities to be individually detected.

Among the interesting differences in the treatment of the subject in these two articles is the relationship of the phenomenon to the periodic law. Here Soddy writes,

> These regularities may prove to be the beginning of some embracing generalisation, which will throw light, not only on radioactive processes, but on the elements in general and the Periodic Law.

Contrast this statement with the one in the 1911 paper in which Soddy seems to find the same phenomenon at variance with the periodic law. It appears, at the time that Soddy wrote these two articles, that he does not know *whether* identical elements and the periodic law fit together; certainly he does not know *how* they fit together.

All in all, the chemical identity of different radioelements is discussed in greater detail in the 1911 paper (Ref. 7) than in this one (Ref. 12). Here it comprises just the last page and a half of a review that covers many aspects of radioactivity in the course of just over 30 pages.

Chemical News, Early 1913 (13)

In this paper, Soddy places the known radioelements in the periodic table. He makes use of the so-called displacement laws developed mainly by Kasimir Fajans (21) and himself (22). Emission of an α particle moves a radioelement two places to the left in the periodic table; emission of a β particle or a "rayless" transformation moves a radioelement one place to the right in the table. Soddy begins the paper by referring to chemically non-separable elements, which he had discussed previously (in Ref. 7). In this later paper, he puts the radioelements in various places in the periodic table, predicting that some will be non-separable from previously known elements. The paper includes several predictions about short-lived species in the various radioactive decay chains. For example, he expects that radium-A and radium-C′ would be non-separable from polonium and radium-C_2 from thallium. Similarly, thorium-A and thorium-C′ would be non-separable from polonium and thorium-D from thallium. He makes a distinction between homologues that are separable, namely radium from barium and polonium from tellurium. These are separable from each other; they belong in the same *group* in the periodic table, but in successive periods.

Thus, this paper remedies one of the "deficiencies" (when viewed with the advantage of hindsight) of the 1911 paper: non-separable elements are no longer "in direct opposition to the principle of the Periodic Law;" they can be reconciled to it. To be sure, this paper does not use the phrase "chemical identity" as did the 1911 paper, but that change in terminology does not in fact represent any retreat from the assertion of chemical identity. As Soddy had written in his review article on radioactivity for 1911 (23)

> These statements [describing radio-elements as non-separable] are not at all, as might be supposed, merely negative expressions of failure due to the difficulties of investigation. The statement, for example, that mesothorium-1 is non-separable from radium completely describes the chemistry of that substance so far as it is known, and indicates, for example, that it is differentiated most definitely from every one of the whole of the rest of the common elements.

Nature, Late 1913 (2)

This letter, published in early December 1913 and comprising just under a solid page of text, introduces the term isotope (24). The letter also defines the term in a way that we would recognize today, despite what we would describe as an incorrect picture of the nucleus. But the title of the letter, "Intra-Atomic Charge," and much of its content is concerned with another, albeit closely related, physical concept that was also aborning at the same time: atomic number. What Soddy called the "Intra-Atomic Charge" is more or less what we would call the nuclear charge, although Soddy (and many others) thought of this as a net nuclear charge, believing that the nucleus contained both positive charges (like α particles) and negative charges (like β particles). That is, the atom, in his mind, had both outer electrons, as in Bohr's model of the atom (also in embryo at this time (25)) and electrons in the nucleus.

A Dutch lawyer and amateur physicist, Antonius van den Broek, had speculated in *Nature*, in a letter published just the week before (26), that the nuclear charge of an element was equal to its atomic number, that is, to its place in the periodic table. Note, by the way, the "direction" of this equality, made clear by Soddy's words (2):

> The intra-atomic charge of an element *is determined by* its place in the periodic table *rather than by* its atomic weight [my emphasis], as concluded by A. van der [*sic*] Broek…

Most scientists today would say that that an element's nuclear charge determines its place in the periodic table, rather than *vice versa*. The point is that the position in the periodic table was not primarily related to atomic weight, but to something that varied more regularly, namely (net) nuclear charge. Soddy had already entertained the possibility that non-separable elements of different atomic weights were responsible for the irregularity of atomic weights in the periodic table (12).

In late 1913 neither the place of an element in the periodic table nor its nuclear charge was known to great precision. Rutherford's scattering experiments left the nuclear charge uncertain by about 20% (26). The ordinal number (place in the periodic table) of the heaviest elements was a bit less uncertain, but only recently. No one was yet sure how many rare earths there were, but the recent papers by Soddy and Fajans suggested that the large number of radio-elements did not all occupy separate places in the periodic table. Fajans had even suggested a term, *Plejade*, for a group of inseparable elements that occupy the same place in the periodic table (27). Soddy explains how these developments in radiochemistry are consistent with that of atomic number, and reckons the intra-atomic charge of uranium to be about 90 rather than the 120 it would be if the nucleus were made up entirely of α particles (thereby making the charge number half of the mass number). Note that the first of two papers by Moseley on the X-ray spectra of the elements (28)—papers generally credited with putting the notion of atomic number on a firm physical footing of (net) nuclear charge—appeared at just about this same time, December 1913.

The last paragraph of Soddy's letter is worth examining in detail:

> So far as I personally am concerned, this has resulted in a great clarification of my ideas, and it may be helpful to others, though no doubt there is little originality in it.

Even if this sentence reflected false modesty, Soddy can not have expected that the next sentence would introduce a term taught to every introductory chemistry student a century later. The paragraph continues

> The same algebraic sum of the positive and negative charges in the nucleus, when the arithmetical sum is different, gives what I call "isotopes" or "isotopic elements," because they occupy the same place in the periodic table.

Soddy may have the wrong nuclear building blocks in mind, but the main idea here retains its validity: moieties that have the same net nuclear charge occupy the same place in the periodic table, whether or not their nuclei differ in other respects. Next he reasserts the chemical identity of isotopes:

> They are chemically identical, and save only as regards the relatively few physical properties which depend on atomic mass directly, physically identical also.

So there, within a few sentences, are the key points of the isotope concept.

Radioactivity, 1913 Review Article (14)

The letter to *Nature* (2) was published before Soddy's review article on radioactivity in *Annual Reports on the Progress of Chemistry* for 1913. So there are already two obstacles to naming the latter paper the breakthrough. One is priority and the other is the diffuse nature of review articles. In general, I would consider a review article a breakthrough paper only if it ties together pieces of a concept or theory that had not previously been assembled. And in principle, the isotope concept as I have described it is a good candidate for such a synthesis, combining as it did chemical evidence assembled over many years and fitting that evidence into the periodic law. But Soddy had completed that synthesis already. Granted, the review article could and did go into the component parts in greater detail, but it was not the first formulation of the crucial synthesis. Indeed, it did not even marshal those components as pieces of evidence in support of the isotope concept.

In any event, the earlier publication would merit recognition as the breakthrough paper unless that publication was obscure. Such was not the case, however, with *Nature*. Granted, *Nature* in 1913 was not the powerful brand in scientific publishing that it is today. After all, even amateurs like van den Broek could get letters into its pages, and quite rapidly too. But prominent members of the scientific community, especially in England, also used letters to *Nature* for rapid communication (29). Clearly, what was published there could not be said to languish in obscurity.

The lead portion of Soddy's review article for 1913 was the reconciliation of the radioelements with the periodic law. The first three and a half pages of the 27-page article were given to the displacement laws, including a large figure. Several of the following pages went into further detail on recent developments of how particular radioelements fit into decay series and/or the displacement law. This portion of the article alludes to the role played by chemically identical but radioactively distinct species in piecing together how the elements fit into the periodic system. It explains the terms isotope and isotopic, and then uses those terms. And it goes on to mention evidence for isotopes outside the radioelements: "F. Ashton" (Aston) had reported a neon of mass 22 along with the usual mass-20 neon.

Concluding Observations

That scientific knowledge is constructed incrementally is a truism that hardly requires defending. In the case of the emergence of the isotope concept, we can see increments within the thought of the single individual with whom the concept is closely associated (deservedly so, in my opinion). Of course that individual did not work alone. In his Nobel address (9), Soddy acknowledges key pieces of evidence about non-separable elements published by McCoy and Ross (30), Strömholm and Svedberg (15), Auer von Welsbach, and others. The fact that a variety of investigators often contribute key pieces of evidence for a particular discovery is the most obvious way in which incrementalism manifests itself in science. A recognized advance is based on a synthesis of key pieces of evidence, sometimes by the discoverer of the latest piece, sometimes (as in the demise of vitalism or the establishment of the germ theory of disease) only after an unofficial consensus after a considerable lapse of time.

In this case, we can observe the evolution of the synthesis of the isotope concept. First Soddy (7, 12) concludes that chemically identical elements that have different physical properties (such as atomic weight and half life) exist. At this point, Soddy had identified a problem that challenges the notion of element and sits uncomfortably with the periodic law. Within a couple of years, though, he (and Fajans) figure out how to fit these elements into the periodic table (13). Less than a year later, Soddy displays considerable (albeit not perfect) insight into the physical quantity that these entities had in common. Whether one uses chemical identity, the periodic law, or this new physical quantity of net nuclear charge to identify elements, Soddy notes that different varieties of the same element exist. And he coins a term for the phenomenon (2, 14).

Not only had he not been working "on" isotopes at any time during this process, he devoted no publication to announcing or proposing the concept; there was no public eureka moment. This is in marked contrast, of course, to publications that announce results at the end of investigations designed to find just such results, and even to publications that announce results at the end of a search for something entirely different. Examples of the former include the detection of gravitational waves (31) and the structure of DNA (32). Examples of the latter include X-rays (33) and the new gas Joseph Priestley called dephlogisticated air (34). Although no one was looking for isotopes, they were not a surprise in the same way that these latter accidental discoveries were surprises. The isotope concept was more of an explanation than a phenomenon, which may help account for why no publication was devoted exclusively or primarily to it. It is clearly not the case that the concept proved to be useful only in retrospect.

In addition to providing an example of incrementalism in science, the development of the isotope concept illustrates the utility of a couple of modes of publication that are sometimes underappreciated. One of these is the publication of negative results (35). The inability of several investigators to separate radioelements was obviously a prerequisite to the realization that they could not be separated by chemical means. This "failure" provided insight into the relationship among these "elements." The fact that such failure was reproducible and known throughout the radioactivity community was important in establishing inseparability as a fact and not an artifact (of deficiency of technique, for example).

The other mode of publication that played a crucial role in this story is the writing of review articles. Review articles are of obvious utility to their readers, whether they are established investigators of a subject or newcomers to it. Here, however, we see the value of review articles to their author. Soddy published annual reviews in the field of radiochemistry starting in 1904. These articles gave him the benefit of intimate knowledge of the variety of radioelements, their behavior, and their chemistry. Far from detracting from publishing primary research, his publication of these secondary research articles put him in a position to make the synthesis described above.

The nature of scientific discovery has been oft debated among scientists, historians and philosophers, particularly in the context of apportioning credit. In such discussions, a key question often is how much of the concept—as understood at the (later) time of the debate—must have been present for it to be considered "discovered." In discussing Soddy's 1911 paper on mesothorium (Ref. 7), I considered an explicit recognition of different "radioelements" being the same element a key part of the isotope concept missing from that paper; I expressed an unwillingness to date the birth of the concept to the recognition of chemical identity and inseparability. Here I explicitly recognize the historical contingencies that permit me to do so, namely the fact that the aspects I identified as "missing" in that paper were present within three years—largely due to the work of same investigator. There was no long gap between "identical elements" and isotopes as there was between the periodic table and atomic number, between evolution by natural selection and the mechanism of transmission of heritable characteristics, and between the hypothesis of continental drift and the mechanism of plate tectonics. Investigators in the field of radioactivity did not have to wonder for long whether the phenomenon of "identical elements" was a real but as yet unexplained aspect of nature or a stumbling block that did not fit their understanding.

Acknowledgment

I would like to thank an anonymous referee for many suggestions that improved the paper.

References and Notes

1. Presented at the 248th National Meeting of the American Chemical Society, San Francisco, CA, Aug. 10, 2014, HIST 10.
2. F. Soddy, "Intra-Atomic Charge," *Nature*, **1913**, *92*, 399-400.
3. Ref. 2 (pp 250-252) and several of the others cited below are collected in A. Romer, Ed., *Radiochemistry and the Discovery of Isotopes*, Classics of Science Series, Dover, New York, 1970. The volume is a collection of important papers on the subject, in English translation if the original publication was not in English, preceded by a lengthy historical essay.

4. American Chemical Society Division of the History of Chemistry, Citation for Chemical Breakthrough Awards, http://www.scs.illinois.edu/~mainzv/HIST/awards/citations_chem-breakthroughs.php (accessed June 19, 2017).

5. J. I. Seeman, "HIST's Citation for Chemical Breakthrough Awards: the First Paper *or* the 'Breakthrough' Paper?", *Bull. Hist. Chem.*, **2013**, *38(1)*, 4-6. J. Gal, "Citation for Chemical Breakthrough Awards: Choosing Pasteur's Award-Winning Publication," *Bull. Hist. Chem.*, **2013**, *38(1)*, 7-12. N. C. Craig, "Charles M. Hall's Persistent Quest of Patents for Refining Aluminum Metal by Electrolysis," *Bull. Hist. Chem.*, **2013**, *38(1)*, 13-18. D. E. Lewis, "Citation for Chemical Breakthrough Award: Mendeleev's Periodic System of Elements," *Bull. Hist. Chem.*, **2014**, *39(1)*, 1-6.

6. J. I. Seeman, Nominations for 2013 Citations for Chemical Breakthrough awards (Nov. 2013 memo).

7. F. Soddy, "The Chemistry of Mesothorium," *J. Chem. Soc. Trans.*, **1911**, *99*, 72-83. Also Ref. 3, pp 179-190.

8. Equipped with the isotope concept, we might call them radioisotopes or radionuclides (the latter preferred by the IUPAC Gold Book, A. D. McNaught and A. Wilkinson, Eds., *IUPAC Compendium of Chemical Terminology*, 2nd ed., Blackwell Scientific Publications, Oxford, 1997. XML online corrected version http://goldbook.iupac.org/ accessed June 19, 2017). But before the isotope concept was established, each new radioactive substance that could not be separated by chemical means and that could be distinguished from all such previously known substances (by half-life, for example) was believed to be a new radioactive element or "radioelement."

9. F. Soddy, Nobel Lecture: "The Origins of the Conception of Isotopes," 1922, Nobelprize.org, http://www.nobelprize.org/nobel_prizes/chemistry/laureates/1921/soddy-lecture.pdf (accessed June 15, 2017).

10. M. Wolfsberg, W. A. Van Hook, and P. Paneth, "A Short History of Early Work on Isotopes" Ch. 1, in *Isotope Effects: in the Chemical, Geological, and Bio Sciences*, Springer, Dordrecht, 2009, pp 1-36.

11. G. B. Kauffman, Ed., *Frederick Soddy (1877-1956): Early Pioneer in Radiochemistry*, D. Reidel, Dordrect, the Netherlands, 1986.

12. F. Soddy, "Radioactivity," *Annu. Rep. Prog. Chem.*, **1910**, *7*, 256-286.

13. F. Soddy, "The Radio-elements and the Periodic Law," *Chem. News*, **1913**, *107*, 97-99. Also Ref. 3, pp 219-226.

14. F. Soddy, "Radioactivity," *Annu. Rep. Prog. Chem.*, **1913**, *10*, 262-288.

15. D. Strömholm and T. Svedberg, "Untersuchungen über die Chemie der radioaktiven Grundstoffe. II.," *Z. anorg. Chem.*, **1909**, *63,* 197-206.

16. Soddy mentions only α particles when discussing estimated atomic weights. Presumably he does not mention the effect of β emissions on atomic weights because he knew that effect to be negligible. The paper includes a decay sequence that involves β emission, and the β particle was already known to be an electron.

17. M. Curie, "Sur le poids atomique du radium," *C. R. Séances Acad. Sci.*, **1902**, *135*, 161-163.

18. W. Ramsay and R. W. Gray, "La densité de l'émanation du radium," *C. R. Séances Acad. Sci.*, **1910**, *151*, 126-128.

19. J. Dalton, *A New System of Chemical Philosophy*, Part I, Manchester, England, 1808, p 143.

20. Soddy had thought he identified a physical property in which atoms of the same element differed. The property was the lifetime of unstable elements, some of which decayed after a very short time and some after a nearly infinite time to yield observed average half lives. (That is, Soddy interpreted the actual decay times of individual atoms of a given radioelement as definite but widely distributed about a certain mean value, whereas we regard the actual decay times of individual radionuclides as indefinite and distributed about the same mean.) See F. Soddy, "The Evolution of Matter as Revealed by the Radio-Active Elements," *Manchester Memoirs*, **1904**, *48*(8), 1-42 on p 21.

21. K. Fajans, "Über eine Beziehung zwischen der Art einer radioaktiven Umwandlung und dem elektrochemischen Verhalten der betreffenden Radioelemente," *Phys. Zeit.*, **1913**, *14*, 131-136; trans. in Ref. 3, pp 198-206. K. Fajans, "Die Stellung der Radioelemente im periodischen System," *Phys. Zeit.*, **1913**, *14*, 136-142; trans. in Ref. 3, pp 207-218. Soddy refers to these two papers in his *Chemical News* article. Another important paper by Fajans would appear in print shortly thereafter: K. Fajans, "Die radioaktiven Umwandlungen und das periodische System der Elemente," *Ber. Dtsch. Chem. Ges.*, **1913**, *46*(1), 422-439.

22. Soddy refers also to the work of Alexander Russell, who was in print with a version of the displacement law before either himself or Fajans: A. S. Russell, "The Periodic System and the Radio-Elements," *Chem. News*, **1913**, *107*, 49-52. As Romer notes (3), Russell relied largely on Soddy's work. In addition, his version of the displacement law is less definite, permitting changes of two places in either direction upon emission of an α particle and of one place in either direction upon emission of a β.

23. F. Soddy, "Radioactivity," *Annu. Rep. Prog. Chem.*, **1911**, *8*, 269-301.

24. Credit for coining the term belongs to Dr. Margaret Todd, a friend of the Soddy family. See A. Fleck, "Frederick Soddy," *Biogr. Mem. Fellows R. Soc.*, **1957**, *3*, 203-216.

25. N. Bohr, "On the Constitution of Atoms and Molecules," *Philos. Mag.*, **1913**, *26*, 1-25.

26. A. van den Broek, “Intra-atomic Charge,” *Nature*, **1913**, *92*, 372-373.

27. K. Fajans, “Die radioaktiven Umwandlungen und die Valenzfrage vom Standpunkte der Struktur der Atome,” *Verhandlungen Dtsch. Phys. Ges.*, **1913**, *15*, 240-259. The *Verhandlungen* were at this time issued with the *Berichte*, so they may be bound with the latter. Fajans writes (p 245) “Für solche Gruppen untrennbarer Elemente, denen eine gemeinsame Stelle im periodischen System zukommt, möchte ich die Bezeichnung Plejade in Vorschlag bringen. [For such groups of inseparable elements which belong in the same place in the periodic system, I would like to propose the term Plejade.]”

28. H. G. J. Moseley, “The High-Frequency Spectra of the Elements,” *Philos. Mag.*, **1913**, *26*, 1024-1034. H. G. J. Moseley, “The High-Frequency Spectra of the Elements. Part II,” *Philos. Mag.*, **1914,** *27*, 703-713.

29. For example, Rutherford, who by this time was certainly among the scientific elite (Fellow of the Royal Society and Nobel laureate), published a letter in *Nature* also responding to van den Broek’s (Ref. 26) and appearing a week after Soddy’s (Ref. 2): E. Rutherford, “The Structure of the Atom,” *Nature*, **1913** (Dec. 11), *92*, 423.

30. H. N. McCoy and W. H. Ross, “The Specific Radioactivity of Thorium and the Variation of the Activity with Chemical Treatment and with Time,” *J. Am. Chem. Soc.*, **1907**, *29*, 1709-1718. Also Ref. 3, pp 128-139.

31. B. P. Abbott *et al.*, “Observation of Gravitational Waves from a Binary Black Hole Merger,” *Phys. Rev. Lett*., **116**, 061102 (2016).

32. J. D. Watson and F. H. C. Crick, “A Structure for Deoxyribose Nucleic Acid,” *Nature*, **1953**, 737-738.

33. W. C. Röntgen, “Ueber eine neue Art von Strahlen,” *Sitz.-Ber. phys.-med. Ges. Würzburg*, **1895**, 132-141.

34. J. Priestley, “Of Dephlogisticated Air, and of the Constitution of the Atmosphere,” Sect. III of *Experiments and Observations on Different Kinds of Air*, Vol. II, J. Johnson, London, 1775, pp 29-61.

35. The issue of negative results, particularly in medical and life sciences, has received considerable attention in the early twenty-first century, leading to journals or sections of journals devoted to publishing such results. The *Journal of Cerebral Blood Flow & Metabolism* instituted a negative results section in 2010: U. Dirnagl and M. Lauritzen, “Fighting Publication Bias: Introducing the Negative Results Section,” **2010**, *29*, 1263-1264. The *Journal of Negative Results in BioMedicine* (http://www.jnrbm.com, accessed June 19, 2017) describes itself as “an open access, peer-reviewed, online journal that provides a platform for the publication and discussion of unexpected, controversial, provocative and/or negative results in the context of current tenets.”

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