## **Rediscovery of the Elements** Soddy and Isotopes



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A Dinner Party in Glasgow, Scotland. During a social gathering in 1913 at the home of his inlaws, Frederick Soddy (1877-1956) (Figure 1) was musing what to call a new concept. For a decade a plethora of radioelements, each with a unique half-life, had been discovered at various universities throughout Europe and North America, and there was no room in the Periodic Table to accommodate them. To solve this problem, Soddy was proposing that radioelements with different half-lives but with the same chemical properties should be confined in one box in the Table. The scholarly Dr. Margaret Todd (1859-1918), an M.D. and novelist friend of his wife, Winifred, suggested<sup>1</sup> such elements be denoted by the Greek words "same place" ("iso-topos") (Figure 2). Soddy shortly announced his idea of "isotopes" (December 4, 1913) in the prestigious journal Nature.2

The Life of Frederick Soddy.<sup>3</sup> A native of Eastbourne in East Sussex, 85 km south of London, Frederick Soddy was trained at the University of Aberystwyth (Wales) and at Oxford University, graduating in 1898. After three years as researcher at Oxford, Soddy accepted the position of Demonstrator at McGill University, Montreal, Canada. After a year he joined the group of Ernest Rutherford



Figure 1. The home of Sir George and Lady Beilby, the parents of Frederick Soddy's wife, Winifred, at 11 University Gardens, Glasgow, Scotland. The home is of Victorian architecture and was built in 1884. Sir George was an industrial chemist who furnished 50 kilograms of uranyl nitrate to Soddy for his experiments. This home served as a popular meeting place for politics, science, business, and leisure. In the drawing-room of this home, the word "isotope" was coined by Soddy at the suggestion of Dr. Margaret Todd.

at that university, who was just beginning to investigate the strange emanation from thorium.<sup>4c</sup> For the next two years the two colleagues established the elemental nature of thoron (radon-220), developed the understanding of radioactive half-lives, and discovered the natural transmutation of the elements.<sup>4c</sup> It was actually Soddy the chemist, well versed in the history of alchemy, who first understood the significance of transmutation, before Rutherford the physicist.<sup>3</sup>

In 1903, Soddy returned to England and joined the research group of William Ramsay (1852–1916; the discoverer of the noble gases) at University College, London, to study the decay by-products of radium. In Ramsay's laboratory, Soddy established that helium was produced during the decay of radium, thus furnishing the first evidence of the transmuta-



Figure 2. This plaque is mounted at 11 University Gardens. The building, now known as the George Service House, presently houses the Humanities Advanced Technology and Information Institute (HATII) of the University of Glasgow.

tion of a known element to another known element; this research was the basis of the future understanding of the alpha particle.<sup>3</sup>

In 1904, Soddy became Professor at the University of Glasgow, where he formulated the concept of isotopes. After 10 years at Glasgow, Soddy moved to the University of Aberdeen (1914–1919), where he co-discovered the element protactinium ("eka-tantalum"). World War I had a distressing effect on Soddy; the death of Henry Moseley<sup>4d</sup> elicited the following from Soddy,3 "When Moseley was killed at Gallipoli I felt enraged. Sometimes I think that something snapped in my brain.... I felt that governments and politicians, or man in general, was not yet fitted to use science." Soddy moved in 1919 to Oxford University, where with a new mission he tried to establish economic policy based on scientific laws. He retired in 1936, after a frustrating period in his life, with a "sense of failure.... in the field of economics." He was at Oxford when he accepted the Nobel Prize in 1921 for his research in radio-elements and isotopy at Glasgow, and for teaching us that atoms could have "identical outsides but different insides."5

**The Discovery of the Radioelements** (see Figure 3). Radioactivity was discovered by Antoine Henri Becquerel when he observed uranium emitted invisible rays that fogged photographic plates wrapped with paper or metal



Figure 3 (LEFT). The decay sequences of the natural radioelements (Fajans and Soddy "Displacement Laws"), patterned after Soddy's original list<sup>10</sup> and graph.<sup>10</sup> Atoms decay by emitting an  $\alpha$ -particle (moving left two units and down four units) or a  $\beta$ -particle (moving right one unit). The list of natural radioelements includes some 40 members and organized into three main series: U-238, originally identified as U1 ("radium series," the black plot), U-235, originally identified as "actinouranium" ("actinium series," the red plot), and Th-232, originally identified as merely "thorium" ("thorium series," the blue plot). The atomic number and mass for each radioelement can be determined from the respective abscissa and ordinate. For each radioelement the original name is given (in addition to the customary elemental abbreviations, Rd = radio, Ms = meso; for example RdAc = radioactinium). (For a more complete description including half-lives of the radioelements, see reference 11).



foil.<sup>4</sup><sup>b</sup> His discovery prompted the search for new radioactive elements by Marie Curie, who canvassed all known elements and determined that of these only uranium and thorium exhibited activity.<sup>4</sup><sup>b</sup> After noticing that ores of uranium were much more radioactive than expected, she launched her classic research with her husband, Pierre Curie, to discover polonium and radium in 1898, and her colleague André Debierne discovered actinium in 1899.<sup>4</sup><sup>b</sup>

The discovery of radioactive elements continued, which became known as the "radioelements." In 1900, William Crookes (1832-1919) - the discoverer of thallium - made an astonishing observation: he found that purified uranium nitrate (prepared either by precipitation with ammonium carbonate or by extraction with ether) exhibited no radioactivity at all!6 (This puzzling observation was shortly explained by Rutherford; vide infra). Instead, radioactivity resided in the separated residue, which Crookes named uranium X, or Ur-X (later named UX1). If Crookes had followed up this work, he would have been the discoverer of the natural transmutation of elements, because UX1 was actually thorium-234, produced by the  $\alpha$ -decay of uranium-238.

**The explanation of UX1 by Rutherford.** Now active in his research at McGill University (where he discovered the element radon)<sup>4c</sup>

Ernest Rutherford attempted to reproduce Crookes' work, but was perplexed by his observation that in his hands purified uranium was still radioactive. By following Crookes' procedure exactly, Rutherford realized the discrepancy was due to Crookes' using the older Becquerel method of radioactivity detection (fogging of wrapped photograph plates) instead of the "electric method" which utilized an ionization chamber pioneered by Pierre Curie<sup>4b</sup> (Rutherford used the Dolezalek electrometer,<sup>7</sup> a refinement of this method<sup>4b</sup>). On the basis of his work at Cambridge<sup>4c</sup> where he defined  $\alpha$ and B-rays, Rutherford realized that pure uranium (and its compounds) was an  $\alpha$ -emitter (whose rays were stopped by paper or foil) and that UX1 was a  $\beta$ -emitter (whose rays passed through Becquerel's and Crookes' paper wrappings). Rutherford designed an ionization chamber in which he could readily distinguish  $\alpha$ -rays and  $\beta$ -rays by alternatively using shields of 0.1 mm aluminum foil (impervious to  $\alpha$ -rays) or 5 mm aluminum foil (impervious to β-rays).<sup>7</sup> (The third type of emission,  $\gamma$ -emission was recently discovered by Paul Villard in 1900, could be inferred by its stoppage using by 50 cm of aluminum.7) After a period of time, an undisturbed sample of purified uranium nitrate produced a fresh supply of UX1.7 Rutherford and Soddy determined the half-life of UX1 to be 22 days.

Figure 4 (ABOVE). Map of Glasgow. Sites of interest in the University of Glasgow area:

Site where Soddy proposed the concept of "isotope," 11 University Gardens - N55° 52.37 W04° 17.45. Joseph Black Building (modern chemistry building), University of Glasgow - N55° 52.32 W04° 17.57 Lord Kelvin's house, University of Glasgow - N55° 52.28 W04° 17.43.

Old Chemistry Building, Main Building, Gilbert Scott Building (now geography), where Soddy performed his work on isotopes - N55° 52.24 W04° 17.25.

Museum Stores, site of Soddy's equipment and models, Thurso Street - N55° 52.17 W04° 17.84.

The thorium and uranium series of radioelements (see Figure 3). Rutherford and Soddy turned to thorium and found it exhibited similar behavior. Reacting thorium nitrate with ammonia (to precipitate thorium hydroxide) gave a filtrate, which when concentrated gave a very active residue they called thorium-X (ThX). Careful study showed ThX in turn transmuted into emanation (thoron), which itself changed to an entire sequence of new radioactive products!<sup>3</sup> After the Christmas holidays of 1901, Rutherford and Soddy returned to study the original purified sample of thorium, which was found to be replenished with the  $\beta$ -emitter ThX. Soon Rutherford and his



Figure 5. This plaque dedicated to Soddy is mounted in the Joseph Black Building, the main chemistry building of the University of Glasgow.

colleague Soddy had carefully outlined a sequence of transmutations:<sup>7</sup>

Th (half-life =  $10^{9}$ yr)  $\rightarrow$  ThX (4 days)  $\rightarrow$ Emanation (54 sec)  $\rightarrow$  ThA (11 hr)  $\rightarrow$ ThB (1 hr)

Did other radioactive elements exhibit a cascade of radioelements? Rutherford and Soddy turned to radium and found the following sequence:<sup>7</sup>

Ra (1500 yr) → Emanation (4 days) → RaA (3 min) → RaB (28 min) → RaC (21 min) → RaD (40 yr) → RaE (6 days) → RaF (143 days)

More researchers join the hunt. The research of Rutherford and Soddy sparked the hunt and discovery of more radioelements by many scientists during the first decade of the 20th century (Figure 3). These researchers included Herbert Newby McCoy (1870-1945; University of Chicago), Alexander Smith Russell (1888–1972; Oxford University), Bertram Borden Boltwood (1870-1927; Yale University), Otto Hahn and Lise Meiter (1879-1968 and 1878-1968, respectively; Kaiser Willhelm Institute, Dahlem-Berlin), Kasimir Fajans (1887-1975; Karlsruhe University), and Frederick Soddy (and colleagues) now at the University of Glasgow (Figures 4–7).3,8,9 The list of radioelements ballooned during the first two decades of the 20th century, reaching a total of almost 40 members, perplexing the scientific world with a"radium series" (RaA through RaG, originating from uranium), a "thorium series" (ThA through ThD, originating from thorium), and an actinium series (AcA through AcD, orig-



Figure 6. At the University of Glasgow, Soddy performed his work in the southeast corner of the Main Building. His laboratory now houses the Geography Department. When he abandoned this working space, it had to be thoroughly scrubbed and reconstructed because of the intense radioactivity; even the doorknobs had to be replaced. Nearby is the historic home of William Thomson (Lord Kelvin, 1824–1907, named after the river that flows nearby).

inating from uranium). These three series included three different emanations of inert gases (radon, thoron, and actinon; all isotopes of radon), as well as later intermediate members named ionium (Th-230), mesothorium1 and -2 (Ra-228 and Ac-228), radiothorium (Th-228), eka-tantalum (Pa-231), etc., and all ending with inert lead.<sup>9</sup> Because the quantities involved were generally unweighable traces, the major way to characterize and identify these elements was by means of their half-lives.

It was unclear just how to organize this motley collection, whose half-lives and random behavior of  $\alpha$ - or  $\beta$ -emission exhibited no apparent pattern. A major additional problem was the impossibility of fitting these new "elements" into the Periodic Table — the situation was bad enough with the rare earths, a dozen unusual elements that seemed to form an independent series (Bohr's atom with the f-subshell was yet to be formulated). So how could one accommodate three times that many rogue radioelements?

The clue of chemical similarity. The key to organization of the radioelements was found through wet laboratory chemistry. As chemists worked their analytical separations, they noticed that there was difficulty in separating some of the members of the radioelements. Whereas the challenge of separating radium from chemically similar barium (solved by the Curies during their discovery of radium<sup>4b</sup>) had been "merely formidable," the problem of separating elements of a certain grouping appeared to be practically impossible, suggesting groupings of virtually indistinguishable chemical behavior.

The Fajans and Soddy Displacement Laws. Meanwhile, the roadmap (Figure 3) by which these radioelements were being transformed into one another was becoming understood. One could trace the zig-zag route of an atom as it followed its predestined path from its progenitor to stable lead by alternatively shedding  $\alpha$ - and  $\beta$ -particles. In early 1913, two defining articles appeared almost simultaneously by Fajans<sup>12b</sup> and Soddy,<sup>13</sup> which stated that all  $\alpha$ -decays resulted in movement two units to the left and all  $\beta$ decays in movement one unit to the right (the so-called Fajans and Soddy "Displacement Laws"). Whereas Fajans' approach was more theoretical, Soddy's conclusions were based on extensive chemical analysis, with a heavy dependence upon final analysis of his student Alexander Fleck (1889–1968) at Glasgow who had proven the chemical identicalness of a number of β-emitters.<sup>14</sup>

In his 1913 articles,<sup>12b</sup> Fajans organized the radioelements in the Periodic Table, lumping similar elements in a "pleiad,"<sup>12b</sup> a name connoting a tight cluster of chemical properties, alluding to the Pleiades, the well known star cluster in the constellation Taurus. Fajans then boldly proposed that there should be an element "UX2" just to the right of Crookes'UX1 — and several months later he and his student Oswald Helmuth Göhring (1889-?) electroplated out the element whose electrochemical



Figure 7. Dr. Joe Conally, our host at the University of Glasgow, looks upon one of Soddy's machines in the Historic Museum ("Museum Stores"). This is the "Statis machine," which has a column with a series of chambers connected by differently sized orifices; as water flows through the column, one can visualize the steady-state distribution of elements in a radioactive ore sample.



Figure 8. This is the main building of the University of Karlsruhe, 12 Kaisserstrasse; the original building of Universitat Fridericiana (N49° 00.56 E08° 24.72), where Lothar Meyer refined his Periodic Table. Behind lay the Chemical Institute (Kollegium) where Kasimir Fajans discovered "brevium"<sup>15</sup> (Pa-234m), the first isotope of element-91 in 1913 (now Kollegiengebäude der Ehrenhof, Englerstrasse 11 - N49° 00.61 E08° 24.72). Nearby is the Ständehaus, where in 1860 the international Chemical Congress was held, which inspired the conception of Dimitri Mendeleev's and Lothar Meyer's Periodic Tables.<sup>44</sup>

potential placed it in this vacant space (Figure 8).<sup>15</sup> This element could be detected because of its high activity, with a short half-life of 1.17 minutes, even though ponderable quantities could not be collected. Because "it was the only element in the 5th group of the last row," he felt justified in giving it a name, "brevium"<sup>16</sup> (now known to be protactinium-234m) (Note 1).

Fajans in his studies was using an electroplating technique whereby he could plate out traces of specific radioelements and could characterize each by determining its half-life. By contrast, Soddy was using wet laboratory procedures and was impressed with the apparent impossibility of chemical separation of various radioelements. Soddy considered the elements of a grouping not "a scattered pleiade," but as chemically identical. While Fajans had grouped the radioelements loosely in a table,<sup>12a</sup> Soddy drew a precise two-axis graph.13 Soddy understood the implications of chemical indistinguishability-for example, ionium (Th-230, the precursor to radium in the uranium sequence) was identical to native thorium (Th-232) (Note 2), even to the point of identical spectra,13 despite their different half-lives  $(7.54 \times 10^4 \text{ and }$ 1.40 x 1010 years, respectively). If one believed Soddy, then the disconcerting problem of stuffing too many elements into the Periodic Table



Figure 9. Otto Hahn Building (Thielallee 63 - N52° 26.85 E13° 17.11), of the present Freie Universität of Dahlem, Germany, previously the Kaiser Wilhelm Institute, where Otto Hahn and Lise Meitner discovered protactinium-231.<sup>24</sup> As has been done with so many buildings in Berlin, this edifice was extensively damaged in World War II but was reconstructed to closely replicate the original architecture. The building is now used for biochemical studies. Much important history has occurred here, including the discovery of nuclear fission (announced by the plaque attached to the turret, seen here to the right). On the second étage (third floor) is the Lise Meitner Hörsaal (lecture hall).

was resolved: different radioelements could be simply lumped into preexisting boxes. The term Soddy coined—isotope—was fully intended to convey the meaning that two species resided at the *identical* location in the Periodic Table, even if they had different atomic masses and halflives. Soddy was proposing an astonishing concept: A specific element, which historically was once considered not fully characterized without a specific atomic mass, now could be variable in its mass!

Corroboration of the predicted different masses for isotopes. It remained to prove that chemically identical elements *did* in fact have different masses. According to both Fajans and Soddy, lead (Pb) from different decay sequences should have different atomic masses. Fajans took the initiative and sent his student Max Ernest Lembert (1891-1925) to the laboratory of Theodore William Richards (1868-1928) at Harvard University, the recognized expert in atomic mass determinations. Richards and Lembert repeated the atomic mass determination for ordinary lead and reproduced the published value of 207.15. Lead from uranium samples from around the world gave a value ranging from 206.40-206.82,17 a value soon confirmed in three more laboratories. There was no doubt about it-lead from radioactive sources was lighter than ordinary lead, and there proved to be no spectroscopic differences. Later, the method of Moseley<sup>4d</sup>

"smoothed over the rough edges of isotopes"9 when his method of X-ray spectroscopy was expanded by Karl Manne Siegbahn (1886–1978, Nobel Laureate in physics in 1924) to include the atomic number of uranium. Siegbahn showed that even though uranium was composed of several isotopes (Note 3), nevertheless the element appeared to exhibit a clearly defined atomic number.<sup>18</sup> The method of mass spectrometry, developed by Francis William Aston (1877–1945, Nobel Laureate in chemistry in 1922), soon allowed the direct observation of isotopes and he wrote the definitive article on the isotopes of uranium, lead, and other radioactive elements.19 Even Fajans, who had long viewed each radioelement as a "new element," by 1923 had abandoned his concept of the scattered pleiads and had accepted the view that isotopes should be tightly boxed in individual cubicles of the Periodic Table.<sup>20</sup>

**Epilogue – the construction of the nucleus.** What was the genesis of the different isotopes of an element? The prevalent view during the first quarter of the twentieth century was that the nucleus was composed of protons and a lesser number of electrons that partially neutralized the positive charge, but it was not known where these electrons resided. Fajans thought all electronic processes (either chemical redox or nuclear decay) must occur in the same region of the atom, since the particles *(continued on page 74)* 

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(electrons) were identical. However, a logical argument of Soddy in 1913 underscored the clear difference between orbiting electrons and nuclear electrons.<sup>21</sup> Soddy compared the oxidation of tetravalent uranous to give hexavalent uranyl (U<sup>4+</sup>  $\rightarrow$  U<sup>6+</sup>) with the radioactive decay of UX1, which loses two electrons to form U2 (Th-234  $\rightarrow$  U-234). If electron loss (oxidation) and beta loss (radioactive decay) were identical processes, then UX1 and U2 should be chemically identical —but instead they could be cleanly separated. Clearly there was something special about "nuclear electrons."

The answer to the riddle of the nuclear electrons was solved by James Chadwick (1891–1974, Nobel Laureate in physics in 1935), who discovered<sup>22</sup> the neutron in 1932. This neutral particle was not simply a combination of a proton and an electron, but an entirely new particle that could eject an electron (to become a proton) in special cases defined by a low proton/neutron ratio. With Bohr's description of electronic structure,<sup>23</sup> a complete picture emerged of the atom and its behavior in the radioelement decay sequences. With this understanding, the story of radiochemistry with its "suicidal success"9 was completed, paving the way for the next chapter-"Nuclear Chemistry" and the birth of the artificial elements. This shall be the topic of a future article in The HEXAGON "Rediscovery" series.

## Notes.

Note 1. "Brevium" was Pa-234m. "Protactinium" (Pa-231) was discovered and isolated in weighable quantities four years later, independently by Hahn and Meitner<sup>24</sup> in Berlin (Figure 9) and Soddy and Cranston<sup>25</sup> in Aberdeen (Figure 10), and named because it was the precursor to actinium (Pa-231 -> Ac-227). Fajans also predicted a pleiad just to the right of the emanation pleiad (radon)12-verified when francium with a half-life of 21.8 minutes was discovered in 1939 by Marguerite Catherine Perey (1909-1975).26 Fajans had a celebrated career but never received the Nobel Prize, perhaps in part owing to political enemies he made in the science world by his brash behavior.27

Note 2. Ironically, it was later found that isotopes *can* have different chemical and physical properties, particularly where a small atomic mass can be a significant difference in the zeropoint energy; deuterium was discovered by Urey in 1931 by distillation of liquid hydrogen<sup>28</sup> and pure heavy water was prepared in 1933 by G. N. Lewis using electrolysis.<sup>29</sup>

Note 3. The possibility of more than one isotope of uranium giving rise to additional decay sequences was first proposed by Auguste



Piccard (1882–1962) of high-altitude (balloon flights) and deep sea (bathyscape) fame. In 1917, he proposed<sup>30</sup> an element actinouranium, incorrectly suggesting a mass of 239; the correct mass of 235 was established by the mass spectral work of Aston,<sup>19</sup> and Rutherford immediately realized<sup>31</sup> that protactinium (discovered by Hahn and Soddy independently) would have a mass of 231 (promptly verified in Hahn's laboratory). Piccard's concept of a naturally occurring isotope of uranium giving rise to the actinium series was acknowledged by Soddy in his Nobel address.<sup>5</sup>

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Figure 10. Marischal College in Aberdeen, Scotland, where Soddy independently discovered<sup>25</sup> "eka-tantalum" (protactinium-231) in 1917 (Broad Street, N57° 08.93 W02° 05.81). Although some preliminary work on eka-tantalum had been done in Glasgow, his finished objective was not completed until he moved here in 1917.

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