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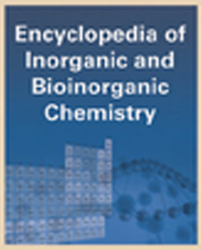
William J. Evans • Timothy P. Hanusa

The Heaviest Metals

Science and Technology of the Actinides and Beyond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Science and Technology of the Actinides and Beyond

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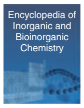

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THE HEAVIEST METALS:

Science and Technology of the Actinides and Beyond

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Encyclopedia of Inorganic and Bioinorganic Chemistry

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Series Preface

The success of the *Encyclopedia of Inorganic Chemistry* (EIC), pioneered by Bruce King, the founding Editor in Chief, led to the 2012 integration of articles from the *Handbook of Metalloproteins* to create the newly launched *Encyclopedia of Inorganic and Bioinorganic Chemistry* (EIBC). This has been accompanied by a significant expansion of our Editorial Advisory Board with international representation in all areas of inorganic chemistry. It was under Bruce's successor, Bob Crabtree, that it was recognized that not everyone would necessarily need access to the full extent of EIBC. All EIBC articles are online and are searchable, but we still recognized value in more concise thematic volumes targeted to a specific area of interest. This idea encouraged us to produce a series of EIC (now EIBC) Books, focusing on topics of current interest. These will continue to appear on an approximately annual basis and will feature the leading scholars in their fields, often being guest coedited by one of these leaders. Like the Encyclopedia, we hope that EIBC Books continue to provide both the starting research student and the confirmed research worker a critical distillation of the leading concepts and provide a structured entry into the fields covered.

The EIBC Books are referred to as *spin-on* books, recognizing that all the articles in these thematic volumes are destined to become part of the online content of EIBC, usually forming a new category of articles in the EIBC topical structure. We find that this provides multiple routes to find the latest summaries of current research.

I fully recognize that this latest transformation of EIBC is built on the efforts of my predecessors, Bruce King and Bob Crabtree, my fellow editors, as well as the Wiley personnel, and, most particularly, the numerous authors of EIBC articles. It is the dedication and commitment of all these people that are responsible for the creation and production of this series and the "parent" EIBC.

Robert A. Scott
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January 2019

Volume Preface

The Heaviest Metals: Science and Technology of the Actinides and Beyond is focused on the elements of highest atomic number—the actinides (Ac–Lr) and transactinides (Rf–Og). Their history spans virtually the entire era of modern chemistry, from the discovery of uranium by Klaproth in 1789 to the confirmation of the names of nihonium, moscovium, tennessine, and oganesson in 2016. Their importance is hard to overstate: collectively, they comprise a quarter of all known elements, and the names of several of them (uranium and plutonium) are widely known to the general public owing to the centrality of their roles in nuclear power generation and weaponry. Radioactivity and nuclear fission were first recognized in an actinide metal (uranium), key discoveries that altered our understanding of the very nature of a chemical element. Of course, many of these metals, including the late actinides and all of the transactinides, are charitably classified as exotics, unlikely ever to appear outside a research laboratory. Nevertheless, their study has refined our knowledge of the effects of relativity on chemical properties in general, and stimulated the development of methods for conducting experiments on an ultratrace scale, with elements that are produced only a few atoms at a time.

An area as extensive as this cannot be covered with justice in a single volume, so we have attempted to strike a balance between broad overviews and more focused topics. *The Heaviest Metals* begins with a counting of the fascinating (and convoluted) discovery of the actinides and transactinides, and then moves on to describe issues with the production of plutonium, the actinide synthesized in greatest quantity, and the complex problems involved in extracting and separating individual actinides. The next section looks at the electronic structure of the actinides and transactinides, chemical properties of the transactinides, and then the question of multiple bonding with these metals. Following this, the challenges in actinide crystallography are detailed, and the application of various spectroscopic techniques (solid-state NMR, Mössbauer, photoelectron, and laser-based spectroscopies) and computational investigations to characterize compounds of these elements are reviewed. The next section examines compounds with metals in the divalent (Th, U, Np, Pu) and pentavalent (U) oxidation states, areas of intense current research activity. The distinctive chemistries of actinide/group 15 complexes, metal borohydrides, supramolecular complexes, and polyoxometalates are also examined. Environmental and health issues are the focus of the next section, where questions of actinide speciation in freshwater and oceans, the biological transformations of actinide ions, and the movement of actinides in subsurface plumes are addressed. Separate chapters describe the emerging field of nuclear forensics, which tracks actinides to prevent theft or illegal disposal, and the multiple benefits provided by actinide radiotherapy. The volume closes with a look at two novel applications: actinide-based catalysis and the superconducting properties of actinide materials.

The Heaviest Metals was intended not only to inform, but also to inspire the reader to imagine new ways in which the elements at the frontier of the periodic table can advance multiple areas of chemistry. If it accomplishes that, our goal for the volume will have been reached.

Finally, we wish to thank the editorial staff at Wiley for their expert shepherding of the project from its earliest conception. Without their steadfast help, it could not have been completed.

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January 2019

IUPAC Periodic Table of the Elements

Key:

atomic number
Symbol
name
conventional atomic weight
standard atomic weight

1	2	3	4	5	6	7	8	9	10	11	12	13	14	15	16	17	18
1 **H** hydrogen 1.008 [1.0078, 1.0082]																	2 **He** helium 4.0026
3 **Li** lithium 6.94 [6.938, 6.997]	4 **Be** beryllium 9.0122											5 **B** boron 10.81 [10.806, 10.821]	6 **C** carbon 12.011 [12.009, 12.012]	7 **N** nitrogen 14.007 [14.006, 14.008]	8 **O** oxygen 15.999 [15.999, 16.000]	9 **F** fluorine 18.998	10 **Ne** neon 20.180
11 **Na** sodium 22.990	12 **Mg** magnesium 24.305 [24.304, 24.307]											13 **Al** aluminium 26.982	14 **Si** silicon 28.085 [28.084, 28.086]	15 **P** phosphorus 30.974	16 **S** sulfur 32.06 [32.059, 32.076]	17 **Cl** chlorine 35.45 [35.446, 35.457]	18 **Ar** argon 39.948
19 **K** potassium 39.098	20 **Ca** calcium 40.078(4)	21 **Sc** scandium 44.956	22 **Ti** titanium 47.867	23 **V** vanadium 50.942	24 **Cr** chromium 51.996	25 **Mn** manganese 54.938	26 **Fe** iron 55.845(2)	27 **Co** cobalt 58.933	28 **Ni** nickel 58.693	29 **Cu** copper 63.546(3)	30 **Zn** zinc 65.38(2)	31 **Ga** gallium 69.723	32 **Ge** germanium 72.630(8)	33 **As** arsenic 74.922	34 **Se** selenium 78.971(8)	35 **Br** bromine 79.904 [79.901, 79.907]	36 **Kr** krypton 83.798(2)
37 **Rb** rubidium 85.468	38 **Sr** strontium 87.62	39 **Y** yttrium 88.906	40 **Zr** zirconium 91.224(2)	41 **Nb** niobium 92.906	42 **Mo** molybdenum 95.95	43 **Tc** technetium	44 **Ru** ruthenium 101.07(2)	45 **Rh** rhodium 102.91	46 **Pd** palladium 106.42	47 **Ag** silver 107.87	48 **Cd** cadmium 112.41	49 **In** indium 114.82	50 **Sn** tin 118.71	51 **Sb** antimony 121.76	52 **Te** tellurium 127.60(3)	53 **I** iodine 126.90	54 **Xe** xenon 131.29
55 **Cs** caesium 132.91	56 **Ba** barium 137.33	57-71 lanthanoids	72 **Hf** hafnium 178.49(2)	73 **Ta** tantalum 180.95	74 **W** tungsten 183.84	75 **Re** rhenium 186.21	76 **Os** osmium 190.23(3)	77 **Ir** iridium 192.22	78 **Pt** platinum 195.08	79 **Au** gold 196.97	80 **Hg** mercury 200.59	81 **Tl** thallium 204.38 [204.38, 204.39]	82 **Pb** lead 207.2	83 **Bi** bismuth 208.98	84 **Po** polonium	85 **At** astatine	86 **Rn** radon
87 **Fr** francium	88 **Ra** radium	89-103 actinoids	104 **Rf** rutherfordium	105 **Db** dubnium	106 **Sg** seaborgium	107 **Bh** bohrium	108 **Hs** hassium	109 **Mt** meitnerium	110 **Ds** darmstadtium	111 **Rg** roentgenium	112 **Cn** copernicium	113 **Nh** nihonium	114 **Fl** flerovium	115 **Mc** moscovium	116 **Lv** livermorium	117 **Ts** tennessine	118 **Og** oganesson

57 **La** lanthanum 138.91	58 **Ce** cerium 140.12	59 **Pr** praseodymium 140.91	60 **Nd** neodymium 144.24	61 **Pm** promethium	62 **Sm** samarium 150.36(2)	63 **Eu** europium 151.96	64 **Gd** gadolinium 157.25(3)	65 **Tb** terbium 158.93	66 **Dy** dysprosium 162.50	67 **Ho** holmium 164.93	68 **Er** erbium 167.26	69 **Tm** thulium 168.93	70 **Yb** ytterbium 173.05	71 **Lu** lutetium 174.97
89 **Ac** actinium	90 **Th** thorium 232.04	91 **Pa** protactinium 231.04	92 **U** uranium 238.03	93 **Np** neptunium	94 **Pu** plutonium	95 **Am** americium	96 **Cm** curium	97 **Bk** berkelium	98 **Cf** californium	99 **Es** einsteinium	100 **Fm** fermium	101 **Md** mendelevium	102 **No** nobelium	103 **Lr** lawrencium

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For notes and updates to this table, see www.iupac.org. This version is dated 28 November 2016.

PART 1
Background

Discovery of the Actinide and Transactinide Elements

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1 INTRODUCTION

"Discovery is new beginning. It is the origin of new rules that supplement, or even supplant, the old … Were there rules for discovery, then discoveries would be mere conclusions."[1] The history of the discovery of the actinides and the transactinides, the 30 elements that comprise the very last part of the present periodic table of the elements, is peppered with rules: new rules, old rules transformed, new rules broken and remade—not necessarily by those doing the research, but often by Nature itself. Furthermore, if we consider the ways in which discoveries are made, they often fall into the categories of planned research, trial and error, or accidental discovery. Add to this a creative and

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observing mind[2] and you can encompass virtually all of the discoveries, and the methods used to further understand and gain more information about how the discovery can be exploited. It would be useful to analyze the following story for these characteristics, for this is the discovery that set in motion the train of events that would expand and change the periodic table forever.

In 1896, Henri Becquerel (1852–1908) reported that the double sulfate of potassium and uranium, formulated by him as [$SO_4(UO)K \cdot H_2O$] using the superscript notation common at the time, emitted radiation capable of penetrating light-opaque paper to expose silver salts. He realized that the so-called phosphorescent material was emitting this radiation by its very nature and not by becoming phosphorescent owing to exposure to light.[3] Subsequent work showed that the radiation could also penetrate thin sheets of aluminum and copper. Becquerel realized at this stage that the radiation was analogous to the newly discovered Roentgen rays.[4] Five additional notes in the same volume of the journal follow the course of his further experiments to show, beyond a doubt, that the radiation was spontaneous and owing to the uranium component of the salt. It was Marie Curie (1867–1934) who eventually named the new phenomenon "radioactivity."

Radioactivity, as evinced by the first actinide to be discovered, was to dominate the scientific, political, economic, and social scenes of the first half of the twentieth century. And during that century, all the rest of the actinides, and most of the transactinides, were to be discovered.

Using radioactivity as the signature by which radioactive atoms could be detected, scientists began to bombard targets with particles such as α-particles and neutrons as they became available, and then to identify the products of these reactions. They gradually surpassed the known limit of atomic number 92 to venture onto an unknown sea, not knowing where it would lead. So far, the journey has led to the discovery of 26 elements beyond uranium, completing the seventh row of the periodic table. This has involved massive amounts of funding, dedicated and persevering work on the part of genius-level individuals, and a surprising degree of international cooperation even during the Cold War. It has led to spectacular discoveries, overturned assumptions and theories, and glimpses of a Nature full of unexpected surprises.

2 WHAT ARE THE ACTINIDES AND THE TRANSACTINIDES?

A simple definition of the groups in question is: the elements beginning with actinium, with atomic number 89, and ending with the last element to be discovered and that completes period 7 of the periodic table, oganesson, with atomic number 118. None of these elements possesses a stable isotope; every actinide and transactinide is radioactive with half-lives that vary from billions of years, such as thorium, ^{232}Th, with a half-life of 1.41×10^{10} years, to μs, such as darmstadtium, ^{267}Ds, with a half-life of 3×10^{-6} s. Table 1 lists these 30 elements (occupying about 25% of the periodic table) in order of the atomic number. However, discovery chronology does not follow from this order.

The first actinide to be discovered, in 1789 by Martin Heinrich Klaproth (1743–1817), was uranium; a century later it was, as well, the first element recognized to be radioactive. Klaproth's alertness to detail accompanied by his pure love of science[5] no doubt prepared him to recognize a new substance when he dissolved the mineral pitchblende in nitric acid, and then neutralized the solution with strong base and observed the formation of a yellow precipitate. Using the tried and true method of heating the precipitate in the presence of a reducing agent, he obtained a black powder that he took for the element, which he named uranium in honor of the newly discovered planet, Uranus.[6]

A glance at Table 1 is quite informative regarding discovery. The first three actinides to be discovered were "lone wolf" affairs: a single discoverer is named, and that brings us to the end of the nineteenth century. It is an entirely different matter for the entire twentieth century: discovery is a team affair, often with long lists of multiple authors: we have entered the age of "big chemistry," characterized by specialized and expensive equipment in a national laboratory. It is easy to see that the Lawrence Berkeley National Laboratory, California, USA (LBNL) exercised a monopoly on actinide discoveries, completing the list with element number 103, lawrencium. Discoveries of the transactinides exhibit more international collaboration but, as we shall see, cooperation during the Cold War was never all sweetness and light.

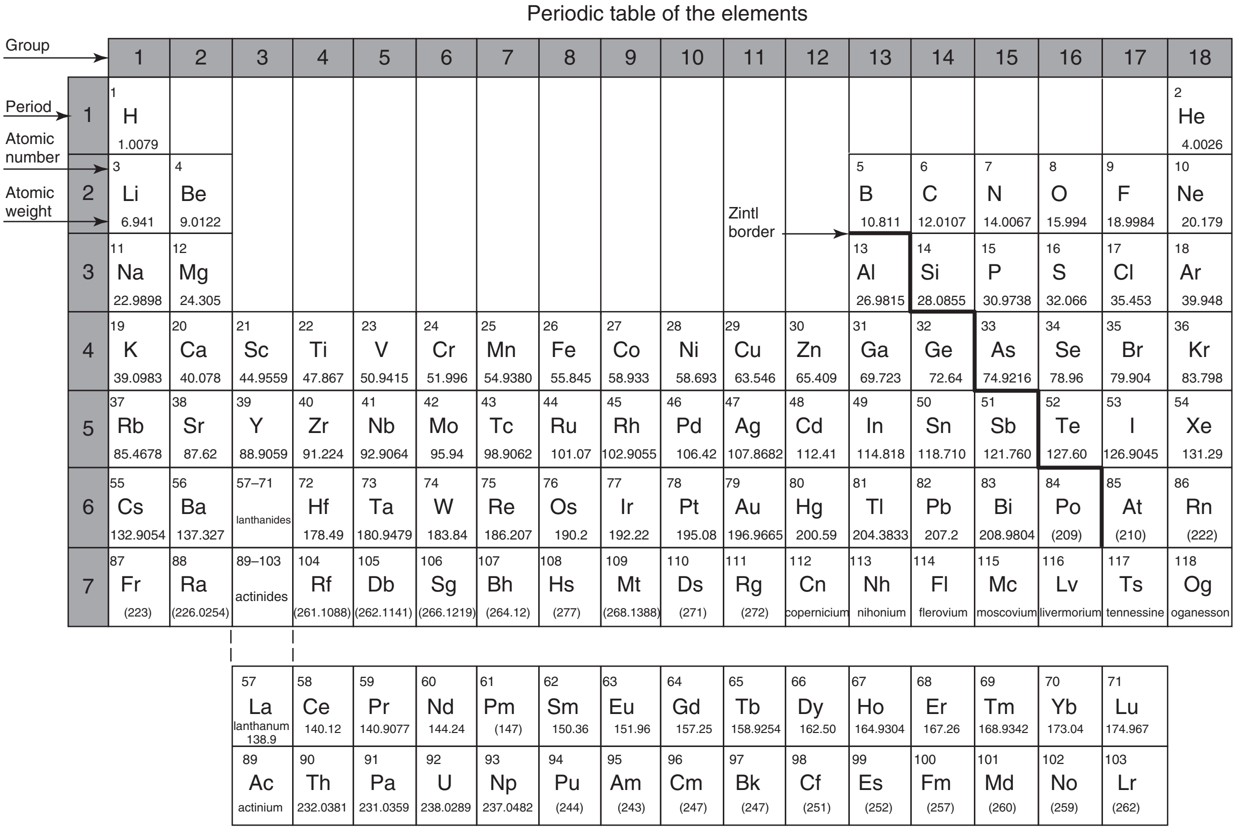
3 THE PLACE OF THE ACTINIDES AND TRANSACTINIDES IN THE PERIODIC TABLE

The modern periodic table is a grid consisting of 7 rows (periods) and 18 columns (groups). Periods 6 and 7 exceed the 18-column model with 32 groups each in the long form, and two offset rows of 15 elements each in the traditional, or medium-long, configuration, used for convenience so that the table will fit on a normal printed page, as shown in Figure 1.

The grid, originally arranged in order of increasing atomic weights of the elements, is now arranged in order of increasing atomic number (the number of protons in the nucleus of an atom, often abbreviated *Z*) in one dimension, and in order of similar chemical properties in the second dimension to form the groups. This grid actually defines the way electrons arrange themselves in atoms in terms of

Table 1 Discovery of the actinides and transactinides

Atomic number	Symbol	Name/symbol	Discoverer	Date of discovery	Place of discovery
89	Ac	Actinium	Debierne	1899	Paris, France
90	Th	Thorium	J. J. Berzelius	1829	Stockholm, Sweden
91	Pa	Protactinium	Hahn, Meitner	1917	Berlin, Germany
			Fajans		Karlsruhe
			Soddy, Cranston, Fleck		Glasgow, Scotland
92	U	Uranium	Martin Klaproth	1789	Berlin, Germany
93	Np	Neptunium	McMillan, Abelson	1940	LBNL, USA
94	Pu	Plutonium	Seaborg, Wahl, Kennedy	1940	LBNL, USA
95	Am	Americium	Seaborg, Morgan, James, Ghiorso	1944	LBNL, USA
96	Cm	Curium	Seaborg, James, Ghiorso	1944	LBNL, USA
97	Bk	Berkelium	Thompson, Ghiorso, Seaborg	1949	LBNL, USA
98	Cf	Californium	Thompson, Street, Ghiorso, Seaborg	1950	LBNL, USA
99	Es	Einsteinium	Choppin, Thompson, Ghiorso, Harvey	1952	LBNL, USA
100	Fm	Fermium	Choppin, Thompson, Ghiorso, Harvey	1952	LBNL, USA
101	Md	Mendelevium	Choppin, Thompson, Ghiorso, Harvey, Seaborg	1955	LBNL, USA
102	No	Nobelium	Flerov and others	1958	JINR, Russia
103	Lr	Lawrencium	Ghiorso, Larsh, Sikkeland, Latimer	1961	LBNL, USA; JINR, Russia
104	Rf	Rutherfordium	Ghiorso, Flerov	1964	LBNL, USA; JINR, Russia
105	Db	Dubnium	Various	1968	LBNL, USA; JINR, Russia
106	Sg	Seaborgium	Ghiorso and others	1974	LBNL, USA
107	Bh	Bohrium	Armbruster, Münzenberg, Hofmann, others	1981	GSI, Germany
108	Hs	Hassium	Armbruster, Münzenberg, Hofmann, others	1984	GSI, Germany
109	Mt	Meitnerium	Armbruster, Hofmann, Münzenberg, others	1982	GSI, Germany
110	Ds	Darmstadtium	Armbruster, Hofmann, others	1994	GSI, Germany
111	Rg	Roentgenium	Armbruster, Hofmann, others	1994	GSI, Germany
112	Cn	Copernicium	Hofmann, others	1996	GSI, Germany
113	Nh	Nihonium	Various	2004	RIKEN, Japan
114	Fl	Flerovium	Various	1999	LLNL, USA; JINR, Russia
115	Mc	Moscovium	Various	2010	LLNL, ORNL, USA; JINR, Russia
116	Lv	Livermorium	Various	2000	LLNL, USA; JINR, Russia
117	Ts	Tennessine	Various	2010	LLNL, ORNL, USA; JINR, Russia
118	Og	Oganesson	Various	2006	LLNL, USA; JINR, Russia



Periodic table of the elements

Period	1	2	3	4	5	6	7	8	9	10	11	12	13	14	15	16	17	18
1	1 H 1.0079																	2 He 4.0026
2	3 Li 6.941	4 Be 9.0122											5 B 10.811	6 C 12.0107	7 N 14.0067	8 O 15.994	9 F 18.9984	10 Ne 20.179
3	11 Na 22.9898	12 Mg 24.305											13 Al 26.9815	14 Si 28.0855	15 P 30.9738	16 S 32.066	17 Cl 35.453	18 Ar 39.948
4	19 K 39.0983	20 Ca 40.078	21 Sc 44.9559	22 Ti 47.867	23 V 50.9415	24 Cr 51.996	25 Mn 54.9380	26 Fe 55.845	27 Co 58.933	28 Ni 58.693	29 Cu 63.546	30 Zn 65.409	31 Ga 69.723	32 Ge 72.64	33 As 74.9216	34 Se 78.96	35 Br 79.904	36 Kr 83.798
5	37 Rb 85.4678	38 Sr 87.62	39 Y 88.9059	40 Zr 91.224	41 Nb 92.9064	42 Mo 95.94	43 Tc 98.9062	44 Ru 101.07	45 Rh 102.9055	46 Pd 106.42	47 Ag 107.8682	48 Cd 112.41	49 In 114.818	50 Sn 118.710	51 Sb 121.760	52 Te 127.60	53 I 126.9045	54 Xe 131.29
6	55 Cs 132.9054	56 Ba 137.327	57–71 lanthanides	72 Hf 178.49	73 Ta 180.9479	74 W 183.84	75 Re 186.207	76 Os 190.2	77 Ir 192.22	78 Pt 195.08	79 Au 196.9665	80 Hg 200.59	81 Tl 204.3833	82 Pb 207.2	83 Bi 208.9804	84 Po (209)	85 At (210)	86 Rn (222)
7	87 Fr (223)	88 Ra (226.0254)	89–103 actinides	104 Rf (261.1088)	105 Db (262.1141)	106 Sg (266.1219)	107 Bh (264.12)	108 Hs (277)	109 Mt (268.1388)	110 Ds (271)	111 Rg (272)	112 Cn copernicium	113 Nh nihonium	114 Fl flerovium	115 Mc moscovium	116 Lv livermorium	117 Ts tennessine	118 Og oganesson

57 La lanthanum 138.9	58 Ce 140.12	59 Pr 140.9077	60 Nd 144.24	61 Pm (147)	62 Sm 150.36	63 Eu 151.96	64 Gd 157.25	65 Tb 158.9254	66 Dy 162.50	67 Ho 164.9304	68 Er 167.26	69 Tm 168.9342	70 Yb 173.04	71 Lu 174.967
89 Ac actinium	90 Th 232.0381	91 Pa 231.0359	92 U 238.0289	93 Np 237.0482	94 Pu (244)	95 Am (243)	96 Cm (247)	97 Bk (247)	98 Cf (251)	99 Es (252)	100 Fm (257)	101 Md (260)	102 No (259)	103 Lr (262)

Figure 1 The standard medium-long form of the periodic table

principal energy levels and sublevels that they occupy, the so-called s, p, d, and f blocks. Not only has it brought order out of the chaos of so many elements with so many different properties, but it also functions as a theoretical tool, a "marvelous map of the whole geography of the elements."[7]

The two rows offset as "footnotes" from the main body of the periodic table, each consisting of 15 elements. The top row, from lanthanum ($Z = 57$) to lutetium ($Z = 71$), along with two elements in the main body of the table, scandium and yttrium, are termed the "rare earths." The 15 rare earths in the offset sit below yttrium with properties so similar to one another that the Czech chemist, Bohuslav Brauner (1855–1935), once proposed that they should all occupy the same space.[8]

Today, we take the placement of the actinides in the table for granted. However, initially, the first-discovered members of this group were placed in the main body of the table with actinium in the yttrium group, thorium under hafnium, protactinium under tantalum, and uranium under tungsten. Any transuranium elements to be yet discovered were expected to fall into place to complete period 6, with the last element in the row, $Z = 104$, fitting under radon.

The differences in chemical properties between some of these supposed homologs soon made this assumption untenable. In 1893, Henry Bassett (ca. 1837–1920),[9] followed by Alfred Werner (1866–1919), who is often given the credit, first suggested that the heavier elements beyond uranium might need an intergroup accommodation similar to that of the rare earths.[10] Decades later, in 1940, when Edwin McMillan (1907–1991) and Philip Abelson (1913–2004) discovered element 93, and shortly afterward, Glenn Seaborg (1912–1999) and his team discovered element 94, they had a surprise waiting. Chemical tests revealed that the properties of both new elements were more similar to those of uranium than to their supposed homologs, rhenium and osmium.[11] At this point in the group's struggle to place the new elements in the periodic table, its extreme utility became spectacularly evident as both a flexible and a predictive theoretical tool. Seaborg took up the Bassett-Werner idea and made it his own:

> I began to believe it was correct to propose a second lanthanide-style series of elements …[starting]… with element number 89, actinium, the element directly below lanthanum in the periodic table. Perhaps there was another inner electron shell being filled. This would make the series directly analogous to the lanthanides, which would make sense, but it would require a radical change in the periodic

> table … [I was told] that such an outlandish proposal would ruin my scientific reputation. Fortunately, that was no deterrent because at the time I had no scientific reputation to lose.[12]

So, the initial stages of discovery of the transuranium elements gave rise to a reconfiguration of the periodic table. The two new elements were appropriately named neptunium and plutonium after the two planets that lay beyond Uranus in the solar system. The rest of the actinides, as they were discovered, fell right into place under their rare earth homologs, and the transactinides, from atomic numbers 104 to 118 populated period 7 to its completion. It remains to be seen how the future treats the superactinides beginning with atomic number 119.[a]

4 THE PRE-URANIUM ACTINIDES: ACTINIUM, PROTACTINIUM, AND THORIUM

4.1 The Discovery of Thorium

Element number 90, thorium, was the first of this trio to be discovered in 1829. One of the most famous chemists of the time, Jöns Jacob Berzelius (1779–1848), Professor at the Karolinska University, Stockholm, in examining a curious mineral sent to him by Jens Esmark (1763–1839), a Norwegian mineralogist, thought that he could discern the presence of a new element. He isolated the impure metal by reducing its fluoride salt with elemental potassium, and named it thorium, after the Scandinavian god, Thor. The mineral was subsequently called thorite.[14] In 1898, working independently, Marie Curie and Gerhard C. Schmidt (1865–1949) reported almost simultaneously that thorium, such as uranium, was radioactive.[15,16]

4.2 The Discovery of Actinium

Seventy years were to pass before the announcement of the discovery of actinium ($Z = 89$), the element that gives its name to the entire actinide series.[17] Parisian André-Louis Debierne (1874–1949) began his studies at the École de Physique et de Chemie and began to study mineral chemistry following the death of his mentor, Charles Friedel (1832–1899). Welcomed into the Curies' laboratory, he began to treat the enormous quantities of pitchblende they supplied to him until he soon discovered a new element; he was one of the youngest chemists ever to do so.[18] He called it actinium from the Ancient Greek word, *aktinos*, meaning beam or ray.

The year 1913 was a landmark one for science: in that year H. G. J. Moseley (1887–1915) conferred a number and identity on every atom by reason of its number of nuclear protons, and Frederick Soddy (1877–1956) discovered isotopes, atoms with differing neutron numbers in atoms with like atomic numbers. He also formulated the law of chemical displacement: α-emitters produce a daughter product two atomic numbers lower and β-emitters one atomic number higher. Moseley's work defined the list of elements still missing in the periodic table, namely elements 43, 61, 72, 75, 85, 87, and 91.[19] Soddy's work solved the puzzle of the myriad of new "elements" spawned by radioactive decay and his chemical displacement law had predictive properties. All of these facts figured weightily in the discovery of protactinium over the period from 1913 to 1917.

4.3 The Discovery of Protactinium

The hunt was now on for the missing element 91. Kasimir Fajans (1887–1975) and Ostwald Helmuth Göhring (b. 1889) took up the challenge. Fajans was the first to succeed in deciphering the radioactive decay cascade of ^{238}U as the following:

$$\mathrm{U_1} \xrightarrow{\alpha} \mathrm{UX_1} \xrightarrow{\beta\text{-}} \mathrm{UX_2} \xrightarrow{\beta\text{-}} \mathrm{U_{II}} \xrightarrow{\alpha} \mathrm{Io} \tag{1}$$

which translates in modern terminology to:

$$^{238}\mathrm{U} \xrightarrow{\alpha} {}^{234}\mathrm{Th} \xrightarrow{\beta\text{-}} {}^{234}\mathrm{Pa^m} \xrightarrow{\beta\text{-}} {}^{234}\mathrm{U} \xrightarrow{\alpha} {}^{230}\mathrm{Th} \tag{2}$$

They found that the substance $\mathrm{UX_2}$, a β-emitter with a very short half-life of about 1 min, did not correspond to any radioisotope already known, realizing that it should occupy a vacant space in the periodic table. Owing to its short half-life, they named this new element *brevium*.

Soon after Fajans's announcement, Otto Hahn (1879–1968) and Lise Meitner (1878–1968), working in Berlin, began to search for longer-lived isotopes of the same element. Hampered by the outbreak of World War I, especially by Hahn's conscription, Meitner carried on alone with a miniscule sample (21 g) of pitchblende, doing preliminary separations. It was only a year later that she received a kilogram sample of radioactive salts from which she was able to isolate an isotope of element ^{231}91 with a half-life of about 32 700 years.[20] They named it protoactinium (later changed to protactinium by IUPAC (International Union of Pure and Applied Chemistry) in 1949), recognizing it as the mother substance of actinium.[b]

In June of that same year, Frederick Soddy and his young student, John Arnold Cranston (1891–1972), published the results[22] of their heat treatments of pitchblende that yielded small sublimated amounts of protactinium for which they were unable to characterize the decay scheme. Obviously, the case of protactinium, tangled by a publication that appeared 5 years before that of Hahn and Meitner, as well as a new claim in the same year, became even more so. Eventually, the priority was awarded to the team that had discovered the longest-lived isotope, Hahn and Meitner, but not without dealing delicately with the aggressive character and imperious temperament of Kasimir Fajans. Cranston and Soddy, having published their papers 3 months after those of Hahn and Meitner, immediately recognized their priority.[23,24]

This little protactinium story was told at some length because it presages the multiple contentious priority disputes to follow: who gets the recognition for the discovery, and who gets to name the new element? The naming, in the end, came to be the most controversial issue, for as paleobotanist Hope Jahren (b. 1969) observes:

> The scientific rights to naming a new species, a new mineral, a new atomic particle, a new compound, or a new galaxy are considered the highest honor and the grandest task to which any scientist may aspire.[25]

5 DISCOVERY OF URANIUM FISSION

5.1 Enrico Fermi's Neutron Bombardment Experiments

The facts that uranium was discovered in 1789 and its radioactivity was recognized in 1896 seem to be almost trivial in light of the shattering discovery of its most important, and most all-encompassing property: its ability to undergo nuclear fission with the consequent release of immense amounts of energy. This property was undreamed of, and in fact dismissed, when, in April of 1934, Enrico Fermi (1901–1954) and his team, the legendary "Ragazzi di via Panisperna," began to bombard uranium with neutrons. Fermi, convinced that knowledge of the atom was in large part complete, decided to investigate the properties of the atomic nucleus. He was one of the first to recognize the tremendous importance of artificial radioactivity, discovered by Frédéric Joliot (1900–1958) and Irène Joliot-Curie (1897–1956), and for which they received the Nobel Prize in Chemistry in 1935.[26] Not possessing a cyclotron, and therefore lacking sufficient irradiated material, he decided to attack the atom with neutrons, discovered only 2 years previously in 1932 by James Chadwick (1891–1974), instead of with α-particles. As neutrons had no electric charge, Fermi reasoned that they would not be repulsed by the nuclear charge and might easily penetrate the nucleus itself. But, as neutrons are not spontaneously emitted by radioactive isotopes, he had to obtain them by bombarding lighter elements, such as beryllium, with α-particles emitted by natural substances, such as radium. The neutron yield was low: just one per every 100 000 α-particles emitted, but undeterred, Fermi personally built the detectors necessary for counting atomic disintegrations. Success came only when, after bombarding all the lighter elements, fluorine and aluminum exhibited neutron-induced radiation.[27] After that, the list of nuclei susceptible to neutron irradiation grew.[28–30] Seven months later, in October, Fermi announced a second crucial discovery: the braking effect of hydrogenous substances, such as water, on the radioactivity induced by neutrons. This amounted to the first step toward the utilization of nuclear energy.

Meanwhile at Rome, Fermi procured a very precious treasure, 1.6 g of radium chloride from which he could extract *emanation* (or radon) that would be necessary for the production of neutrons. Further work by Fermi and his team led to two seemingly new elements[31] with atomic numbers 93 and 94 owing to neutron absorption by ^{238}U, and subsequent double-β-emission according to the following schemes:

$$^{239}\mathrm{U} \rightarrow {}^{239}93 + \beta^- \rightarrow {}^{239}94 + \beta^- \qquad (3)$$

Radiochemical tests showed that the activity of ^{239}U produced particles with properties that did not belong to any elements that preceded them in the periodic table. Believed to be eka-rhenium and eka-iridium, they were placed in period 7 of the table.

5.2 Criticism of the Fermi Group's Interpretation of Results

The Fermi group's announcement raised sharp criticism in scientific circles. In addition to the two "transuranic elements" they thought they had identified, they had found a good half-dozen others with a variety of chemical properties difficult to place in the periodic table as they had to be untangled from uranium's ongoing normal decay producing its own short-lived daughter products.[32] In fact, a chemist at the University of Fribourg, Ida Tacke Noddack (1896–1978), criticized Fermi's experimental judgment in only searching for elements in the neighborhood of element 92. She said that all elements should be searched for, even lighter ones. She did not hesitate to declare that she strongly doubted that the products Fermi identified were transuranium elements, but suggested nuclear fission instead.[33] Her idea was unacceptable in the physics world, deemed highly speculative and lacking a theoretical basis. "Everyone knew" that atoms just did not fly apart in such a manner!

Things remained unresolved. A year later, Otto Hahn and Lise Meitner repeated Fermi's experiments using better facilities and they confirmed Fermi's results. Furthermore, according to them, they were also able to observe traces of elements 95, 96, and 97 that they provisionally called *eka-iridium*, *eka-platinum*, and *eka-aurum*.[34] However, as time went on, Irène Joliot-Curie and her Yugoslav coworker, Pavel Savitch, published some papers documenting their concentration on only one of the products of neutron irradiation, that with a half-life of 3.5 h, and after a few false starts conclusively stated that the product in question strangely resembled lanthanum, an already known element lodged in the middle of the periodic table. However, they never declared that they had actually found lanthanum, only a possible transuranic element that resembled lanthanum![35,36] They could not imagine that they actually had lanthanum. Reality was hidden in plain sight!

5.3 Fission at Last!

The last of these papers made Hahn sit up and take notice: perhaps the almost forgotten suggestion by Ida Noddack was right after all. So later in 1938, after more experimentation and rethinking, Hahn and his colleague Fritz Strassmann (1902–1980) finally admitted that, as chemists, they realized they were dealing with radiobarium and radiolanthanum, but as physicists they added, "we cannot bring ourselves to take such a drastic step, which goes against all previous laws (a word that Hahn later changed to 'experiences') of nuclear physics."[37]

Hahn communicated his conclusions by letter to Lise Meitner who was in exile in Sweden, fleeing the Nazi racial persecution, and she, with her nephew Otto Frisch (1904–1979), in their famous walk in the woods, worked out a theory whereby the positive charge of the uranium nucleus was large enough to overcome the effect of the nuclear surface tension almost completely, allowing the nucleus to fall apart at the slightest provocation. They also worked out the fact that the mass loss on nuclear division would be about one-fifth the mass of a proton, exactly equivalent to the correct and enormous energy predicted by Albert Einstein's (1879–1955) relationship, $E = mc^2$.[38,39]

Meanwhile, Enrico Fermi had already received his Nobel Prize in Physics for 1938, awarded *for his demonstrations of the existence of new radioactive elements produced by neutron irradiation, and for his related discovery of nuclear reactions brought about by slow neutrons.*[c.] The citation is very cautious in using the words "new radioactive elements," initially interpreted erroneously by Fermi as transuranium elements. But, in light of subsequent interpretations, he had actually discovered nuclear fission without knowing it, and actually produced new radioactive isotopes of elements previously known!

5.4 The Impact of Uranium Fission on the Modern World

By the irony of fate (or, some would say, of blindness), Enrico Fermi, in looking for transuranium elements, found nuclear fission. At about the same time, physicist Paul Scherrer (1890–1969), working in Zurich, had an even closer encounter with fission. "He bombarded thorium … with neutrons and saw the fission fragments that Meitner and Frisch had identified. But Scherrer wouldn't believe his eyes. He thought his Geiger counter was malfunctioning. What wasn't expected wasn't seen."[d.] Fermi, working in Fascist Rome in 1933, or Scherrer working in Switzerland, could have handed (or have seized from them) the information the Nazis would need to build a super-weapon 6 years earlier than the actual recognition of fission and its potential had they realized the evidence that was right before their eyes. Their "slight oversights" had a profound and beneficial effect on the rest of the world.

When word of the reality of nuclear fission broke upon the world, Niels Bohr (1885–1962) in Copenhagen struck his head with his fist and exclaimed. "Oh, what fools we were that we did not see this before." And in Paris, Irène Joliot-Curie cried out, "What fools we were!"

In 1941, just 2 years after the discovery of fission, Hans von Halban (1908–1964) and Lew Kowarski (1907–1979), two French exiles from the Curie Institute working in Cambridge but under the mentorship of Frédéric Joliot in France, were the first to establish that it was possible to sustain a chain reaction starting with natural uranium.[e.] Simultaneously, two other Cambridge physicists, Norman Feather (1904–1978) and Egon Bretscher (1901–1973), hypothesized that the chain reaction could have military applications. By now, it was recognized that the fissionable nucleus was the ^{235}U isotope of element 92, only seven parts in 1000 in naturally occurring uranium. They also hypothesized that the more abundant isotope, ^{238}U, could be transmuted by neutron absorption into a new, hitherto unknown, element which would not only be fissionable, but would also have a long half-life according to a pathway almost identical to Equation 3:

$$^{238}\text{U} + \text{n} \rightarrow {}^{239}\text{U} \rightarrow {}^{239}93 + \beta^- \rightarrow {}^{239}94 + \beta^- \quad (4)$$

What would follow from these discoveries was an international race for the ultimate weapon carried on in wartime under the shroud of utmost secrecy. Although research on the peaceful uses of atomic energy was also on the docket, it had low priority when it came to building the atom bomb. Heavy water, deemed essential for the propagation of a chain reaction owing to its moderating (slowing down) properties on neutrons, was in short supply. The largest production plant, Norsk Hydro, was in the hands of Nazi Germany. Although many top scientists abhorred the idea of such a weapon, the Allied governments knew that they could not allow Germany to beat them in the race and use this weapon for world domination. As Frederick Soddy remarked presciently in 1904: "The man who put his hand on the lever by which a parsimonious nature regulates so jealously the output of this store of energy would possess a weapon by which he could destroy the earth if he chose."[f.]

6 THE BERKELEY HEGEMONY

To understand how the University of California at Berkeley eventually became the epicenter of the discovery