

## PRODUCTION AND CHEMISTRY OF TRANSACTINIDE ELEMENTS

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### Summary

Remarkable progress in synthesizing new transactinide elements and in studying chemical properties of those elements has been achieved in the last decade. This article gives a brief summary of the reported syntheses and nuclear properties of the

transactinide elements as well as chemical investigation of those elements. Experimental techniques of single atom detection after in-flight separation with electromagnetic separators have made a breakthrough in production and identification of new transactinide nuclides. Development of automated rapid chemical separation techniques based on one atom-at-a-time scale has also considerably contributed to the progress of chemical studies of the transactinide elements. Some key experiments exploring new frontiers of the production and chemical characterization of the transactinide elements as well as the state of the art in these experimental studies are demonstrated. Prospects of extending nuclear and chemical studies of the heaviest elements in near future are shortly considered.

## 1. Introduction

Presently, we know more than 20 artificial transuranium elements. According to the actinide concept, the 5f electron series ends with element 103, lawrencium (Lr), and a new 6d electron transition series is expected to begin with element 104, rutherfordium (Rf). The elements with atomic numbers  $Z \geq 104$  are called transactinide elements. The periodic table of the elements is shown in Figure 1. The currently known transactinide elements, elements 104 through 112, are placed in the periodic table under their respective lighter homologues in the 5d electron series, hafnium (Hf) to mercury (Hg). Elements from 113 to 118 with the exception of 117 synthesized would be in the successive 7p electron series, although the discoveries of elements with  $Z \geq 112$  are still waiting to be confirmed.

	1	2	3	4	5	6	7	8	9	10	11	12	13	14	15	16	17	18
1	1 H																	2 He
2	3 Li	4 Be											5 B	6 C	7 N	8 O	9 F	10 Ne
3	11 Na	12 Mg											13 Al	14 Si	15 P	16 S	17 Cl	18 Ar
4	19 K	20 Ca	21 Sc	22 Ti	23 V	24 Cr	25 Mn	26 Fe	27 Co	28 Ni	29 Cu	30 Zn	31 Ga	32 Ge	33 As	34 Se	35 Br	36 Kr
5	37 Rb	38 Sr	39 Y	40 Zr	41 Nb	42 Mo	43 Tc	44 Ru	45 Rh	46 Pd	47 Ag	48 Cd	49 In	50 Sn	51 Sb	52 Te	53 I	54 Xe
6	55 Cs	56 Ba	57 La	72 Hf	73 Ta	74 W	75 Re	76 Os	77 Ir	78 Pt	79 Au	80 Hg	81 Tl	82 Pb	83 Bi	84 Po	85 At	86 Rn
			⋮															
			Lanthanide	57 La	58 Ce	59 Pr	60 Nd	61 Pm	62 Sm	63 Eu	64 Gd	65 Tb	66 Dy	67 Ho	68 Er	69 Tm	70 Yb	71 Lu
			⋮															
			Actinide	89 Ac	90 Th	91 Pa	92 U	93 Np	94 Pu	95 Am	96 Cm	97 Bk	98 Cf	99 Es	100 Fm	101 Md	102 No	103 Lr
			⋮															
7	87 Fr	88 Ra	89 Ac	104 Rf	105 Db	106 Sg	107 Bh	108 Hs	109 Mt	110 Ds	111 Rg	112	113	114	115	116		118

Figure 1: Periodic table of the elements (2007).

Searching for and producing new elements are very challenging subjects in recent advanced nuclear and radiochemistry. How many chemical elements may be synthesized on earth? How can they be produced? How long can they survive? Which properties do determine their stability? What are their chemical and physical properties? And how are the orbital electron configurations affected in the strong electric field of heavy atoms? These are the most fundamental questions in science.

One of the most fundamental properties of the transactinide elements (nuclei) is their relatively high stability. According to the theoretical nuclear models including nuclear shell structure, the existence of an enhanced stability in the region of nuclei with  $Z = 114$  (or possibly 120, 122, or 126) and neutron number  $N = 184$  has been predicted as the next doubly magic spherical nucleus beyond  $^{208}\text{Pb}$  ( $Z = 82$ ,  $N = 126$ ); the so-called island of stability surrounded by a sea of instability has been expected there. The recent theoretical calculations predict another stabilized region of the deformed nuclei at around  $Z = 108$  and  $N = 162$  due to hexadecapole deformation in the ground state. A most recent experiment has produced and identified the nucleus  $^{270}\text{Hs}$ , hassium (Hs), with  $Z = 108$  and  $N = 162$  and evaluated its half-life of 22 s that is remarkably long for a transactinide nuclide. The measured  $\alpha$ -decaying particle energy ( $Q_\alpha$  value) of  $^{270}\text{Hs}$  well fits with theoretical estimates, providing direct evidence for this new island of stability. This region may constitute a bridge or reef toward the expected island of around  $Z = 114$  and  $N = 184$ .

Studies of chemical properties of the newly-synthesized transactinide elements are extremely interesting and challenging subjects in modern nuclear and radiochemistry. One of the most important questions is to clarify the position of the transactinide elements in the periodic table. It is also of special interest to assess the magnitude of the influence of relativistic effects (see: *Radiochemistry and Nuclear Chemistry*, Appendix 2) on chemical properties. According to the calculations of electron configurations of heavy atoms, it is predicted that sudden changes in the structure of electron shells may appear due to relativistic effects which originate from the increasingly strong Coulomb field of a highly charged atomic nucleus. Therefore, it is expected that heavier elements show a drastic rearrangement of electron configurations in their atomic ground states, and as electron configurations are responsible for chemical behavior of elements, such relativistic effects can lead to unexpected chemical properties. Increasing deviations from the periodicity of chemical properties based on extrapolation from lighter homologues in the periodic table are consequently predicted. It would be no longer possible to deduce detailed chemical properties of the transactinide elements simply from the position in the periodic table. The aim of chemical study of the transactinide elements is to explore the new frontiers of inorganic chemistry, i.e., the chemistry of the elements in the 7th period.

Over the past decades, there has been a great progress in experimental investigation of the chemical properties of the transactinide elements as well as in the synthesis of heavier elements extending the periodic table still further and the chart of nuclides toward higher  $Z$  and  $N$ . After a short summary of the discovery and synthesis of the transactinide elements, the present article highlights some recent topical works on the study of production and chemistry of the transactinide elements and provides a brief outlook for future developments.

The elements heavier than fermium (Fm) with  $Z = 100$  cannot be produced by neutron capture reactions even at high flux nuclear reactors and must be made at accelerators using heavy-ion-induced reactions with the rate of an atom at a time. Thus, the elements heavier than Fm are often called “the heaviest elements”. Their chemistry has also to be explored with one atom at a time. In chemical aspects, the transactinide elements with  $Z = 104–112$  are clearly characterized as the 6d transition elements beyond the actinide series (see: *Chemistry of the Actinide Elements*). From the nuclear point of view, however, there is no definite border between elements 103 and 104. Thus, some important and related topical results on nuclear properties of heavy nuclei with  $Z \geq 100$  are contained in this article.

## 2. Brief History of Discovery

The discovery and synthesis of transactinide elements reported are summarized in Table 1. The names and symbols up to element 111, roentgenium (Rg), are approved by International Union of Pure and Applied Chemistry (IUPAC) based on the reports of the Transfermium Working Group (TWG) and Joint Working Party (JWP) that consist of scientists appointed by both IUPAC and International Union of Pure and Applied Physics (IUPAP).

In the discovery of transactinide elements, special experimental techniques, such as a parent-daughter  $\alpha$ - $\alpha$  correlation technique, were developed to identify both atomic number and mass number of transactinide nuclides, because half-lives of transactinides produced were too short and the number of atoms produced is too small to allow any chemical separation and the ordinary identification method of  $Z$ . The  $\alpha$ -decay of an unknown new species was measured and was correlated in time with the  $\alpha$ -decay of a known daughter nuclide, thus establishing their genetic relation. Additional  $\alpha$  correlations with subsequent decay of granddaughter or even great-granddaughter nuclides made possible the definite identification of the synthesized nuclide. The method requires rather sophisticated detection and timing techniques, but it provides unequivocal identification because the parent nuclide has to be one helium atom heavier than its known daughter nuclide. This technique was pioneered by Ghiorso and coworkers at Lawrence Berkeley Laboratory (LBL), and they discovered Rf to seaborgium (Sg). It was also used in the discoveries of the nuclides beyond bohrium (Bh), each with half-lives of only a microsecond to a few seconds, made by Gesellschaft für Schwerionenforschung (GSI), Germany; Flerov Laboratory for Nuclear Reactions (FLNR), Dubna, Russia; and The Institute of Physical and Chemical Research (RIKEN), Japan.

Atomic number	Element*	Symbol	Year of discovery	Institute	Production reaction
104	Rutherfordium	Rf	1969	LBL <sup>a)</sup>	$^{249}\text{Cf}(^{12}\text{C}, 4\text{n})^{257}\text{Rf}$
105	Dubnium	Db	1970	LBL	$^{249}\text{Cf}(^{15}\text{N}, 4\text{n})^{260}\text{Db}$
			1971	FLNR <sup>b)</sup>	$^{243}\text{Am}(^{22}\text{Ne}, 4\text{n})^{261}\text{Db}$
106	Seaborgium	Sg	1974	LBL	$^{249}\text{Cf}(^{18}\text{O}, 4\text{n})^{263}\text{Sg}$
107	Bohrium	Bh	1981	GSI <sup>c)</sup>	$^{209}\text{Bi}(^{54}\text{Cr}, \text{n})^{262}\text{Bh}$
108	Hassium	Hs	1984	GSI	$^{208}\text{Pb}(^{58}\text{Fe}, \text{n})^{265}\text{Hs}$
109	Meitnerium	Mt	1982	GSI	$^{209}\text{Bi}(^{58}\text{Fe}, \text{n})^{266}\text{Mt}$
110	Darmstadtium	Ds	1995	GSI	$^{208}\text{Pb}(^{62}\text{Ni}, \text{n})^{269}\text{Ds}$
111	Roentgenium	Rg	1995	GSI	$^{209}\text{Bi}(^{64}\text{Ni}, \text{n})^{272}\text{Rg}$

112			1996	GSI	$^{208}\text{Pb}(^{70}\text{Zn}, n)^{277}112$
113			2004	FLNR	$^{243}\text{Am}(^{48}\text{Ca}, 3n)^{288}115 \rightarrow ^{284}113$
			2004	RIKEN <sup>d)</sup>	$^{209}\text{Bi}(^{70}\text{Zn}, n)^{278}113$
114			1999	FLNR	$^{244}\text{Pu}(^{48}\text{Ca}, 3n)^{289}114$
115			2004	FLNR	$^{243}\text{Am}(^{48}\text{Ca}, 3n)^{288}115$
116			2000	FLNR	$^{248}\text{Cm}(^{48}\text{Ca}, 4n)^{292}116$
118			2006	FLNR	$^{249}\text{Cf}(^{48}\text{Ca}, 3n)^{294}118$

\* As of 2007, the elements beyond  $Z = 111$  are not yet approved by IUPAC.

- a) Lawrence Berkeley Laboratory, presently Lawrence Berkeley National Laboratory (LBNL)
- b) Flerov Laboratory of Nuclear Reactions
- c) Gesellschaft für Schwerionenforschung
- d) The Institute of Physical and Chemical Research

Table 1: Discovery of transactinide elements.

The elements Rf to Sg as shown in Table 1 were produced using the so-called hot fusion reaction, based on actinide targets with light heavy-ion beams. The reaction products, recoiling from the target due to the momentum transfer from the projectiles were stopped in a gas cell and then transported with a gas-jet transport system to a measurement system where their parent-daughter  $\alpha$ - $\alpha$  correlations were observed. To access the new region beyond Bh whose expected half-lives and production cross sections drop still further, another type of reaction, the so-called cold fusion reaction using target nuclei near the doubly magic  $^{208}\text{Pb}$  with appropriate neutron-rich heavy-ion beams, and fast and sensitive detection methods allowing the identification of new nuclides or even new elements by the observation of single atoms were developed. The fast single-atom detection method consists of in-flight separation using an optical recoil mass separator and the above parent-daughter  $\alpha$ - $\alpha$  correlation technique. To approach to the further neutron-rich region where the island of stability is expected around  $N = 184$ , the FLNR group is now performing an extended series of experiments bombarding actinide targets with the neutron-rich  $^{48}\text{Ca}$  beam. The discoveries of elements 112 through 118 except for element 117 have been claimed. Unfortunately, the experiments at FLNR suffer a disadvantage in that the observed nuclear decays are not genetically linked to the region of known nuclei.

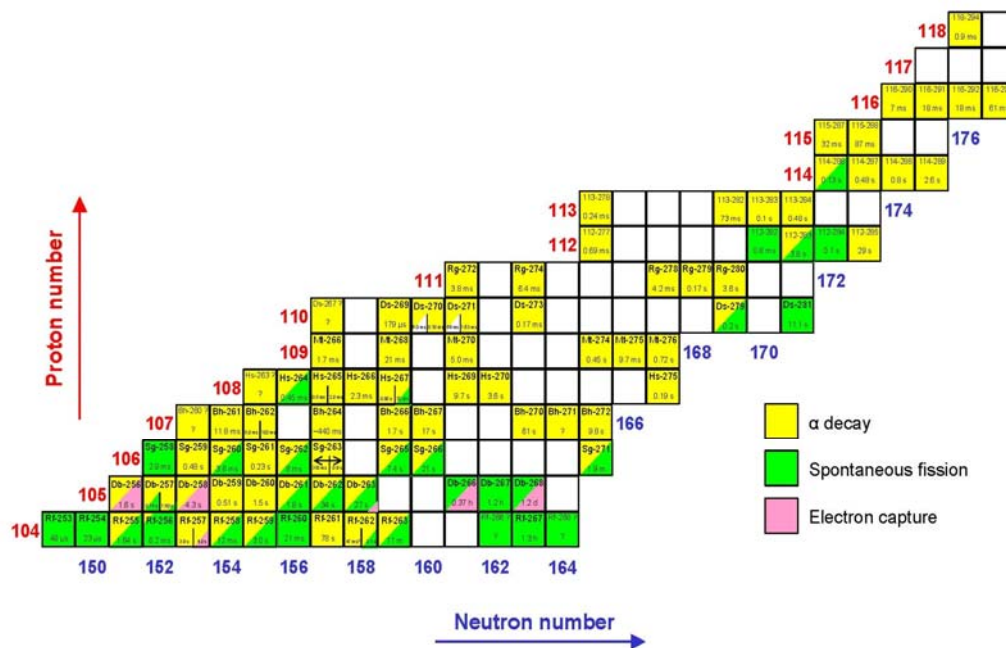


Figure 2: Chart of the transactinide nuclides. Decay modes are represented by specific colors.

The criteria that must be satisfied for the discovery of a new chemical element to be recognized, established by TWG and JWP, requires the data with a high degree of internal redundancy and of the highest quality and also demands clear observations of known descendent or production of previously unknown intermediates through cross bombardments. The claims for the discoveries of the elements 112, 113, 114, 115, 116, and 118 have been submitted to IUPAC for the official approval of their discovery.

Figure 2 shows the chart of the transactinide nuclides (Magill *et al.* 2006 and Oganessian 2007). The data obtained at FLNR with the  $^{48}\text{Ca}$ -induced reactions are located in the neutron-rich region, while the nuclides synthesized by cold fusion reactions are in the neutron-deficient region in each  $Z$ .

### 3. Production and Nuclear Decay Studies of Transactinides

#### 3.1. Heavy-ion Fusion Reaction

The only successful method so far to produce transactinides is the heavy-ion-induced complete fusion reactions. (Recently, the production of long-lived neutron-rich transactinides in deep inelastic collisions of transuranium nuclei is theoretically studied within the dynamical model based on multidimensional Langevin equations.) The minimum projectile energy required to overcome the Coulomb barrier in these reactions generally exceeds the reaction  $Q$ -value (see: *Radiochemistry and Nuclear Chemistry*) necessary for the compound nucleus formation. Successful production of highly fissionable heavy nuclei mainly depends on two factors: fusion cross section  $\sigma_{\text{fusion}}$  and survival probability  $P_{\text{survival}}$ ,

$$\sigma_{\text{production}} = \sigma_{\text{fusion}} \times P_{\text{survival}} \quad (1)$$

The first term is the yield of compound nucleus formation or the fusion cross section used in the heavy-ion reaction, while the second term is the survival probability of the produced transactinides in the process of de-excitation. Fusion cross section can be approximated by the equation

$$\sigma_{\text{fusion}} = \pi \tilde{\lambda}^2 \sum_{l=0}^{l_{\text{fusion}}} (2l+1) T_l P_{\text{CN}}, \quad (2)$$

where  $\tilde{\lambda}$  is the de Broglie wave length of the incident heavy ion,  $l$  is the orbital angular momentum, and  $T_l$  indicates the transmission coefficient of the  $l$ -th partial wave through the fusion barrier. The angular momentum  $l_{\text{fusion}}$  indicates  $l$  of the compound system up to which the partial waves contribute to the fusion. In the production of heavy nuclei, additional factors that enhance or decrease the yields of transactinides should be considered; they are fusion enhancement at sub-Coulomb-barrier energy and dynamical hindrance to fusion.  $P_{\text{CN}}$  is the probability of the compound nucleus formation influenced by the above factors. On the de-excitation process of heavy nuclei, competition between particle emission (mainly neutron) and fission (the latter destroying transactinide nuclei),  $\Gamma_n / \Gamma_f$ , should be taken into account, where  $\Gamma_n$  and  $\Gamma_f$  denote decay widths for particle emission and fission, respectively.

The combined fused system, compound nucleus, carries a certain amount of excitation energy. In accordance with the excitation energy, synthesis reactions are in general classified into the following two types: cold and hot fusion. Cold fusion reactions are characterized by relatively low excitation energies of approximately 10–20 MeV at the Coulomb barrier. The reactions of the doubly magic nuclei  $^{208}\text{Pb}$  (or  $^{209}\text{Bi}$ ) with medium-heavy ion projectiles, for example,  $^{54}\text{Cr}$ ,  $^{58}\text{Fe}$ ,  $^{62,64}\text{Ni}$ , and  $^{70}\text{Zn}$  allow to form cold compound nuclei. Only one or two steps of fission–evaporation competition,  $\Gamma_n / \Gamma_f$ , are necessary to dissipate the excitation energy. A high survival probability expected is the main advantage of this type of reactions. As shown in Table 1, elements 107 to 113 have been produced in the cold fusion reactions. However, with an increase of the projectile mass, the formation cross sections of heavy products drastically decrease due to dynamical fusion hindrance. Another question concerns the decay properties of the produced nuclei. With the increase of the atomic number of the compound nucleus,  $Z_{\text{CN}}$ , neutron-deficient nuclei are produced in the cold fusion. The half-lives of the products strongly decrease with the increase of  $Z_{\text{CN}}$ , because the stability region is expected in the neutron-rich region.

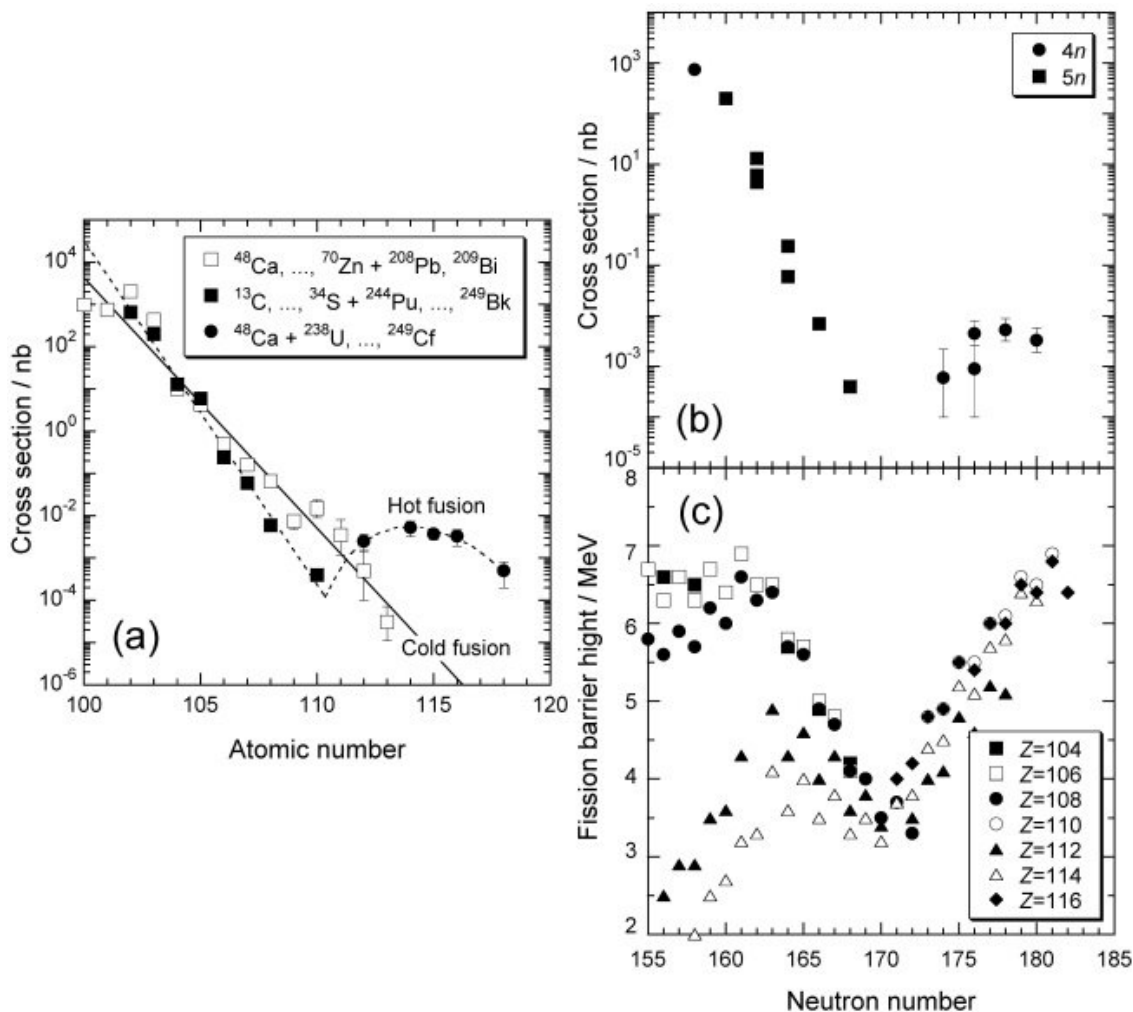


Figure 3: (a) Maximum production cross-sections of the 1n-evaporation channel in cold fusion reactions and those of the 4n- or 5n-evaporation channels in hot fusion reactions are plotted as a function of the atomic number of the compound nucleus. (b) Hot fusion cross sections for the production of nuclides with  $Z \geq 102$  using a variety of heavy-ion beams and (c) predicted fission barrier heights as a function of neutron number of the transactinide nuclei (after Oganessian 2007).

In contrast, hot fusion reactions are characterized by compound nucleus excitation energies of typically 40–50 MeV, when heavy actinides, such as  $^{238}\text{U}$ ,  $^{242,244}\text{Pu}$ ,  $^{243}\text{Am}$ ,  $^{245,248}\text{Cm}$ ,  $^{249}\text{Bk}$ ,  $^{249}\text{Cf}$ , and  $^{254}\text{Es}$  as target nuclei and neutron-rich light heavy-ions, such as  $^{18}\text{O}$ ,  $^{22}\text{Ne}$ ,  $^{26}\text{Mg}$ ,  $^{34,36}\text{S}$ , and  $^{48}\text{Ca}$  are used as the projectiles. Three to five neutrons are emitted from the compound nucleus formed in hot fusion reactions, which drastically reduces the survival probability in the cooling process. However, it is expected that the dynamical fusion hindrance is considerably smaller than in the case of cold fusion reactions. Because of the neutron-richness of the actinide targets, hot fusion reactions can produce neutron-rich nuclides that have relatively long half-lives. Thus the nuclides produced in hot fusion reactions are often used in the chemical study of transactinide elements.



To decrease the dynamical hindrance, it is desirable to make use of more asymmetric reactions and to obtain nuclides in the neutron excess region by using neutron-rich nuclides both for target and projectile nuclei. Figure 3(a) shows the maximum production cross sections of the main channels in each reaction type as a function of  $Z_{CN}$ , indicating that the cross section exponentially decreases with the increase of  $Z_{CN}$ . When  $Z_{CN}$  changes from 104 to 113, the cross section decreases by about a factor of  $10^5$ . The hot fusion reactions have smaller cross sections by almost one order of magnitude than the cold fusion reactions. It should be noted, however, that cross sections observed in the  $^{48}\text{Ca}$ -induced reactions on  $^{244}\text{Pu}$ ,  $^{243}\text{Am}$ ,  $^{248}\text{Cm}$ , and  $^{249}\text{Cf}$  are about three orders of magnitude larger than the ones expected from the extrapolation of the dashed line. As mentioned above, the survival probability  $P_{\text{survival}}$  depends mainly on  $\Gamma_n / \Gamma_f$ .

$$P_{\text{survival}} \approx \prod_{i=1}^x \left( \frac{\Gamma_n}{\Gamma_f} \right)_i \approx \prod_{i=1}^x \exp[(B_f - B_n)/T]_i, \quad (3)$$

where  $B_f$  and  $B_n$  are fission barrier and neutron separation energy (also abbreviated sometimes by  $S_n$ , see: *Radiochemistry and Nuclear Chemistry*) of the compound nucleus, respectively,  $T$  is the nuclear temperature, and  $x$  is the number of emitted neutrons. As shown in Eq. 3,  $P_{\text{survival}}$  strongly depends on the difference  $(B_f - B_n)$ . In the transactinide region, the  $B_f$  value is influenced by the magnitude of shell correction energy, so that  $P_{\text{survival}}$  depends strongly on the neutron number of the compound nucleus. The observed higher cross sections in the  $^{48}\text{Ca}$ -induced hot fusion reactions would be due to expected higher fission barriers around  $N = 162$  and  $184$  [see Figure 3(b)].

Half-lives of the transactinides decrease rapidly with increasing  $Z_{CN}$  as depicted in Figure 4. The transactinides produced so far decay almost immediately after they have been synthesized. Therefore new instrumental methods for detecting and identifying the nuclides have been required. The half-lives of the product nuclei produced in the  $^{48}\text{Ca}$ -induced reactions are about two orders of magnitude longer than those of other nuclei.

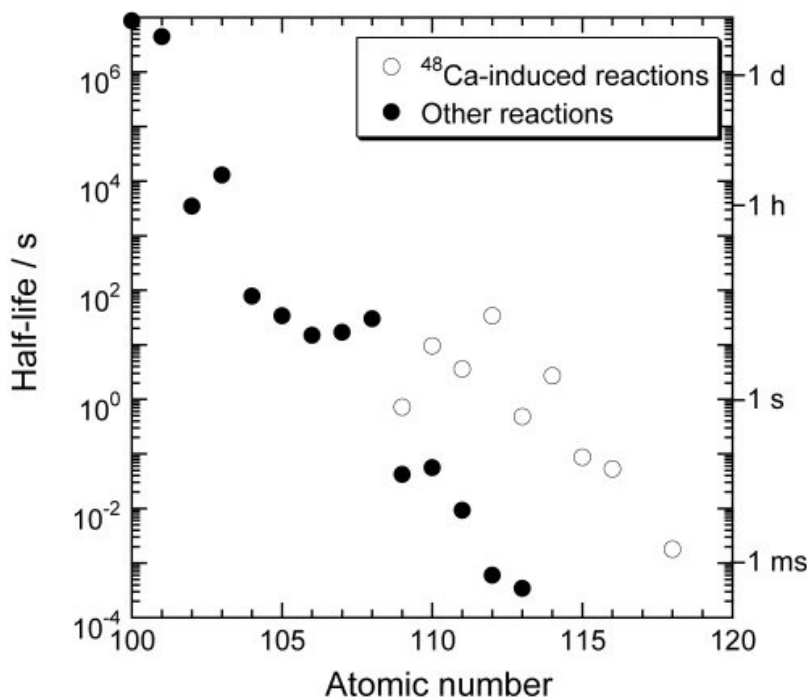


Figure 4: Half-lives of the longest-lived nuclide of each element versus atomic number of the products. The open circles indicate the longest half-life of the nuclides in each atomic number produced in the  $^{48}\text{Ca}$ -induced reactions.

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