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Discoveries of isotopes by fission

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Abstract. Of the about 3000 isotopes presently known, about 20% have been discovered in fission. The history of fission as it relates to the discovery of isotopes as well as the various reaction mechanisms leading to isotope discoveries involving fission are presented.

Keywords. Fission; isotopes.

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1. Introduction

About 3000 different isotopes have been discovered until now. A recent compilation summarized details of the discovery of all isotopes [1–4] including the year, laboratory and country of discovery as well as the production mechanism used to produce the isotopes. Fission, one of the largest contributing production mechanisms, accounts for about 20% of all the discovered isotopes. Several different methods to induce fission have been used over the years in the discovery of isotopes. Initially, fission was used by neutrons generated from sources, charged-particle accelerators, or from reactors. Later on, isotopes were also discovered following fission induced by low-energy charged particle reactions or high-energy spallation. Spectroscopy of fragments from spontaneous fission sources also contributed to the discovery of new isotopes. More recently, most of the very neutron-rich isotopes have been discovered by projectile fission. After a brief summary of the discovery of fission process itself, these production mechanisms will be discussed. The paper concludes with an outlook on future discoveries of isotopes using fission.

2. Discovery of fission

The first observation of fission was not recognized as such. When Fermi bombarded a uranium sample with an 800 mCi beryllium–radon source he interpreted the observed

activities as the potential discovery of elements heavier than uranium [5]. He drew this conclusion from the inability to assign the activities to a known heavy element: "This negative evidence about the identity of the 13 min-activity from a large number of heavy elements suggests the possibility that the atomic number of the element may be greater than 92... A careful search for such heavy particles has not yet been carried out, as they require for their observation that the active product should be in the form of a very thin layer. It seems therefore at present premature to form any definite hypothesis on the chain of disintegrations involved" [5].

This interpretation was questioned by Noddack shortly after: "Der Beweis, daß das neue Radioelement die Ordnungszahl 93 hat, ist also noch keineswegs geglückt, da Fermi ihn nur durch ein unvollkommen durchgeführtes Ausschlußverfahren versucht hat." (Proof that the new radioelement has a Z of 93, has not been established yet, as Fermi deduced it from incomplete exclusion arguments.) [6]. Noddock even hinted at the possibility of fission: "Es wäre denkbar, daß bei der Beschießung schwerer Kerne mit Neutronen diese Kerne in mehrere größere Bruchstücke zerfallen, die zwar Isotope bekannter Elemente, aber nicht Nachbarn der bestrahlten Elemente sind." (It would be conceivable that heavy nuclei which are bombarded with neutrons break up into several larger fragments which are isotopes of known elements but not neighbours of the irradiated elements.) [6].

Further doubt on the production of transuranic elements in neutron bombardment of uranium was expressed by Meitner *et al* in 1937 [7]. They observed three different decay processes which they assigned to the decay of uranium: "Also müssen die Prozesse Einfangsprozesse des Uran 238 sein, was zu drei isomeren Kernen Uran 239 führt. Dieses Ergebnis ist mit den bisherigen Kernvorstellungen sehr schwer in Übereinstimmung zu bringen." (Therefore these processes must be capture processes of uranium 238, which would lead to three isomeric nuclei of uranium 239. This result is hard to understand within the current understanding of nuclei.) [7].

In spite of these concerns, Hahn, Meitner and Strassmann continued to publish papers convinced that they had formed new transuranium elements [8,9]. In 1938, Fermi received the Nobel prize for the apparent discovery of transuranium elements and in his acceptance speech delivered on December 12, 1938, he stated: "We concluded that the carriers were one or more elements of atomic number larger than 92; we, in Rome, use to call the elements 93 and 94 Ausenium and Hesperium respectively. It is known that O Hahn and L Meitner have investigated very carefully and extensively the decay products of irradiated uranium and were able to trace among them elements up to the atomic number 96" [10]. However, in the write-up of the acceptance speech he acknowledged the potential misidentification in a footnote: "The discovery by Hahn and Strassmann of barium among the disintegration products of bombarded uranium, as a consequence of a process in which uranium splits into two approximately equal parts, makes it necessary to reexamine all the problems of the transuranic elements, as many of them might be found to be products of a splitting of uranium" [10].

Commonly, the paper by Hahn and Strassmann submitted on December 22, 1938 and published on January 6, 1939 [11] is credited with the discovery of fission. However, in this paper entitled *Über den Nachweis und das Verhalten der bei der Bestrahlung des Urans mittels Neutronen enstehenden Erdalkalimetalle* (On the proof and properties of the alkaline earth metals in the irradiation of uranium by neutrons) Hahn and Strassmann did

not dare to announce the discovery of fission: "Was die 'Trans-Urane' anbelangt, so sind diese Elemente ihren niedrigeren Homologen Rhenium, Osmium, Iridium, Platin zwar chemisch verwandt, mit ihnen aber nicht gleich. Ob sie mit den noch niedrigeren Homologen Masurium, Ruthenium, Rhodium, Palladium chemisch gleich sind, wurde noch nicht geprüft. Daran konnte man früher ja nicht denken. Die Summe der Massenzahlen Ba = Ma, also z. B. 138 + 101, ergibt 239!" (As far as the "trans-uranium elements" are concerned, they are chemically similar to their homologues rhenium, osmium, iridium, platinum, but not equal. If they correspond to the even lower homologues technetium, ruthenium, rhodium, palladium has not been tested. This is not something that one could have thought of earlier. The sum of the Ba+Ma mass numbers (138+101) is 239!).

And they go on to state: "Als Chemiker müßten wir aus den kurz dargelegten Versuchen das oben gebrachte Schema eigentlich umbenennen und statt Ra, Ac, Th die Symbole Ba, La, Ce einsetzten. Als der Physik in gewisser Weise nahestehende 'Kernchemiker' können wir uns zu diesem, allen bisherigen Erfahrungen der Kernphysik widersprechenden, Sprung noch nicht entschließen. Es könnten doch noch vielleicht eine Reihe seltsamer Zufälle unsere Ergebnisse vorgetäuscht haben." (As chemist we should rename Ra, Ac, Th to Ba, La, Ce based on the above described experiments. As "nuclear chemists" close to physics, we cannot take this step, because it contradicts all present knowledge of nuclear physics. It still could be possible that a string of weird coincidences could have simulated these events.)

Only about a month later Hahn and Strassmann did take this step in the paper *Nachweis der Entstehung aktiver Bariumisotope aus Uran und Thorium durch Neutronenbestrahlung; Nachweis weiterer aktiver Bruchstücke bei der Uranspaltung* (Proof of the production of active barium isotopes from uranium and thorium by neutron irradiation; Proof of further active fragments of uranium fission) [12] where they presented conclusive evidence for the observation of barium in uranium fission: " Endgültiger Beweis für das Entstehen von Barium aus dem Uran" (Final proof for the production of barium from uranium). This paper was submitted on January 28, 1939 and published in the journal *Naturwissenschaften* on February 10, 1939.

However, the first interpretation of their results in terms of fission had been submitted a few weeks earlier (January 16, 1939) and published one day later (February 11, 1939) by Lise Meitner and Otto Frisch in a paper entitled: *Disintegration of Uranium by Neutrons: a New Type of Nuclear Reaction* [13] in which they state: "On the basis, however, of present ideas about the behaviour of heavy nuclei, an entirely different and essentially classical picture of these new disintegration processes suggest itself. On account of their close packing and strong energy exchange, the particles in a heavy nucleus would be expected to move in a collective way which has some resemblance to the movement of a liquid drop. If the movement is made sufficiently violent by adding energy, such a drop may divide itself into two smaller drops." [13].

3. Production methods

The various probes that have been used over the years to induce fission and then detect and identify new isotopes among the fission fragments are listed in table 1.

Table 1. Approximate number of isotopes discovered in various fission reactions. The years of the first and most recent (last) discovery are also listed.

Production method	First	Last	Number of isotopes
Neutrons from sources or accelerators	1939	1962	30
Reactor neutrons	1946	1993	220
Spallation	1948	2003	40
Charged particles	1958	2007	30
Spontaneous fission	1970	2012	30
Projectile fission	1994	2014	220
Total	1939	2014	570

The number of total isotopes discovered with each method is only an approximate number because the initial assignments of the discovery of isotope project [14] have been made over a period of five years during which the criteria for discovery evolved. Currently, all assignments are being revisited to apply a unique set of standards to all isotopes.

Overall about 570 isotopes have been discovered in fission during the last 75 years corresponding to ∼20% of all known isotopes. Most of the isotopes are on the neutron-rich side of the chart of isotopes as shown by the yellow squares in figure 1.

3.1 *Source- and accelerator-neutron-induced fission*

The various groups searching for transuranic elements and ultimately discovering fission used neutrons from radium–beryllium sources to induce fission of uranium. The first

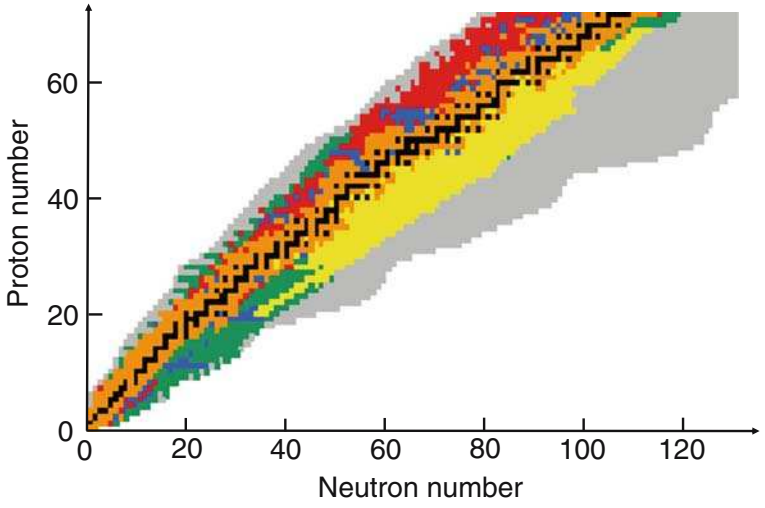

Figure 1. Section of the chart of nuclei indicating the different production methods in the discovery of isotopes. The methods used were mass spectroscopy (black), light-particle-induced reactions (orange), fusion (red), spallation (blue), projectile fragmentation (green) and fission (yellow).

identified fission fragment isotope was ^{140}Ba described by Hahn and Strassmann in their 1939 paper stating the final proof for fission [12]. Based on the measured half-life of the daughter nucleus, it was tentatively assigned to ^{140}Ba: "Was die anderen Barium isotope aus dem Uran anbelangt, so läßt sich für das Ba IV vielleicht die Hypothese machen, daß es die Muttersubstanz des in der Literatur beschriebenen Radiolanthans von 3146 Stunden Halbwertszeit mit dem vermutlichen Atomgewicht 140 ist." (Concerning the other from uranium produced barium isotopes, it is hypothesized that Ba IV may be the parent of the radioactive lanthanum which was reported with a half-life of 3146 h with the probable atomic weight of 140.) [12,15].

It is interesting to note that ^{140}Ba was the only isotope discovered using a neutron source. In 1936, Heyn had already used neutrons produced by bombarding beryllium and lithium with accelerated deuterons to discover ^{62}Cu [16] and immediately after the discovery of fission, he was the first to apply this method to induce fission and identify new fission fragments (^{88}Rb and ^{139}Cs) [17].

In 1951, Kofoed-Hansen and Nielsen discovered ^{90}Kr, ^{90}Rb and ^{91}Rb at the Copenhagen cyclotron by transferring the fission fragments directly into an ion source of an isotope separator [18]. The technique of this ground-breaking experiment was later termed isotope separation on-line (ISOL) and used for the discovery of hundreds of isotopes.

3.2 *Reactor-neutron fission*

Between 1940 and 1980 the discovery of new fission fragments was dominated by fission induced by neutrons from reactors (figure 2). A large number of isotopes were discovered during the Manhattan project but the results were published only after the data were declassified [19–21].

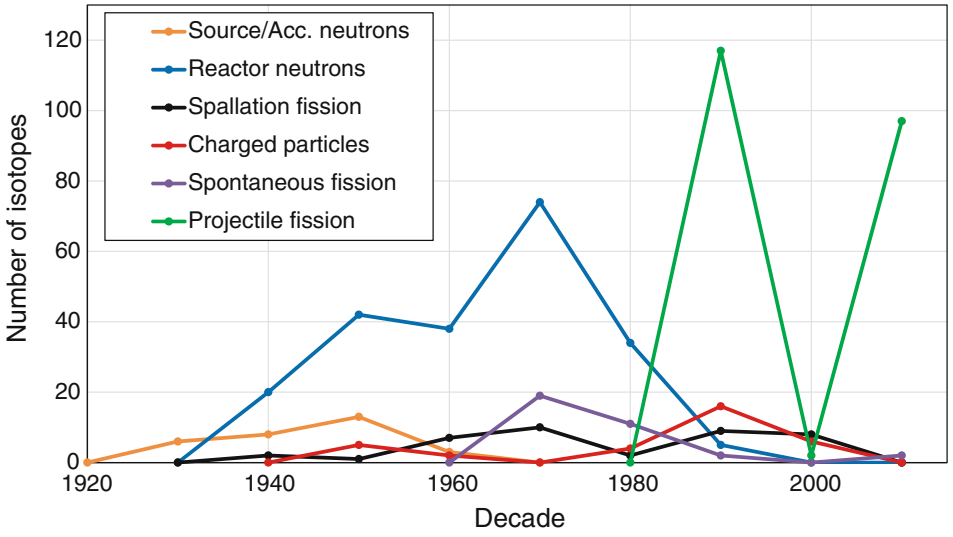

Figure 2. Isotope discoveries using different production methods as a function of time.

Most of the isotopes were discovered at reactors during the 1970s when ISOL method was used to separate the fission fragments. The first new isotopes (^{97}Y, ^{99}Zr and ^{101}Nb) were identified in 1970 with the gas-filled on-line mass separator at the FRJ-2 reactor in Jülich [22]. Subsequently, discoveries were reported from several systems around the world: OSIRIS [23] at the R2-0 reactor in Studsvik, Sweden, SOLIS [24] at the Soreq Nuclear Research Centre in Yavne, Israel, the separator at the research reactor in Munich, Germany [25], LOHENGRIN [26] and OSTIS [27] at the high-flux reactor in Grenoble, France, TRISTAN [28] at the research reactor in Ames, Iowa, and later at the high-flux beam reactor at Brookhaven National Laboratory and finally KUR-ISOL [29] at the reactor of Kyoto University in Japan. The listed references in this and the other sections refer only to the isotope discovery first reported for each method.

3.3 *Spallation fission*

As the beam energy of particle accelerators increased it was realized that fission could also be induced by bombarding heavy elements with high-energy deuterons and α-particles [30]. ^{66}Ni and ^{67}Cu were discovered by irradiating bismuth with 200 MeV deuterons from the Berkeley 184-inch cyclotron [31].

Similar to neutron-induced fission at reactors, most of the isotopes discovered by spallation reactions were identified by the ISOL method. Between 1967 and 2003, about 35 isotopes were first observed at on-line mass separators at Orsay [32] and Grenoble [33], and at CERN-ISOLDE [34].

3.4 *Charged particle fission*

Surprisingly, fission induced by low-energy charged particles was not used to explore new isotopes until about 10 years after the higher energy spallation reactions. In 1958, Alexander *et al* discovered ^{114,116,117}Ag and ^{114,115}Pd by bombarding uranium with 15 MeV deuterons at the Massachusetts Institute of Technology [35]. This approach became productive only when it was coupled with an ISOL device for separation and identification of the fission fragments. From 1988 until 2007, about 25 new isotopes were discovered at the IGISOL Facility [36] in Jyväskylä, Finland, LISOL Facility [37] in Louvain-la-Neuve, Belgium and the JAERI-ISOL Facility [38] in Tokai, Japan.

3.5 *Spontaneous fission*

During the 1970s and 1980s, approximately 30 isotopes were identified by studying the decay of ^{252}Cf spontaneous fission sources. Coincidence experiments of fission fragments with conversion electrons, X-rays and γ-rays were performed at the Lawrence Radiation Laboratory, Berkeley [39] and University of Texas [40] while chemical separation techniques [41] and the ISOL method [42] were used at the Idaho National Engineering Laboratory.

In the 1990s, ^{104}Zr [43] and ^{151}Ce [44] were discovered by measuring γ-ray cascades from a ^{248}Cm source at the Argonne National Laboratory and the Center de Recherches Nucleáires in Strasbourg, respectively. More recently, ^{155}Pr and ^{157}Nd were identified for

the first time with the Canadian Penning Trap mass spectrometer using a ^{252}Cf source at the Argonne National Laboratory [45].

3.6 *Projectile fission*

The most productive method for identifying new fission fragments is clearly the projectile fission of uranium ions accelerated to relativistic energies. Coupled with a high-resolution fragment separator, this method allows the unique mass and Z identification of fission fragments simultaneously. In the 1994 ground-breaking experiment at GSI, ^{238}U was accelerated to 750 MeV/nucleon by the heavy-ion synchroton SIS and fragments were measured with the FRS fragment separator. A total of 52 new fission fragments were identified in this experiment [46]. In three subsequent experiments another 7 [47], 58 [48] and 37 [49] isotopes were discovered.

The only other facility that is presently capable of utilizing this method is the radioactive ion-beam factory (RIBF) at RIKEN. So far, a total of 49 new isotopes have been discovered by bombarding beryllium and lead targets with a 345 MeV/nucleon ^{238}U beam and identifying the fragments with the fragment separator BigRIPS [50,51]. The discovery of another 13 isotopes has so far only been reported in an annual report [52].

Finally, although not used to produce new isotopes directly, projectile fission has recently been used in an innovative way at RIBF. Identified neutron-rich fission fragments from projectile fission were bombarded in-flight a secondary target where even more exotic nuclei were produced by projectile fragmentation. With this method, Wang *et al* [53] were able to discover ^{141}Ag.

4. Future discovery potential

The approximately 3000 presently known nuclides do not represent even half of the nuclides predicted to be particle-bound. Recently, Erler *et al* [54] have estimated that about 7000 nuclides could exist. Figure 3 shows the chart of nuclides where the stable nuclides are indicated by black squares, presently known nuclides are shown in green and additional nuclides predicted to be particle-bound by Erler *et al* [54] are shown in yellow. Two thousand of the predicted nuclides are in the region above $N > 184$ and will most likely never be discovered. Probably it is also reasonable to assume that another 500 along the neutron dripline in the region above $Z = 50$ will remain out of reach. Thus, about 1500 nuclides could be discovered in the future. To produce these nuclides, different experimental techniques have to be applied depending on the region in the chart of nuclides. Although the proton dripline has essentially been reached and even crossed in some areas, a couple of hundred nuclides are still to be discovered which are proton-unbound but have finite measurable lifetimes. As indicated in figure 3 this region of the chart can be reached with either fusion evaporation reactions (grey arrows) or projectile fragmentation reactions (red arrows).

In the future, for nuclides which cannot be reached with a combination of stable targets and beams, it is conceivable that fusion–evaporation reactions with radioactive beams can be a viable alternative. The region of superheavy elements is currently being explored only with fusion–evaporation reactions with stable beams. However, it has

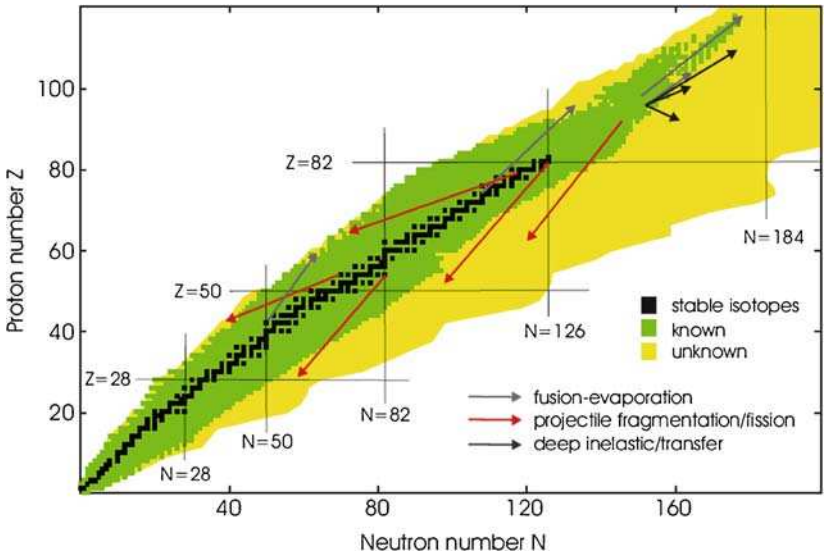

Figure 3. Chart of nuclides. Stable nuclides are indicated by the black squares, presently known nuclides are shown in green and additional nuclides predicted to be particle bound are shown in yellow. The arrows indicate different reaction mechanisms which might be used in the future to explore unknown regions [59].

been pointed out that fusion–evaporation reactions with radioactive beams [19,20] as well as deep inelastic/transfer reactions (black arrows) [20–22] could be used to populate neutron-rich superheavy nuclides. The largest unexplored region of the nuclear chart is clearly the region of neutron-rich nuclides. These nuclides can most likely be reached with projectile fragmentation or fission reactions. It is obvious that these new discoveries will only be possible with continued efforts to build new facilities with an advanced accelerator and newly developed separation and detection techniques such as the radioactive ion-beam factory (RIBF) at RIKEN [55], the facility for antiproton and ion research (FAIR) at GSI [56] and the facility for rare isotope beams (FRIB) at MSU [57,58].

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