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Discovery of Nuclei at and Beyond the Proton Dripline

M. Thoennessen

(*National Superconducting Cyclotron Laboratory and Department of Physics & Astronomy*
Michigan State University, East Lansing, MI 48824, USA)

Abstract: In contrast to the neutron dripline the proton dripline has been reached almost across the whole nuclear chart. However, because of the Coulomb barrier relatively long-lived isotopes can exist beyond the proton dripline. It is estimated that about 200 new isotopes at and beyond the proton dripline should be able to be discovered in the future. A brief review of the discovery of proton-rich nuclides as well as an outlook for the discovery potential in the future is presented.

Key words: proton dripline; isotope discoveries

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1 Introduction

A few years ago a comprehensive compilation of the discovery of all isotopes was assembled for the first time[1] and updates for 2014[2] and 2015[3] have been published. An overview of the project is available online at Ref. [4]. The compilation chronicled the growth of the chart of nuclides away from the valley of stability towards the neutron and proton driplines. Fig. 1 shows the ten-year average of the number of nuclides discovered per year (top panel) and the integral number of nuclides discovered so far (bottom panel). In addition to the total number of nuclides (black, solid lines), the figure also displays the evolution for near-stable (red, short-dashed lines), proton-rich (purple, dot-dashed lines), neutron-rich (green, long-dashed lines) and transuranium (blue, dotted lines) nuclides.

Overall, since about 2010 the number of isotopes discovered per year has increased significantly, predominantly due to the production and observation of a large number of neutron-rich nuclides. While the rate had dropped to below 20 per year between 2007 and 2009, it has now recovered to about 30 per year. At the same time discoveries of proton-rich nuclides continue to decline, falling in 2013 to below 4 per year for the first time since 1937. Nevertheless, by the end of 2014 there were still more proton-rich nuclides (1276) known than neutron-rich nuclides (1210).

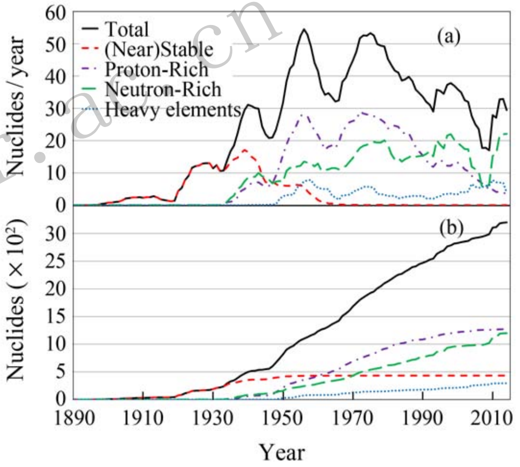

Fig. 1 (color online) Discovery of nuclides as a function of year. The top panel shows the 10-year running average of the number of nuclides discovered per year while the bottom panel shows the cumulative number. The total number of nuclides shown by the black, solid lines are plotted separately for near-stable (red, short-dashed lines), proton-rich (purple, dot-dashed lines), neutron-rich (green, long-dashed lines) and transuranium (blue, dotted lines) nuclides (from Ref. [3]).

The discovery of proton-rich nuclides can be broadly attributed to four different production mechanisms. After the development of the first particle accelerators by Cockroft and Walton[5] and Lawrence and Livingston[6] they were produced in low-energy nuclear reactions using light particles, like protons, neutrons and α-particles. As the energy of the particle accelerators increased over time additional reaction channels opened up. With the completion of the 184-inch cyclotron at Berkeley in 1947, beam energies of more than 100 MeV became available. Charged particles

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Biography: M. Thoennessen(1959–), male, USA, Professor, working on experimental nuclear physics, thoennessen@nscl.msu.edu.

accelerated to these energies could penetrate and excite the nucleus so that it would emit a large number of nucleons in the process. In this "spallation" of the nucleus many new radioactive isotopes could be populated that were not reachable in low-energy reactions. The first report of a spallation reaction was presented in an abstract of the proceedings of the meeting of the American Physical Society in July 1947 by Cunningham *et al.*[7] The term spallation for this process was first suggested by Sullivan and Seaborg a month later[8].

In the quest for new transuranium elements in the 1940s it became clear that the fusion of heavy ions would be the best method to produce elements beyond the ones produced by neutron capture or light-ion induced reactions. The first acceleration of heavy ions was achieved by Miller *et al.* at the Crocker Laboratory of the University of California at Berkeley in 1950. The intensity of the accelerated ^{12}C and ^{16}O nuclei was not sufficient to induce a detectable number of fusion reactions[9]. However, only a couple months later, Ghiorso *et al.* succeeded in identifying ^{246}Cf by bombarding ^{238}U with ^{12}C ions. The compound nucleus ^{250}Cf evaporated four neutrons to populate ^{246}Cf which then subsequently was identified by its α-radioactivity[10].

Soon thereafter it was realized that heavy-ion fusion-evaporation reactions were not only an excellent tool to produce new elements but that they also could be used to populate light neutron-deficient nuclei. Since these early attempts fusion-evaporation reactions have become one of the most productive reaction mechanisms to discover new nuclides. In addition to the almost 200 transuranium and superheavy nuclei well over 500 neutron-deficient nuclides were first identified in heavy-ion fusion evaporation reactions.

Finally, projectile fragmentation reactions contributed to the discovery of proton-rich isotopes. This technique was first used at Berkeley by Symons *et al.*[12] in 1979 to produce neutron-rich fragments by bombarding carbon targets with a 205 MeV/u ^{40}Ar beam. The resulting fragments were detected in a zero-degree magnetic spectrometer and identified in a ΔE-E silicon detector telescope[11]. It took another seven years before Langevin *et al.* discovered the first proton-rich isotopes (^{23}Si, ^{27}S, and ^{31}Ar) in a fragmentation reaction at GANIL in Caen, France.

2 Proton-rich nuclides

In the following subsections some of the discoveries along the proton drip lines will be highlighted. In light nuclei, the dripline has been crossed, and short-lived nuclei decay instantaneously by one or multiple protons. In heavier nuclides around $Z = 25$, two-proton radioactivity has been observed. Several new isotopes have recently been reported in the region around the double magic $Z = 50$, $N = 50$ nucleus ^{100}Sn. Above $Z = 50$, one-proton radioactivity is a common property in most odd-Z elements up to bismuth.

2.1 $Z < 13$

The first nucleus discovered beyond the proton dripline was ^{9}B. It was discovered in 1940 in the ^{9}Be(p,n) charge exchange reaction by Haxby *et al.*[13] at the Westinghouse Research Laboratories in East Pittsburgh, Pennsylvania. They deduced the separation energy from the onset of neutron emission as a function of beam energy. The first nucleus unbound with respect to two-proton emission was ^{6}Be, again populated in a charge exchange reaction. In 1958, Bogdanov *et al.*[14] bombarded enriched ^{6}Li with 9.6 MeV protons from the 1.5 m cyclotron of the U.S.S.R. Academy of Sciences Nuclear Energy Institute and measured the energy of the emitted neutrons by time-of-flight.

The presently only known three- and four-proton emitters ^{7}B and ^{8}C were discovered in 1967 at Berkeley and 1974 at the Kernforschungsanlage Jülich, Germany, respectively. McGrath, Cerny, and Norbeck populated ^{7}B in a three-nucleon transfer reaction – ^{10}B(^{3}He,^{6}He)^{7}B – where the excitation energy spectrum of ^{7}B was derived by measuring the ^{6}He ejectiles in a four-counter semiconductor telescope[15]. Robertson *et al.*[16] used the four-neutron transfer reaction ^{12}C(α,^{8}He)^{8}C and measured the ^{8}He ejectiles in a double-focusing magnetic analyzer.

None of these experiments actually measured the emitted proton(s). The first experiment to discover a new isotope by detecting the protons in coincidence with the fragments using the invariant mass method was performed in 2004 at GANIL. Zerguerras *et al.*[17] studied the break-up events from a secondary ^{20}Mg beam and reconstructed the ground state of ^{18}Na from the protons and ^{17}Ne fragments.

2.2 $Z \sim 25$

Proton radioactivity was first observed in the decay of an excited state of ^{53}Co which also corresponded to the discovery of this isotope. Jackson *et al.*[18] used the fusion evaporation reaction ^{16}O(^{40}Ca,p2n) to form ^{53}Co in 1979 at the Harwell variable energy cyclotron. These results were confirmed by Cerny *et al.*[19] at the Berkeley 88-inch cyclotron who also observed delayed protons following the reaction ^{54}Fe(p,2n)^{53}Co. The two papers were submitted on the same date and published sequentially in the same issue of Physics Letters B. It should be noted that Cerny was a co-author on both papers.

The discovery of two-proton radioactivity was also almost simultaneously reported from two laboratories. Pfützner *et al.*[20] submitted first evidence of the two proton decay of ^{45}Fe on May 17, 2002 from the fragmentation of a 600 MeV/nucleon ^{58}Ni beam with the FRS at GSI in Darmstadt, Germany. Only four days later, Giovinazzo *et al.*[21] submitted their results of the fragmentation of 75 MeV/nucleon ^{58}Ni at the SISSI-LISE3 facility at GANIL. ^{45}Fe itself had already been discovered six years earlier by Blank *et al.*[22] at GSI.

2.3 $Z \sim 40$

The most recent observation of a new isotope along the proton dripline was the identification of ^{59}Ge with the Coupled Cyclotron Facility (CCF) of the National Superconducting Cyclotron Laboratory at Michigan State University submitted on June 16, 2015. Ciemny *et al.*[23] identified 4 events of ^{59}Ge in the fragmentation of 150 MeV/nucleon ^{78}Kr. At the 5th International Conference on Proton-Emitting Nuclei held July 6-10, 2015 at the Institute of Modern Physics of the Chinese Academy of Sciences in Lanzhou, Gernhäuser *et al.*[24] reported preliminary results of the discovery of ^{90}Pd, ^{92}Ag, ^{94}Cd, ^{96}In, ^{98}Sn, and ^{104}Te.

The 1995 discovery of ^{103}Sb by Rykaczewski *et al.*[25] has recently been questioned by the non-observation of events for this isotope in the fragmentation reactions of ^{124}Xe beams of 1 GeV/nucleon at GSI[26–27] and 345 MeV/u at the RIBF accelerator complex and the BigRIPS separator at RIKEN, Japan[28].

2.4 $Z > 51$

The first observation of ground state proton radioactivity was achieved in 1982 at the UNILAC at GSI by Hofmann *et al.* They observed position-time correlations of implanted residues and delayed protons in a detector array at the end of the velocity separator SHIP following the fusion evaporation reaction ^{96}Ru(^{58}Ni,p2n)^{151}Lu[29]. Since then a total of 31 isotopes exhibiting proton radioactivity have been discovered in the mass range between antimony and bismuth.

Fig. 2 shows a section of the chart of nuclides for proton-rich isotopes between antimony and holmium. Proton emitters are indicated by the dark gray boxes and the calculated proton dripline is shown by the thick black line. The figure shows that there are several proton-bound even-Z elements that have not been discovered yet. In addition, for the odd-Z elements there are still unknown isotopes between the last β-decaying isotope and first proton-emitting isotope.

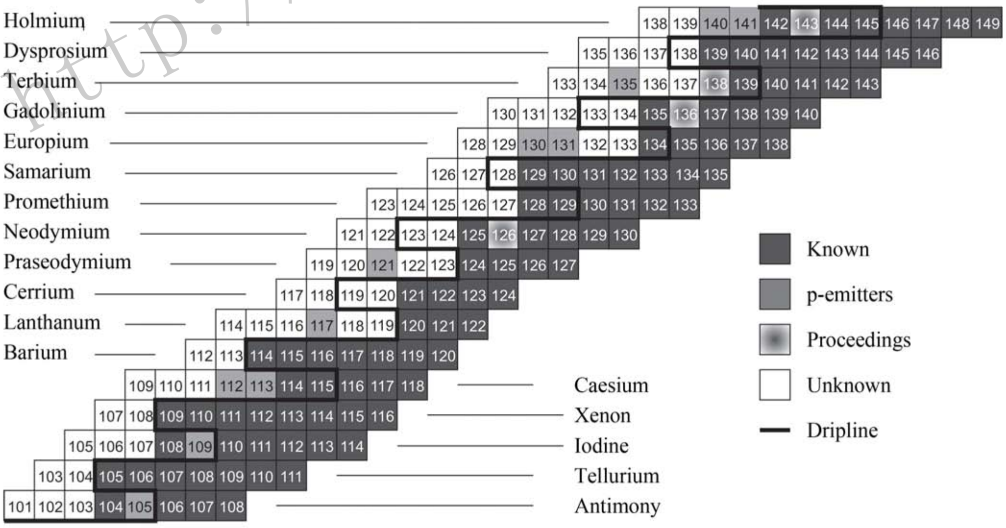

Fig. 2　Section of the chart of nuclides for proton-rich isotopes between antimony and holmium. Beta- and proton-emitting nuclides are shown as dark and light gray boxes. Nuclides reported only in conference proceedings are shown by the white-to-black shaded boxes while unknown nuclides are shown in white. The thick black line corresponds to the calculated proton dripline.

Isotopes that so far have only been reported in conference proceedings or internal reports are shown as white-to-black shaded boxes. Souliotis had presented the identification of ^{126}Nd, ^{136}Gd, ^{138}Tb, and ^{143}Ho as well as ^{150}Yb and ^{153}Hf which are not shown in the figure, at the International Conference on Achievements and Perspectives in Nuclear Structure held in Aghia Palaghia, Crete, Greece, July 11-17, 1999[30], however, the results were subsequently not published in the refereed literature. The observation of ^{143}Ho was also reported in an annual report by Seweryniak *et al.*[31]. In addition, although the discovery of ^{144}Tm was presented at several conferences[32–34] it was never published in a refereed journal.

The exploration of proton-rich isotopes in the rare earth region has been a major research focus of nuclear physics in China. As shown in Fig. 3 eleven new isotopes in this mass region have been discovered at the Institute of Modern Physics of the Chinese Academy of Sciences in Lanzhou.

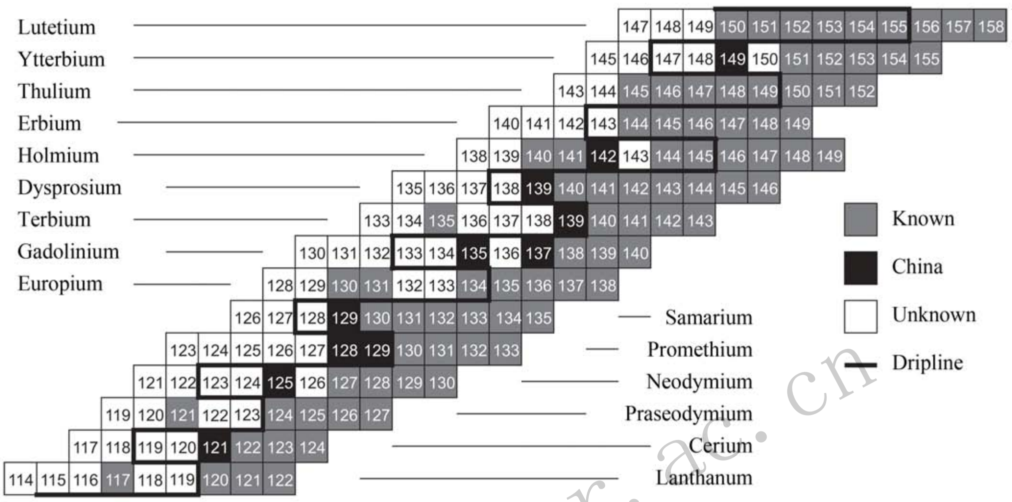

Fig. 3 Section of the chart of nuclides for proton-rich isotopes between lanthanum and lutetium highlighting isotopes discovered at the Institute of Modern Physics of Chinese Academy of Sciences in Lanzhou, China.

3 Future

As of September 1, 2015, the discoveries of 3 208 isotopes have been published in refereed journals. They represent not even half of the nuclides predicted to be particle bound as recently estimated by Erler *et al.*[35]. Although the proton dripline has essentially been reached and even crossed in some areas, there are still several hundred nuclides to be discovered which are either proton bound or unbound but have finite measurable lifetimes. Fig. 4 shows a section of the chart of nuclides for proton-rich isotopes between calcium and mercury. It displays presently known nuclei (black squares), the dripline calculated with the empirical mass formula of Tachibana *et al.*[36] (dashed line), and a simple extrapolation of the lifetime limit of $\sim 10^{-9}$ s (solid line). The grey squares show the estimated reach of the future Facility for Rare Isotope Beams (FRIB)[37–38] currently under construction at Michigan State University assuming a production limit of approximately one nucleus per day. The figure shows that FRIB will be able to map out the whole dripline including all even-Z nuclei up to $Z = 80$. FRIB has the potential to produce well over 200 new nuclides most of them beyond the proton dripline. About 100 nuclei will still be out of reach. In addition to FRIB other next generation radioactive beam facilities will contribute to the discovery of new proton-rich isotopes. At RIKEN, the Radioactive Ion-Beam Factory RIBF is already in operation[39] while at GSI the Facility for Antiproton and Ion Research FAIR is under construction[40]. Most likely most of the isotopes will be produced in projectile fragmentation reactions. Th-

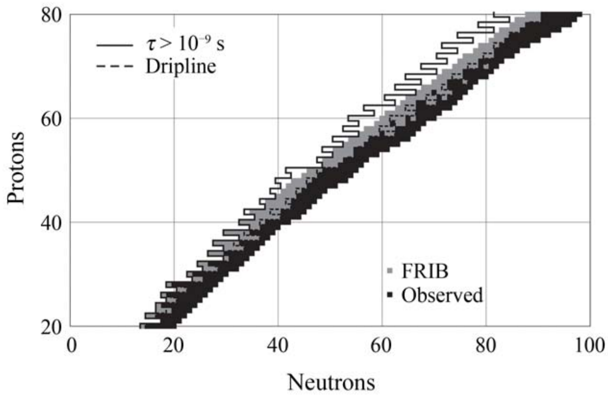

Fig. 4 Section of the chart of nuclides for proton-rich isotopes between calcium and mercury. Known isotopes are shown in black and isotopes predicted to be discovered at FRIB are shown in gray. The dripline is indicated by the dashed-line while the solid line corresponds to an estimated lifetime limit of about a nano-second.

us equally critical for new discoveries at RIBF, FAIR, and FRIB are the next generation fragment separators, BIG-RIPS[41–42], the Super FRS[43], and the FRIB fragment separator[44], respectively.

In addition to these facilities there are two other major facilities with projectile fragmentation capabilities being planned, both of them in Asia. The High-Intensity Heavy Ion Accelerator Facility (HIAF) being designed at the Institute of Modern Physics of the Chinese Academy of Sciences will be based on a heavy ion superconducting linac, an accumulation booster ring and a multifunction storage ring system[45]. The future Korean Rare Isotope Beams Accelerator Facility (KRIA) will have rare isotope beams with energies of up to 250 MeV per nucleon produced in projectile fragmentation as well as high quality and intense re-accelerated ISOL beams[46].

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