

# Big Science: The Discovery of Tennessine

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#### Oak Ridge National Laboratory recognizes and thanks our partner institutions in this collaborative effort

The Joint Institute for Nuclear Research, Dubna, Russia Vanderbilt University The University of Tennessee, Knoxville Lawrence Livermore National Laboratory The Research Institute for Advanced Reactors, Dimitrovgrad, Russia University of Nevada, Las Vegas

## BIG SCIENCE: The Discovery of Tennessine

The international effort to create superheavy element 117—Tennessine—began in late 2004 when Yuri Oganessian of the Joint Institute for Nuclear Research (JINR) in Dubna, Russia, proposed the experiment to the Department of Energy's Oak Ridge National Laboratory.

Oganessian had developed a "hot fusion" method of bombarding actinide targets with a neutron-rich calcium-48 beam that had resulted in the synthesis, and thus discoveries, of elements 114, 115, 116 and 118. A "cold fusion" method of bombarding lead or bismuth with heavy ions had produced elements 106 through 113.



Yuri Oganessian

Synthesizing element 117 to fill the remaining square in row seven of the Periodic Table required a target of the radioisotope berkelium-249 (Bk-249), which in theory would produce the short-lived element if relentlessly bombarded with a calcium-48 beam at JINR's Gas-Filled Recoil Separator.

Berkelium-249, however, is an exclusively man-made material attainable as a byproduct of the production of californium-252 (Cf-252), another synthetic radioisotope used in medicine and industry. ORNL is the world's only source of Bk-249 in sufficient quantities for the proposed experiment. Bk is typically produced as a byproduct of Cf-252, which was not in production in February 2005, when Oganessian visited ORNL. Still, a collaboration between JINR, ORNL, Lawrence Livermore National Laboratory, and Vanderbilt University was proposed to await the next campaign to separate Cf-252 for the Department of Energy's isotope program.

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Vanderbilt's advocate for the element 117 experiment was physics professor Joe Hamilton, who has a history of collaboration with ORNL going back to the establishment of the University Isotope Separator at Oak Ridge in the early 1970s and the Joint Institute for Heavy Ion Research that involved ORNL, Vanderbilt and the University of Tennessee in the early 1980s.



Jim Roberto

When a Cf-252 campaign materialized at ORNL in 2008, Hamilton informed Oganessian and introduced him to Jim Roberto, ORNL's director of Scientific and Technology Partnerships, whose materials research interests include heavy element nuclear physics. Roberto, in turn, set in motion the effort to retrieve Bk-249 for the element 117 experiment during the Cf-252 campaign. Roberto and Oganessian agreed to coordinate the Bk-249 effort with the JINR accelerator's availability.

Lawrence Livermore National Laboratory also joined the project. LLNL had collaborated with JINR in the discoveries of superheavy elements 114, 116 and 118. Element 116 was later named livermorium after the California lab.

ORNL's nuclear expertise and capability to produce exotic radioisotopes originated with the laboratory's World War II mission to demonstrate the production of plutonium in a nuclear reactor. That Manhattan Project work was rapidly accomplished: The X-10 Graphite Reactor reached criticality on November 4, 1943, and demonstrated the production and separation of gram quantities of plutonium. Large-scale plutonium production was undertaken at Hanford, Wash., and a plutonium test device was detonated in the New Mexico desert on July 16, 1945.

The scientists who were assembled for the urgent wartime project lost no time in mining the reactor's potential for scientific research and established important groundwork for nuclear medicine, neutron scattering science, and the nuclear power industry. E.O. Wollan performed early neutron diffraction experiments at the Graphite Reactor, which he pursued with Clifford Shull. Shull eventually shared the Nobel Prize for Physics based on this pioneering neutron science work.

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Chemical separation science was also integral to the success of the Manhattan Project. One byproduct of the quest for a nuclear weapon was the separation of the theorized lanthanide element 61 at ORNL. The team of discoverers, which included Charles Coryell, Larry Glendenin and Jacob Marinsky, named the new element "promethium," inspired by the mythological Prometheus, who stole fire from heaven to benefit mankind.

Radiochemistry and isotope production have remained leading scientific capabilities at ORNL. ORNL is one of the world's largest producers of rare radioisotopes, with much of the work centered at the High Flux Isotope Reactor, where targets are irradiated, and the adjoining Radiochemical Engineering Development Center, where the radioisotopes are processed, separated and purified.



The accelerator complex at the Joint Institute for Nuclear Research, Dubna, Russia.

The radioactive, neutron-emitting isotope Cf-252 is produced through an 8-month exposure of americium and curium targets to the world's highest steady-state neutron flux. After being irradiated at HFIR, the targets are processed at REDC. Berkelium-249 can also be obtained in the process.

With a Cf-252 program on the horizon, Roberto, Oganessian and Hamilton met in Nashville in 2008 to plan the experiment. The Department of Energy approved the separation campaign in November, including production of

Bk-249 for research purposes (i.e., discovering element 117). ORNL and JINR agreed on a schedule for the experiment including transfer of the Bk-249 to JINR, dedicated accelerator time for the experiment at JINR, and collaboration in the related research.

Roberto asked Kzrysztof Rykaczewski, a nuclear physicist in ORNL's Physics Division, to help facilitate the international scientific effort. Rykaczewski is a veteran nuclear spectroscopist who has discovered more than 60 isotopes, 10 of them at ORNL, and represents decades of nuclear physics research and experience in discovering new alpha and proton emitting isotopes. A native of Poland who speaks Russian, Rykaczewski was also comfortable working across borders.

The Bk-249 targets emerged from HFIR in January 2009 to begin the nearly six months of processing, three of those simply to decay to a point they could be handled. Processing requires several more months, which for the purposes of the 117 experiment represented a considerable tax on Bk-249's 330-day

half-life. REDC produced 22 milligrams of Bk-249 with impurities of less than one part in 10<sup>7</sup>, an achievement that impressed researchers on the receiving end at Dubna.

REDC technicians captured a photo of the rare radioisotope in a vial, the 22 milligrams of green fluid competing with the hot cell's yellow lithium bromide patina. REDC technicians completed the processing by mid-June, and the shipping phase commenced. The carefully packaged and labeled shipment was separated into five parcels resembling large paint buckets.



Rykaczewski worked with ORNL's Isotope Business Office and a shipping company to prepare the radioisotopes for transit, which included translating documents and obtaining approvals and permits. It was new territory: Isotope shipments to Dubna typically involved materials with half-lives of hundreds of years. Bk-249's was less than a year. The packages embarked on a nerve-wracking odyssey that included two round trips across the Atlantic after the shipment was turned away twice. More time was lost when the parcel was delayed for various, mostly bureaucratic, reasons.

Eventually, the Bk-249 parcels were delivered to Dimitrovgrad's Research Institute for Atomic Reactors for target preparation in what still is the fastest transfer of actinides between the United States and Russia on record.

There were technical hitches, as well: The Russian team, which had requested the radioisotope in the form of berkelium nitrate, initially had difficulty extracting the Bk-249. They questioned at one point whether they really had Bk-249 and eventually resorted to a strong concoction of three acids, called "tsar vodka," to retrieve the radioisotope. The team then hand-painted the radioisotope onto the target because the original plan to electroplate it produced unsatisfactory results.

Time on the Bk-249's 330-day half-life was ticking away the whole time, but the hand-painted target worked well, and on July 28 the targets were flown to Dubna to make up time, and the calcium-48 beam was applied to the target at JINR's Gas-Filled Recoil Separator.

In addition to the Bk-249, ORNL sent research staff members to Dubna and provided detectors, instruments and digital electronics for detecting shorter-lived isotopes, the products of knowledge gained from decades of nuclear physics work at ORNL. After 150 days of irradiation with a beam of 6 trillion calcium-48 ions per second, the team reported it had detected six atoms of element 117, which then decayed into elements 115, 113, 111, 109, 107 and 105. *"I really wanted to be sure. We were looking for a very few events, and we knew what to look for in our previous work."* — Krzysztof Rykaczewski

Rykaczewski, who spent weeks in Russia with the experiment, describes obtaining the results:

"I really wanted to be sure. We were looking for a very few events, and we knew what to look for from our previous work. We were sending 6 trillion projectiles per second and on average we got one event per month—a chain of correlated alphas as they decay in milliseconds at first, then longer into seconds. It is a very distinct signature correlated in space and in time. This is how we detect superheavy molecules—the sequence of decays.

"The probability that it is random is 10<sup>-6</sup>," or one in a million, Rykaczewski said. "It cannot be random."

The data analysis of the thousands of candidate reactions by Dubna and LLNL researchers supported the existence, however fleeting, of the half dozen atoms of element 117. Authors representing JINR, RIAR, ORNL, LLNL, Vanderbilt and the University of Nevada, Las Vegas announced the discovery on April 9, 2010, in Physical Review Letters. The publication also noted a strong rise in stability for superheavy isotopes (above Z=111), supporting the theorized Island of Stability (see "Significance of element 117" on page 9).

Confirmation experiments followed the announcement. JINR, ORNL, LLNL, Vanderbilt and the University of Tennessee, Knoxville (UTK) observed an additional 14 atoms of element 117 at JINR in 2012 and 2013 using berkelium from ORNL.

A group of scientists from 16 institutions in Australia, Finland, Germany, India, Japan, Norway, Poland, Sweden, Switzerland, the United Kingdom and the United States conducted confirmation experiments to independently verify the discovery of element 117. This independent sighting of element 117,

which observed two atoms, was presented in a Physical Review Letters study published in May 2014. The research involved the production—again—of berkelium at ORNL and bombardment with high-power calcium ion beams in an accelerator at GSI Helmholtz Centre for Heavy Ion Research in Darmstadt, Germany. University of Tennessee professor Robert Grzywacz, a former Eugene Wigner Fellow at ORNL, worked with ORNL to develop data acquisition technology that was used in the confirmation experiments.

On Dec. 30, 2015, a committee comprising members of the International Union of Pure and Applied Physics (IUPAP) and the International Union of Pure and Applied Chemistry announced the criteria for the discovery of a new element had been met and invited the collaborators to propose a name and symbol to replace element 117's working name of ununseptium.

ORNL physicists developed this detection system for superheavy element research.



There was precedent for naming new elements after geographic locations relevant to their discoveries (e.g., americium, californium, livermorium, dubnium). The name ultimately proposed—"tennessine"—aligns with the halogen group of the Periodic Table—fluorine, chlorine, bromine, iodine and astatine in column 7A.

The symbol would be Ts. (The more intuitive Tn is a discontinued but sometimes used symbol for tungsten.)

Because institutions in the state of Tennessee played a major role in the experiment, the collaboration agreed that tennessine would be an appropriate name for ununseptium, and on June 8, 2016, IUPAC's Inorganic Chemistry Division published a provisional recommendation for the name tennessine and symbol Ts.

After a five-month public review period, element 117 officially joined the Periodic Table as tennessine on November 28, 2016, "in recognition of the contribution of the Tennessee region, including Oak Ridge National Laboratory, Vanderbilt University, and the University of Tennessee at Knoxville, to superheavy element research, including the production and separation of unique actinide materials for superheavy element synthesis."

In the same announcement, IUPAC officially named element 115 moscovium in recognition of the Moscow region, home to JINR, and element 118 oganesson, in recognition of Yuri Oganessian's pioneering contributions to superheavy element research.

In addition to the berkelium-249 for element 117, ORNL-produced radioisotopes were used in the discoveries and confirmations of superheavy elements 114 (Pu-244), 115 (Am-243), 116 (Cm-248) and 118 (Cf-249).

#### Significance of element 117

In the current periodic table, elements beyond uranium (atomic number of 92) are increasingly unstable and decay rapidly into other elements.

Nuclear physicists beginning with nuclear pioneer Glenn Seaborg have theorized an "island of stability" beyond the current periodic table where new superheavy elements would exhibit longer lifetimes. Such an island would extend the periodic table to even heavier elements, and the increased lifetimes would make chemistry experiments and potential applications for these heretofore unknown elements increasingly practical.

Element 117 was the only missing element in row seven of the periodic table. On course to the island of stability, researchers initially skipped element 117 due to the difficulty in obtaining the berkelium target material. The observed decay patterns in the new isotopes from the element 117 experiments bring scientists closer to the island of stability and continue a general trend of increasing stability for superheavy elements with increasing numbers of neutrons in the nucleus. The discovery of element 117 provides strong evidence for the existence of the island of stability.



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### A Special Eugene P. Wigner Distinguished Lecture

Recognizing the scientific discovery and naming of element 117 (tennessine) Friday, January 27, 2017 • 3 to 5 p.m. Iran Thomas Auditorium • Oak Ridge National Laboratory

#### Welcome and Introduction

Dr. Thom Mason, Director, Oak Ridge National Laboratory

#### Eugene P. Wigner Distinguished Lecture Discovering Superheavy Elements

Dr. Yuri Oganessian Scientific Leader, Flerov Laboratory of Nuclear Reactions, Joint Institute for Nuclear Research

#### **Question and Answer Session**

Dr. Yuri Oganessian Dr. Thom Mason

#### **Screening of Element 117 Video**

#### Remarks

Dr. Thom Mason Dr. Timothy Hallman, Associate Director of Science, Office of Nuclear Physics, US Department of Energy Dr. Victor Matveev, Director, Joint Institute for Nuclear Research Governor Bill Haslam, State of Tennessee

#### **Announcement of Donation of Periodic Tables**

Dr. Thom Mason Governor Bill Haslam

#### **Concluding Remarks**

Dr. Thom Mason

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Reception in the Spallation Neutron Source Lobby