IUPAC/IUPAP COMMITTEE MEDIATES CUSTODY BATTLE OVER HEAVY ELEMENTS

Unlike babies, which are usually named at birth, a number of heavy elements have remained anonymous even though some are nearly 30 years old. These nameless orphans have been the objects of bitter custody battles: Three major research groups—located at the University of California at Berkeley, the Joint Institute for Nuclear Research in Dubna, Russia, and the Laboratory for Heavy Ion Research (GSI) in Darmstadt, Germany—have competed intensely for years to produce the heavy elements and have filed conflicting claims for discovery. But given the difficult nature of the experiments on such short-lived isotopes, it hasn't always been clear which group has really produced a given element.

Seeking to resolve this impasse, the International Unions of Pure and Applied Chemistry and of Pure and Applied Physics established a Transfermium Working Group in 1985 to consider which group should be accorded priority for discovering each element. The TWG published its report in the spring of 1992 in *Progress in Particle and Nuclear Physics*,[1], and again this August in the IUPAC journal, *Pure and Applied Chemistry*.[2] But its judgment has been accepted by only two of the three major research groups involved.

In statements published in reply, Glenn Seaborg and Albert Ghiorso,[2,3] leaders of most of the Berkeley experiments, raise a number of objections; they especially protest the TWG conclusion that Berkeley and Dubna should share the credit for elements 104 and 105. In separate comments, representatives of the Dubna and Darmstadt groups accept the conclusions of the TWG, although each group expresses some reservations: Yuri T. Oganessian and Ivo Zvara of the Dubna group feel[2,4] that the TWG had underestimated the importance of some of their papers on elements 103–105. And Peter Armbruster and his colleagues at GSI in Darmstadt don't fully agree[2,5] with the TWG decision to accord "major" rather than full credit to Darmstadt for element 108.

Composition of the working group

The TWG was not the first group to take a stab at resolving the conflicting claims for elements. In 1974 IUPAC and IUPAP set up a committee to examine rival US–Soviet claims for elements 104 and 105. The push for such a committee came largely from Georgi Flerov and his colleagues from the Dubna group. But the committee, composed of experts from the US, the USSR and other countries who were not involved in the relevant experiments, never met or issued a report.

For the TWG, IUPAC and IUPAP appointed people who were deemed to be informed but impartial and who had no connection to the work being studied or to the countries where it was done. IUPAP took the initiative for the study and named seven physicists to the working group. IUPAP invited the participation of IUPAC, which in turn appointed two chemists. Seaborg and Ghiorso complain that the working group had no *nuclear* chemists. Over the period from 1987 until its report was issued in 1991, the working group met seven times, for about one week each time, and between meetings the members corresponded extensively.

Even though elements 101, 102 and 103 were named long ago, the TWG considered all elements with atomic number $Z > 100$, because the group felt that some questions of priority needed to be resolved for each one, according to the TWG's chairman, Denys Wilkinson of the University of Sussex, England. Wilkinson stressed to us that the group was concerned solely with priority in discovery and not with the assignment of names.

But, of course, the name is the game. The group that first produces a new element traditionally earns the right to propose its name. The final recommendation for element names is made by the IUPAC Commission on Inorganic Nomenclature, although the commission is not obligated to take the proposed name; practicality and common usage are considered as well.

The first task of the TWG was to settle on the criteria for priority in discovery, but the members found that they could not come up with a rigid set of rules. In its report, the TWG does discuss the kinds of chemical or physical properties that would help identify an unknown element, and also measurements that would help prove that the measured property was indeed associated with the unknown. But the group also points out that a discovery is not always black-and-white; rather, conviction builds up in stages as evidence accumulates from different experiments. "There may be cases," the TWG states in its report, "in which it would be unjust to assign an absolute priority in the 'discovery' of a new element but where the credit should be appropriately apportioned." Indeed the group gave joint or shared credit for four of the nine elements it considered.

The second task was to assign priorities. For this, the working group studied the relevant papers and met with each research group at its laboratory. In their report the TWG members provide what they call a "discovery profile," summarizing the key experiments they feel led to the identification of a particular element.

Elements 101–103

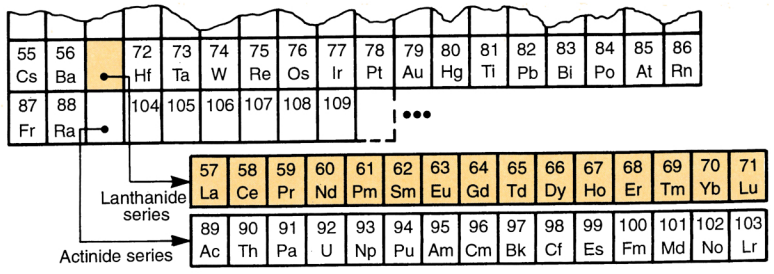
The first three elements the TWG considered were those that were already named. In 1955 a Berkeley group that included Ghiorso and Seaborg used an ion-exchange technique to separate an element with $Z = 101$. The Berkeley group proposed that this new element be called mendelevium, and the IUPAC Commission on Inorganic Nomenclature accepted the name. The TWG members were not, however, fully convinced by the 1955 paper; they still assigned priority to Berkeley but based it mostly on a 1958 experiment that identified a different isotope using essentially the same chemical method supplemented with proof of a link between the element-101 parent and its fermium daughter. Ghiorso and Seaborg argue that their 1955 experiment involved a standard chemical technique that, on its own, constituted acceptable identification. The distinction matters because the 1958 experiment was done by a different set of researchers at Berkeley, only one of whom was also in the group that did the 1955 work.

The first evidence for element 102 was reported in 1959 by a collaboration of researchers from Argonne National Laboratory, the Atomic Energy Research Center in Harwell, England, and the Nobel Institute of Physics in Stockholm. This group proposed the name nobelium, for Alfred Nobel. IUPAC immediately accepted the name, but it turned out that the data were not strong enough to support the claim. Subsequently, first the Berkeley group and then the Dubna group claimed credit for element 102. The Soviet

researchers proposed the name joliotium, while the Berkeley team suggested that the name nobelium be retained. "Nobelium" has stuck.

The TWG concluded that two experiments in 1966 by the Dubna group produced the first convincing evidence for element 102. The TWG discusses the Berkeley work of 1958 and 1961 in its discovery profile but does not mention that work in stating its conclusion. The omission has disappointed Ghiorso and Seaborg.

Both Berkeley and Dubna have laid claim to element 103 over the years. Berkeley proposed the name lawrencium, and the Commission on Inorganic Nomenclature recommended that name. The Dubna researchers, critical of the Berkeley experiment, felt that the commission's decision was hasty and made an alternative, tentative proposal to call this element rutherfordium. The TWG, however, traces a complex chain of experiments in which confidence in the discovery was gained only through accumulation of evidence from both rivals. The TWG thus proposes that credit be shared. Ghiorso and Seaborg agree fully and feel that the TWG treatment of this discovery was the best in the report.



Heavy elements at the end of the periodic table. The priority for discovering elements with atomic numbers 103 through 109 has been disputed, so they have been nameless. As a result of the report of the Transfermium Working Group, researchers at GSI in Darmstadt have proposed names for elements 107 to 109.

The fight over 104 and 105

The competition between Berkeley and Dubna is perhaps the most intense when it comes to elements 104 and 105. In the days when those elements were discovered the Berkeley team was led by Ghiorso and the Dubna group by Flerov. Ghiorso and his colleagues proposed the names rutherfordium and hahnium for elements 104 and 105, while Flerov's team put forward kurchatovium and nielsbohrium, respectively. IUPAC has made no recommendation regarding those names. The TWG recommended that Berkeley and Dubna share credit for both elements.

For element 104 the working group credits experiments by Berkeley that determined, the geneological alpha-decay sequence; such experiments are unequivocal, if done correctly. Regarding the Dubna claims, the TWG cites in particular a series of experiments directed by Zvara. Element 104 is the first element beyond the actinides—that is, those elements between actinium (Z = 89) and lawrencium (see the portion of the periodic table above)—and the experiments by Zvara's group exploited the expectation that it would form a chloride that was volatile, unlike those formed by the actinides. The recoil products from the bombardment of a plutonium target by ^{22}Ne ions entered a region where they could react with gaseous chloride compounds. By separating the chlorides from the gas stream, the Dubna team hoped to isolate any atoms of element 104 that had linked up with chlorine. Researchers from Dubna also did a series of experiments trying to identify element 104 from its spontaneous fission. In both types of experiments there was considerable confusion about the halflives of the elements that were being produced. However, the TWG did some calculations on the original Dubna data and concluded that Zvara and his colleagues had actually seen an isotope of element 104 (^{259}104) in its radiochemical experiments, even though the group had misinterpreted its halflife. "The significant point is not the determination of a halflife," the report states, "but rather the transmission of a spontaneous fission activity in situations where Z = 104 could be formed and the absence of it when only products with Z < 104 are possible."

It is the TWG decision regarding element 104 that has brought the strongest reaction from Ghiorso and Seaborg. The pair feel that the basis of the Dubna claims for many years was the evidence from spontaneous-fission experiments, rather than Zvara's chemical experiments. The fission experiments found evidence for an 80-millisecond activity, which Ghiorso and Seaborg assert even the Dubna researchers no longer think exists. The TWG report did not mention those experiments. Regarding Zvara's volatility experiments, Ghiorso and Seaborg question whether a large enough fraction of the isotope ^{259}104 decays by spontaneous fission for the Dubna group to have observed it. The US members of the abortive 1974 IUPAC–IUPAP committee on 104 and 105, in a paper written on their own initiative but only published many years later,[6] also expressed the opinion that the Dubna volatility experiments were not convincing. Ghiorso and Seaborg feel that Zvara's technique should not be described as "chemical," because the separation of the chlorides involved a mechanical sweeping aside of the volatile elements. They also view as "highly irregular" the retrospective treatment of the Dubna data by the TWG.

Aaldert H. Wapstra of NIKHEF (the National Institute for Nuclear and High-Energy Physics) in Amsterdam, who served on the TWG, defends the group's conclusion. He pointed out to us that Ghiorso and Seaborg criticize the TWG in the case of element 101 for not taking into account the chemical evidence, but then fault the TWG for relying too heavily on the chemical evidence for element 104. He added that the working group had done retrospective calculations on the data from other research groups as well.

The dispute over element 105 concerns which group produced its evidence first. The TWG concludes that "independent work reported in 1970 from Berkeley . . . and from Dubna . . . was essentially contemporaneous and equally convincing." Ghiorso and Seaborg disagree, claiming that the Dubna work followed the Berkeley experiment by a year. Zvara points out that the TWG also describes some papers from Berkeley as "contemporaneous" with ones from Dubna that preceded them by about a year.

The TWG accorded credit for element 106 to a collaboration between Berkeley and Lawrence Livermore National Laboratory. Just this September, at the Actinide '93 conference, held in Santa Fe, New Mexico, a different Berkeley group, led by Ken Gregorich and Darleane Hoff-

man, reported the production of the same isotope of 106. At the same conference a collaboration between researchers from Dubna and Lawrence Livermore reported producing an unexpectedly long-lived isotope of 106. (See the accompanying news story.)

Elements 107 and 109 have been credited to Darmstadt, and no one has publicly objected. The TWG also gives "major credit" for 108 to the Darmstadt researchers, because their evidence can stand alone whereas the Dubna experiment is convincing only when combined with some cross-section measurements made at Darmstadt. The Darmstadt group feels that it deserves sole credit for $Z = 108$ but is committed to going along with the conclusions of the working group.

Assigning names

Since the TWG report appeared, only the GSI researchers have proposed names, for elements 107 through 109. They struck an interesting compromise to fulfill their obligation to make some kind of joint proposal with Dubna for element 108: In essence, they would let the Russians have one name out of the three they proposed—but it would not be the one for element 108. Specifically, the GSI group members proposed for element 107 the name nielsbohrium, one of the names that the Dubna group had picked for another element (105); for element 108, they put forth the name hassium, after the Latin name for the state of Hesse, where GSI is located; and for 109 they chose meitnerium. So far attempts to broker a compromise on the names for elements 104 and 105 have failed.

The IUPAC has not yet recommended any names. In fact, at an IUPAC meeting in Lisbon in August, there was some pressure for the International Commission on Inorganic Nomenclature not to make any recommendations concerning names at this point. One group putting pressure on the commission is the Committee on Inorganic Nomenclature of the American Chemical Society. Paul Karol of Carnegie Mellon University, who serves on that committee, asserts that some members of the US nuclear chemical community are disturbed by the findings of the TWG and by the absence of a nuclear chemist in the group.

The TWG is standing firmly behind its conclusions. A letter from the TWG follows the comments by the three research groups in the August issue of *Pure and Applied Chemistry*. Stressing that its members had no vested interest in the outcome of its deliberations and that each had devoted long hours of study to the subject, the group declares that it does not intend to engage in point-by-point rebuttals of the objections raised by Ghiorso and Seaborg. The TWG states that "after detailed examination of all the criticisms from Berkeley we do not find it necessary in any way to change the conclusions of our report."

—BARBARA GOSS LEVI

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EXPERIMENTS FIND A RELATIVELY LONG-LIVED ISOTOPE OF ELEMENT 106

There's more to research on heavy elements than simply producing them. Recent experiments continue to explore their properties. In a welcome collaborative effort, researchers from the Joint Institute for Nuclear Research in Dubna, Russia, and from the Lawrence Livermore National Laboratory announced at the Actinides '93 Conference in Santa Fe, New Mexico, in September that they had produced an isotope of element 106 that emits an alpha particle with a particularly low energy within a decay time between 10 and 30 sec. That's long compared with the lifetimes of microseconds to milliseconds that are more typical of the elements at the upper end of the periodic table.

The long-lived isotope, $^{266}106$, is still not as stable as the nuclides expected to populate the long-sought "island of stability," which should exist around atomic number $Z = 110$ and neutron number $N = 180$, according to the most recent calculations. Because of nuclear shell effects, some nuclides in that mass region are expected to live more than a thousand years. But forming nuclei in the region is still a formidable challenge.

Some recent theories have predicted that short of the island of stability, there might be what Rayford Nix of Los Alamos National Laboratory calls a "rock of stability." The important feature of the theories is the inclusion of deformed nuclear shape, that is, an average potential energy surface that is deformed and that gives rise to new shell structure. Calculations incorporating such deformed shells indicate that the potential energy surface contains a dip that stabilizes the nucleus against decay by alpha emission or by spontaneous fission, especially for $Z = 108$ and $N = 162$.

The Dubna–Livermore collaboration, led by Yuri Lazarev and Ronald Lougheed, set out to check that prediction. The group was not able to reach the particularly favorable isotope, $^{270}108$, but it could produce two nearby isotopes of element 106 by bombarding a curium-248 target with neon-22 projectiles accelerated at Dubna's U400 cyclotron. The researchers found six decay chains that they attributed to the alpha decay of the isotope $^{266}106$ and the subsequent spontaneous fission of its daughter nuclide. Their identification was based on establishing genetic links between those decays. The data are consistent with an alpha-decay lifetime between 10 and 30 sec and a branching ratio for spontaneous fission about equal to or less than 0.5. Another new alpha-decaying isotope produced by the Dubna–Livermore team, $^{265}106$, also shows a halflife on the order of a second.

Two sets of calculations based on the deformed-shell model have indicated that $^{266}106$ should have a relatively long halflife. The observed values agree much more closely with the theoretical calculations of a group from the Soìtan Institute for Nuclear Studies in Warsaw, Poland, led by Adam Sobiczewski, than with those of Peter Möller and Nix at Los Alamos. Nix comments that the approaches of the two groups differ only in details and that lifetime estimates can be off by many orders of magnitude. In any case, the longer life of this isotope should facilitate measurements of its other properties.

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