Criteria for New Element Discovery:

by Sigurd Hofmann

Allure and romance are rarely expected in an article presented under the bulky headline "Criteria that must be satisfied for the discovery of a new chemical element to be recognized." However, the members of the Transfermium Working Group (TWG) worked out a most fascinating publication on a difficult subject during the years from 1988 to 1991 [1].

In order to solve problems related to the discovery of transfermium elements, which included claims up to element 109 at that time, IUPAC and the International Union of Pure and Physics (IUPAP) had jointly launched TWG for working out appropriate criteria and rules so that existing claims for discovery could be settled and that in the future the priority of discovering new elements could be decided timely and unambiguously. As a result, the 1991 article on the criteria for discovery of a new element was prepared by A.H. Wapstra as secretary in the name of the TWG, consisting of:

- D.H. Wilkinson (IUPAP; UK), Chairman
- A.H. Wapstra (IUPAP; Netherlands), Secretary
- I. Ulehla (IUPAP; Czechoslovakia), Secretary
- R.C. Barber (IUPAP; Canada)
- N.N. Greenwood (IUPAC; UK)
- A. Hrynkiewicz (IUPAP; Poland)
- Y.P. Jeannin (IUPAC; France)
- M. Lefort (IUPAP; France)
- M. Sakai (IUPAP; Japan)

It is to the credit of the members of TWG that the article became an exciting lecture on the spirit behind research, in particular on the study of new heavy and superheavy elements and nuclei. Two most impressive extracts of the TWG report are presented in the following:

> "The centuries-old history of the definition and discovery of chemical elements has a deep scientific and general fascination. This is because the problem is of an essentially finite scope: there can only be a limited number of species of atomic nuclei containing different numbers of protons that can

Transfermium Working Group visit the Berkeley laboratory, 19-23 June 1989. The photo shows the nine members of TWG and Glenn Seaborg as the host of the group. *Front row: Ivan Ulehla (Czechoslovakia, co-secretary), Denys Wilkinson (UK, chairman), Glenn Seaborg (USA, leader of LBNL), Yves Jeannin (France). Back row: Marc Lefort (France), Norman Greenwood (UK), Andrzej Hrynkiewicz, (Poland), Mitsuo Sakai (Japan), Robert Barber (Canada), Aaldert Wapstra (co-secretary, Netherlands). Jeannin and Greenwood were named by IUPAC, the others by IUPAP. The TWG has held the following meetings, of which the first and last were "private", with the remainder in the laboratories of chief concern: 3-5 February 1988, Nonant (France); 12-17 December 1988, Darmstadt (Germany); 19-23 June 1989, Berkeley (USA); 12-16 February 1990, Dubna (Russia); and 16-20 April 1990, Prague (Czechoslovakia).*

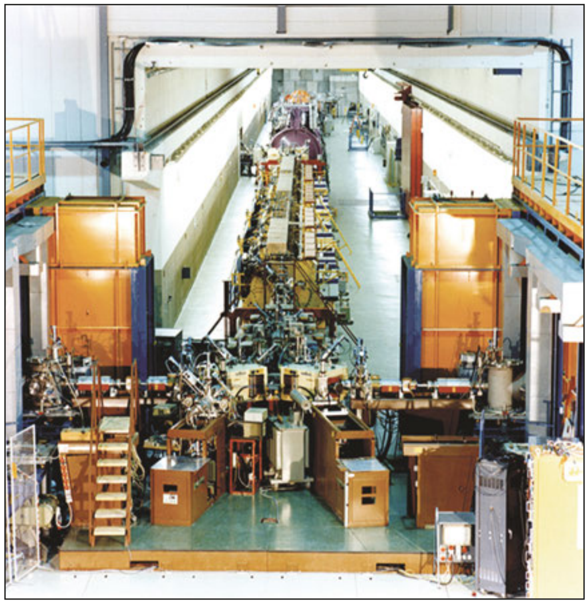
be imagined to have an existence, though perhaps only fleeting, in the chemical sense. But although the problem is of finite scope, we do not know what the scope is: we do not as yet know how many elements await discovery before the disruptive Coulomb force finally overcomes the nuclear attraction. In this sense, the problem is open although of finite scope, unlike the number of continents upon the surface of the earth where we know with certainty that none still awaits discovery. These considerations give to the discovery of new elements an importance, an allure and a romance that does not attach to the discovery of, say, a new comet or a new beetle where many more such discoveries are to be anticipated in the future."

And:

"... the insight that they [the new elements] give into the details of the construction of Nature's most complex nuclear edifices and the laws that govern their construction, explains the great investment of material and, most particularly, human resources into the discovery of new elements. Lives are committed over decades to this enterprise, and this is not surprising. Nor is it then surprising that, although from the point of view of science itself (except that of the "science of history") and the associated advance of human understanding it does not matter who makes the discovery, immense importance is attached, personally, institutionally and nationally, by those engaged in the enterprise, to the public recognition of their discoveries."

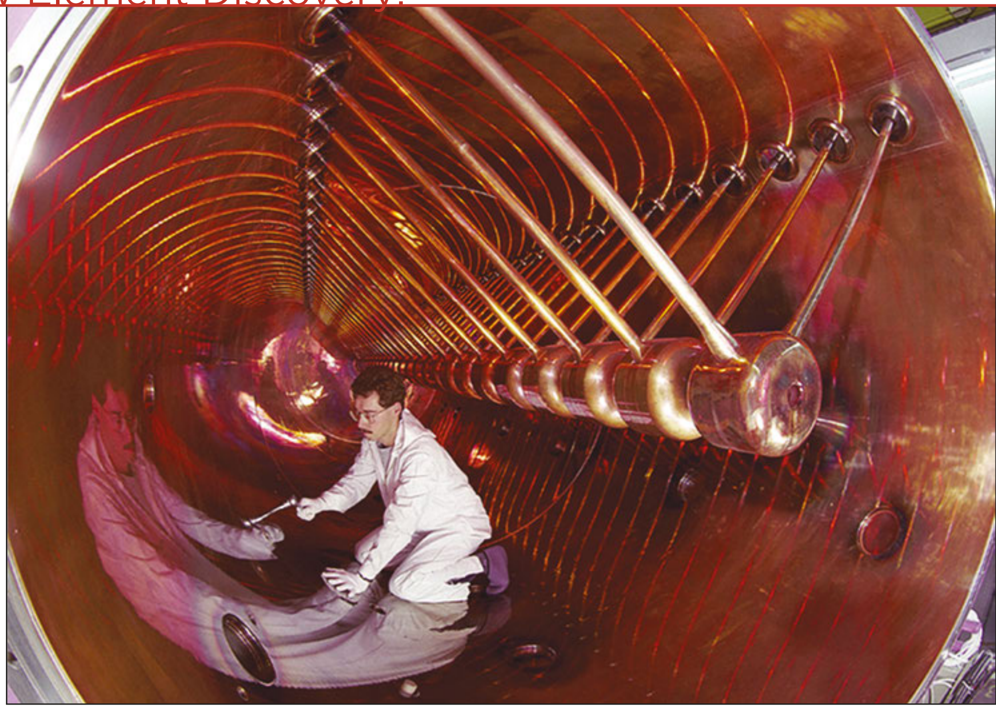
Beyond personal, institutional, and even national controversies, scientific work needs clear and unambiguous data and results. Those related to new elements are obtained in big laboratories having adequate instrumental equipment available. These are necessary prerequisites because all new elements beyond uranium were discovered by observation of the nuclear reactions which produced them. The last eighteen elements were discovered in fusion reactions and the more recent ones in fusion with heavy ions. Now, one may ask why ambiguities arise when this research is performed by outstanding scientists working in highly advanced laboratories with the most sophisticated technical equipment?

The answer is related to the complicated matter, and to the fact that only a few atoms are produced, in extreme cases only one, so that statistical fluctuations are huge. Another complication with identification of the produced nuclei arises through the variety of decay modes and wide range of possible lifetimes. In principle, all known nuclear decays, except radioactive neutron emission, have to be considered for isotopes in the region of the heaviest elements: β^+ or electron capture, β^- and α decay, radioactive proton emission, and spontaneous fission, as well as decays from the groundstate and from isomeric states. On one hand, this variety of decay modes opens a rich field for experimental research as well as theoretical studies. On the other hand they can be a source for errors and incorrect or misleading interpretations. In addition, all these studies are hampered by relatively high backgrounds from various nuclear reactions other than complete fusion. To make matters worse, energetically possible evaporation of reaction neutrons, protons and/or α particles after fusion contributes additional uncertainty.



The "UNIversal Linear Accelerator" (UNILAC) setting at the Gesellschaft für Schwerionenforschung, GSI, in Darmstadt, Germany, where the first beam was generated at the end of 1975.

Furthermore, experimental deficiencies like beam and target impurities, insufficient detector resolution, and electronic disturbances, together with the large statistical fluctuations and sometimes also the application of uncertain theoretical predictions may result in interpretation of a measurement which later cannot be substantiated. The worst that could happen, and indeed happened more than once in the scientific work of laboratories, is that individuals may not resist the temptation to manipulate data. This may be explained by an obsession for recognition as well as a motivation to satisfy the expectations of sponsors.



Inside the UNILAC at the GSI

All these experimental and human imperfections can lead to ambiguities and controversies which do not allow for an immediate recognition of an experimental result as being correct or, in the case of conflicting data, to determine which is correct.

In order to solve the problems related to the assignment of priority of discovery of elements 101 to 109, TWG applied its 1991 criteria to the existing claims of these elements. The result was published in 1993 [2]. The following years saw a number of consultations and exchanging of letters in order to find agreement between IUPAC, IUPAP, and the involved laboratories. At that time this included Lawrence Berkeley Laboratory, (now Lawrence Berkeley National Laboratory (LBNL)), in California, USA, the Joint Institute for Nuclear Research (JINR) in Dubna, Russia, and the Gesellschaft für Schwerionenforschung, (GSI) in Darmstadt, Germany. Eventually, a final IUPAC Recommendations for the names and symbols of these elements were published in 1997 [3]. These names and symbols as they appear now in the Periodic Table of the Elements were also accepted by IUPAP and the involved laboratories.

Discussion on the priority of discovery of elements up to 109 was still ongoing in 1995 and 1996 when discovery of further elements 110, 111, and 112 was claimed by an international collaboration working at the Separator for Heavy Ion Reaction Products (SHIP) at GSI. A review of all the results was presented and published in 2000 [4]. The most recent results of this series of experiments have been published in 2002 [5]. In 1999, discovery of element 114 was announced by international research collaborations working at the energy filter VASSILISSA and the Dubna Gas-Filled Recoil Separator (DGFRS) at JINR. Subsequently, international collaborations working at DGFRS announced claims for discovery of elements 116, 118, 115, 113, and 117 in chronological order in the years up to 2010. The most recent discovery was published in [6]. A summary of all results of the Dubna experiments has been presented in [7]. Another claim on the discovery of element 113 was submitted by a Japanese-Chinese collaboration working at the Gas-filled Recoil Separator (GARIS) at the Institute of Physical and Chemical Research (RIKEN) at Saitama near Tokyo in 2004 [8]. Subsequently, most of the experiments were repeated at the claiming laboratories, in many cases independently at other research centres as well. Most of the results were confirmed.

The claims for discovery of elements from 110 to 118 were investigated by four newly established Joint Working Parties (JWPs) of IUPAC and IUPAP in the years from 2001 to 2016. In six IUPAC Technical Reports, the JWPs assigned priority of discovery of the elements from 110 to 118 on the basis of the criteria published by TWG in 1991. The decisions were accepted by IUPAC, IUPAP, and in particular by the involved laboratories. Discovery of elements 110 to 112 was assigned to the collaboration at GSI, 113 to the collaboration at RIKEN and 114 to 118 to the collaborations at JINR. According to the rules for the 'Naming of New Elements' published as IUPAC Recommendations in 2002 [9] with an update from 2016 [10] these elements received their names as suggested by the discoverers. These names and symbols from darmstadtium, Ds, for element 110 to oganesson, Og, for element 118 are listed in IUPAC's recent Periodic Table of the Elements.

The remarkable growth of research on superheavy nuclei and elements based on the development of

Foundation meeting of the JWG in Egelsbach near Darmstadt, Germany, 20-22 May 2017. Left to right: Sigurd Hofmann (Chair), Sergey Dmitriev, Jacklyn Gates, Natalia Tarasova (2017 President of IUPAC) proudly keeping the Chart of Nuclei in her hands, Bruce McKellar (2017 President of IUPAP), James Roberto, Hideyuki Sakai (Vice Chair), and Claes Fahlander, respectfully holding the Periodic Table of the Elements.

intensive beams of rare but stable isotopes, the use of neutron rich radioactive targets of actinides, and highly sensitive detection methods was difficult to envisage at the time when criteria and rules for assigning priority of discovery of new elements were set up in 1991. Therefore, not all criteria take into account specific advantages or any problems and difficulties adherent to the production and identification methods being in use now.

A new Joint Working Group (JWG) was created by IUPAC and IUPAP at the beginning of 2017 with the task to review the criteria and rules worked out by the 1991 TWG in the light of the experimental and theoretical advances in the field. The new JWG consists of six members:

- Sigurd Hofmann (IUPAC; GSI, Germany), Chairman
- Hideyuki Sakai (IUPAP; RIKEN, Japan), Vice Chairman
- Sergey N. Dmitriev (IUPAC; JINR, Russia)
- Claes Fahlander (IUPAP; Lund University, Sweden)
- Jacklyn M. Gates (IUPAP; LBNL, US)
- James B. Roberto (IUPAC; ORNL, US)

Three members were suggested by IUPAC and three by IUPAP. The restriction excluding the appointment of members from a claimant laboratory which applies to JWPs does not apply to our JWG, which is not evaluating any claims. Two members are nuclear chemists and four nuclear physicists. This reflects that nuclear decay properties are eventually needed for the identification of an isotope, although chemical separation may have been or will be performed. The importance of the mutual dependence of chemistry and physics becomes apparent in cases when radioactive isotopes of actinides are needed for the irradiation. An impressive example is the production and purification of the isotope ^{249}Bk by chemists of the Oak Ridge National Laboratory, Tennessee, USA. This isotope with a half-life of only 327 days was needed for the synthesis of element 117 at JINR. The work of those chemists was honoured with naming this element after the state of their laboratory, tennessine.

Our first meeting with participation of the then respective Presidents Natalia P. Tarasova of IUPAC and Bruce H. J. McKellar of IUPAP took place in Egelsbach near GSI in Darmstadt, Germany, 20-22 May 2017 (see photo). After election of a chairman and a vice chairman, both presidents communicated the Terms of Reference for JWG. A second four-hour meeting was arranged during the 3rd International Symposium on Super-Heavy Elements in Kazimierz Dolny, Poland, on 11 September 2017.

At the first meeting in Egelsbach we exchanged opinions on the subject and discussed a possible layout of our report. Considering the update of the criteria in the context of recent discoveries and expected future search experiments for new elements we felt it also necessary to present a short retrospective on the elaborate work of experimentalists and JWPs according to the Chinese proverb: "*If you want to learn about the future, you have to look into the past.*"

The retrospective research revealed that the task of our JWG differs from that of the TWG functioning during the years 1988 to 1991. At that time, criteria were developed for assigning priority of discovery of

Festive christening of element 112 as copernicium, Cn, at the GSI in Darmstadt, Germany, on 12 July 2010.

elements for which discovery was already claimed up to meitnerium with element number Z = 109. Our present aim was to work out criteria for assigning discovery of new elements beyond oganesson, Z = 118, which may be discovered in the future. Therefore, it was essential to estimate on the basis of known data and theoretical studies which new elements will most likely be searched for, what their decay properties may be, and which methods could be applied for production and identification using present and near-future technology.

The detailed study of the publications claiming discovery of a new element revealed that in general the nature of the experimental data is such that an absolutely secure identification in a first attempt is rarely possible when new regions in the chart of nuclei are explored, where decay chains produced by subsequent α decays are not connected to known nuclei, or the small number of produced nuclei does not allow for a convincing identification by characteristic X-rays. Only the combination of information from various irradiations and measurements results in a secure identification. The term "discovery profile" was already coined for this method of linking results from different obtainable measurements in different laboratories in the 1991 TWG report. This was the procedure from the end of the 1990s and the beginning of the new millennium through which the isotopes of new elements with proton numbers from 114 to 118 could be safely and relatively quickly identified. These isotopes were located in the theoretically predicted and now experimentally confirmed region of spherical superheavy nuclei, also known as the island of stability.

A similar situation may occur in the future when experimentalists will search for new regions of isotopes of new elements beyond the known region of spherical superheavy nuclei at Z = 114 and neutron number N = 184. Such new regions could arise from the stabilizing effect of a next single or double shell closure for spherical nuclei, from a special arrangement of the energy levels for protons and/or neutrons for deformed nuclei, or due to bubble or donut like arrangements of the nucleons.

Identification of new elements is relatively straightforward when the produced isotopes can be identified *via* a sequence of subsequent α decays genetically connected to known daughter products and using known reactions, *e.g.* fusion evaporation reactions, and established identification methods. In this case only one measured decay chain could be sufficient for safely assigning priority of discovery. This is way the first isotopes of the elements from bohrium, Z = 107, to nihonium, Z = 113, were identified.

However, the identification of an isotope using genetic relations is not always straight forward. The assignment of a decay chain genetically linked to a potentially known isotope requires that no other possible candidate with similar properties exists that could be mistaken for the isotope that is believed to be well known. Similarly, in irradiations of a target with heavy ions it is *a priori* not possible to assign a measured decay chain to the product of a fusion reaction and in particular not to an isotope produced by evaporation of only neutrons.

This relatively simple example already reveals the difficulties which can arise with the interpretation of experimental results. In our JWG report we discuss the criteria listed in the 1991 TWG report in the light of the present technical possibilities and improved physical and chemical knowledge obtained from the successful production and identification of isotopes in the region of superheavy nuclei and elements. We discuss in detail various identification methods and point out specifically the experimental problems which can arise and which can hamper the interpretation of the measurements. Also considered are physical properties of the reaction for production and properties of the nuclear decays. All this information is intended as guidance to be considered by experimentalists and future JWPs. It became obvious that our report cannot present a list for checking fulfilled and failed criteria where the number of fulfilled criteria decides on the discovery. It rather reveals that for each criterion its weight and its applicability to a certain experimental result have to be carefully evaluated and that a number of relevant criteria often need to be combined for elaboration of a discovery profile.

We do not consider our report as a review of all experimental and theoretical studies performed in the field since 1991. This information can be found in various review articles and in the specific publications in *Pure and Applied Chemistry*. Study of the large number of publications and reports needed time. Similar to situations in research or other enterprises, the development of ideas and discussion with an exchange of views also needs time.

To develop rules and criteria for safe and timely decisions on priority of discovery of a new element could not be achieved in a few days of meeting. On the other hand, the long distances between our home institutions made frequent meetings difficult. Therefore, we tried to approach an ideal form of communication using email. This way we could finish our report after one year of work. It was sent for further consideration to the Presidents of IUPAC and IUPAP in May 2018 [11].

Our look into the past led us to reflect with admiration on the obtained results in the field of research on superheavy elements and nuclei during the nearly thirty years since 1991. The publications convey not only the pure physical or chemical results but also the enthusiasm and pride of the authors, with uncertainty usually hidden between the lines. We realized that in complicated situations the best way for solving open problems is communication in addition to the application of rules and criteria. This is similar to situations which happen to us in our everyday life. Communication often helps to overcome apparent injustice and to improve collaboration through the exchange of knowledge.

Although nobody can look into the future we believe that exciting work is still ahead of us. Primarily it is the search for isotopes of new elements near and beyond the presently known region of superheavy nuclei and elements but also the detailed study of nuclear, atomic, and chemical properties of isotopes of the already known ones. Exploring the unknown gives us '*... the insight ... into the details of the construction of Nature's most complex nuclear,* atomic, and chemical *edifices and the laws that govern their construction'.*

Finally, I would like to thank my colleagues in the Joint Working Group for a fruitful period of discussion and sharing thoughts and ideas on an important theme of allure and romance.

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