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ON THE DISCOVERY OF THE ELEMENTS 110–112

(IUPAC Technical Report)

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On the discovery of the elements 110–112* (IUPAC Technical Report)

Abstract: The IUPAC/IUPAP joint working party on the priority of claims to the discovery of elements 110, 111, and 112 has reviewed the relevant literature pertaining to the several claims. In accordance with the criteria for the discovery of elements, previously established by the 1992 IUPAC/IUPAP Transfermium Working Group, it was determined that the claim by the Hofmann *et al.* research collaboration for the discovery of element 110 at GSI has fulfilled those criteria. For elements 111 and 112, the collaboration of Hofmann *et al.* produced high-quality data with plausible interpretations, but confirmation by further results is needed to assign priority of discovery for these elements. The working party was not convinced that claims of other collaborations have satisfied the discovery criteria.

INTRODUCTION

In 1998 it was decided that a small working party of four independent experts drawn from IUPAC and IUPAP would be assigned to establish priority of claims to the discovery of elements 110, 111, and 112. The three laboratories primarily involved in the studies were contacted in May 1998 requesting papers relevant to the discoveries, with the understanding that a working party will have been formed to carefully review those materials. In January 1999, members were notified of their nomination to the joint working party (JWP). All accepted, and the next month, the formal charge was issued. Documents submitted by the claimant laboratories were delivered to the group.

The task of the working party was to review the documentation, to make judgment on the priority claims and to report to the two Unions through Prof. John Corish, President of IUPAC's Inorganic Division. The working party's report was requested to be suitable for open publication. It was furthermore suggested that a starting point would be consideration of the criteria for judging such claims as discussed by the prior Transfermium Working Group that considered elements 101–109. Any deficits in documentation could be compensated for by way of the literature or by correspondence.

When the priorities of the claims have been established, the appropriate group for each element would be asked to suggest a name formally.

CRITERIA USED

The "Criteria that must be satisfied for the discovery of a new chemical element to be recognized" (91TWG, 92TWG) established by the IUPAP/IUPAC Transfermium Working Group (TWG) served as a guide. Sections particularly relevant to our deliberations on elements 110–112 are partially reproduced here for convenience. The last sentence, balancing a justifiably conservative stance with the need for reasonable flexibility, is especially germane to our deliberations and has been italicized by us for emphasis. The intent is not to set a higher standard for "discovery" than applies elsewhere in science but rather to conform to a uniform, consistent basis for definitive observation and interpretation.

"Discovery of a chemical element is the experimental demonstration, beyond reasonable doubt, of the existence of a nuclide..."

*Elements 110, 111, and 112, under the IUPAC systematic (provisional) naming system, are referred to as ununnilium, ununium, and ununbium, respectively.

“The TWG realizes that the term ‘reasonable doubt’ is necessarily somewhat vague... Confirmation demands reproducibility... In the case of the new elements the TWG attaches considerable importance to reproducibility and would indeed like to be able to suggest that no new element should be recognized officially until the data upon which the claim is based have been reproduced, preferably in another laboratory and preferably by a different technique. However, it cannot: ...it would appear unreasonable to apply such a demand of demonstrated reproducibility in all rigidity. We do not believe that recognition of the discovery of a new element should always be held up until the experiment or its equivalent have been repeated, desirable in principle as this may be. *However, we would waive this requirement only in cases where the data are of such a nature that no reasonable doubt is possible (for instance for data with a high degree of internal redundancy and of the highest quality), and under circumstances where a repetition of the experiment would imply an unreasonable burden.*”

Our assessments were further influenced by the archive discovery profiles for elements 107, 108, and 109, each characterized by having a very small number of events. All initial claims involved either subsequent confirmation and/or included clear observations of known descendents or production of previously unknown intermediates through cross bombardments.

DISCOVERY PROFILES

We follow the thoughtful guidance, procedures, and format introduced by the TWG in presenting discovery profiles: historical accounts of relevant publications on each element appended by our opinion(s) as to the value of the evidence on the basis of the criteria. Our resources were articles submitted by research groups and laboratories in response to solicitation by IUPAC and also other relevant publications routinely available in research libraries or through modern electronic search techniques. As is customary in scientific analysis, more credence was given to resources that had already been successfully subjected to critical refereeing (see Bibliography). Each profile begins with a reprise of the pertinent 92TWG content, if any. The element atomic number is in boldface followed by enumerated comment labels.

Element Z=110

110; 01-03 (92TWG)

Prior TWG Conclusion: “The Dubna experiment (87Og99) describes interesting preliminary work but is insufficient to give confidence that element 110 has been produced”.

110; 04 The collaboration of Hofmann et al., 95Ho01

The fusion–evaporation reaction using a ^{62}Ni beam on an isotopically enriched ^{208}Pb target produced four chains of alpha-emitting nuclides following the presumed formation of $^{269}110 + n$. The heavy residue is separated from nonfusion residues in-flight by the electromagnetic SHIP velocity filter which spatially localizes, through position-sensitive detectors, the product and its radioactive progeny. Even in the first chain to be measured, the second and third consecutive alpha energies and delay times are in concordance with previously studied ^{265}Hs and ^{261}Sg . The redundancy of the consecutive alpha energies and delay times in the second through fourth chains measured is very reassuring. Even more so is the observation of fourth and fifth alpha particle energies and delay times in the last two chains observed that are in very good agreement with the known properties of descendants ^{257}Rf and ^{253}No .

JWP ASSESSMENT: Element 110 has been discovered by this collaboration.

110; 05 The Berkeley papers 95Gh01, 95Gh02

Essentially contemporaneous with **110; 04** above, evidence for the production of $^{267}110$ was offered by the Lawrence Berkeley Laboratory based on one particular sequence of events resulting from the cold fusion reaction of a bismuth target with a beam of cobalt ions. The gas-filled magnetic spectrometer SASSY2 was employed to separate the fusion recoils and their decay products from other nuclides. The

drawback in interpreting the resulting sequence as convincing evidence for the production of element 110 is the need to postulate unproven steps and to explain unfavorable observations. There was, for example, an unfortunate electronic failure which necessitated use of a detector that was disabled for a fraction of a millisecond. The underperforming electronics becomes an explanation for missing the detection of intermediate ^{263}Hs whose decay mode and half-life are unknown. Confidence in assigning alpha particle energies is diminished by degradation of the resolution in the detection system. Position-sensitive detectors work in favor of the interpretation. Unfortunately, it was not possible to obtain additional beam time.

JWP ASSESSMENT: Although the authors' conjectures that production of the claimed element is the simplest explanation consistent with the observations, the criteria needed—high quality of data and internal redundancy—have not been met.

110; 06 *The collaboration by Lazarev et al., 96La01*

The bombardment of ^{244}Pu by ^{34}S produced, by a hot fusion pathway, one chain of spatially and temporally correlated alpha-emitting nuclides that was assigned to the product $^{273}110$. The evaporation residues were separated in-flight by a recoil separator. We include this example for illustration purposes, but it will also be cited in reference to element 112 below. The investigators interpret their results as a sequence of five alpha-emitting nuclides beginning with $^{273}110$. However, only three of the alpha particles are observed. A number of additional incomplete chains are also noted, but with lesser confidence.

110; 07 *The collaboration by Hofmann et al. 98Ho02*

The bombardment of enriched ^{208}Pb by ^{64}Ni produced $^{271}110$ by the fusion–evaporation process, losing one neutron from the compound nucleus. The SHIP facility was employed, and nine localized chains of alpha emitters were observed and characterized with position-sensitive detectors. This study was very influential in the thinking of the joint working party. The high quality of the data and the internal redundancy in the nine chains served as a benchmark against which similar studies for this element and other elements were compared during the JWP deliberations.

Element Z=111

111; 01-02 (92TWG)

Prior TWG Conclusion: “No data indicating formation of element 111 are available”.

111; 03 *The collaboration of Hofmann et al., 95Ho03*

In bombardments of ^{209}Bi targets with ^{64}Ni using the velocity selector SHIP facility to discriminate in favor of the fused product, $^{273}111$, three sets of localized alpha-decay chains were observed with position-sensitive detectors. The origin was assigned to the isotope $^{272}111$, one neutron removed from the compound nucleus.

Applying criteria to the case of element 111, the data are of the highest quality. However, there is internal redundancy with just two pairs of data. Chains 2 and 3 have mutually concordant alpha energies, but ones ascribed to the previously unknown ^{264}Bh . Chains 1 and 3 also have mutually concordant alpha energies, but these are ascribed to the previously unknown ^{268}Mt . There is no redundancy involving properties of known daughters for verification purposes.

Assignment of members of each full chain, unlike the examples of elements 107–109, is burdened by the profuse need to invoke (uncertain) isomeric states with different alpha energies and different lifetimes. For example, $^{272}111$ decays to two isomeric states of ^{268}Mt ; ^{268}Mt decays to two isomeric states of ^{264}Bh ; ^{264}Bh possibly decays to two isomeric states of ^{260}Db , only one of which agrees well with the known decay of ^{260}Db . ^{260}Db decays to ^{256}Lr , but the sole observation with a concordant decay energy occurred with a time delay of 66 s. Although statistically consistent with the known mean-life (35 s), the value is arguably sufficiently convincing.

For chain 1, two of its four alphas have insufficient energy information and the third alpha is of sufficiently different energy and lifetime compared to chains 2 and 3 to make the sequence assignment fragile.

Chain 2 is most compelling, matching the known ^{260}Db energy and lifetime. Unambiguous observation of its daughter ^{256}Lr in this sequence would have been sufficient to secure the discovery.

Chain 3 has a mismatch in ^{260}Db 's alpha energy compared to event 2; its daughter appears quite late (66 s) compared to the known 35 s mean-life; and the second full alpha energy (10.221 MeV) disagrees with that of event 2's second full alpha energy (10.097 MeV).

JWP ASSESSMENT: The results of this study are definitely of high quality but there is insufficient internal redundancy to warrant certitude at this stage. Confirmation by further results is needed to assign priority of discovery to this collaboration.

Element Z=112

112; 01-03 (92TWG)

Prior TWG Conclusion: "Data reported so far are insufficient to indicate that a new element has been produced."

The work of the Marinov *et al.* collaboration rests on analysis of fission tracks from tungsten targets irradiated with multi-GeV protons. Production of superheavy elements is premised on the interaction of heavy energetic spallation fragments with tungsten target nuclei to produce very long-lived fusion residues. Chemical separations were done, and some mass spectrometry studies were interpreted as evidence of molecular clusters containing element 112. The techniques employed are mostly indirect, few controls or studies of blanks were performed, and the need to invoke a collection of questionable and/or novel, speculative mechanisms is required to buttress the interpretation.

112; 04 The collaborations of Marinov *et al.*, 91Ma01, 92Ma01, 93Ma01

This series of papers continues, with a different set of collaborators, to refer to past evidence for the secondary production of very long-lived eka-mercury ($Z=112$) from the spallation of tungsten with high-energy protons. Chemical separation of mercury is assumed to carry its heavier analog. The specificity of the separation for mercury (its radiochemical purity) is upheld only very indirectly. Mass measurements were performed. That the masses scattered between 308 and 318 necessitates invoking formation of a variety of molecular ions whose production likelihood is never verified through blank runs nor with normal mercury. Interpretation of spontaneous fission tracks requires energies significantly lower than expected for binary fission. The authors accommodate this problem by invoking quaternary fission. A critical review of this collection of papers was very problematic because of the dominant referencing to supportive experimental and theoretical literature by the same author(s). Each reference is very sparing in the presentation of high-quality data yet liberal in confident claims for having proven their points.

JWP ASSESSMENT: The situation pertaining to these collaboration results has not changed substantially since the TWG judgment. If anything, it has become weaker because independent attempts to duplicate the process of fusion with secondary residues from high-energy proton irradiations of heavy targets have failed to find yields of elements more than a half dozen atomic numbers greater than that of the target (71Ka01, 73Ba01, 73Ge01) rather than the three dozen or more invoked by Marinov *et al.*

112; 07 The collaboration of Hofmann *et al.*, 96Ho01

Using the electromagnetic velocity filter SHIP, fusion-like residues of the reaction of ^{70}Zn with enriched ^{208}Pb targets were measured. Two chains of localized alpha-emitters were identified as originating with $^{277}112$. The quality of the data is very high. However, regarding the complete criteria, there is only one incidence of redundancy, that assigned to the previously uncharacterized isotope ^{269}Hs that appears in both events; there is no redundancy involving known daughters.

Redundancy is arguably and unfortunately confounded by the effects of isomerism. The two observed alphas from $^{277}112$ involve different states and lead to yet two other very different decay branch-

es in $^{273}\text{110}$. One of these events leads to the known isotope ^{265}Sg , but no further confirmation with descendants ^{261}Rf nor ^{257}No is observed. The other event does lead to the latter two nuclides, but the ^{261}Rf alpha energy observed is 0.24 MeV too high despite a 0.02 MeV resolution, imposing weakened persuasiveness of the assignment. Its claimed daughter is, however, observed with the expected energy.

The first two alphas in the chains show no redundancy. In chain 1, the ^{265}Sg alpha energy agrees well with the known value, but the appearance delay is a concern because it is more than three times the known half-life of 7.4 s. Event 2 reports a ^{261}Rf alpha energy that is in significant disagreement with known energies, posing uncertainty with the assignment. The last alpha in chain 2 agrees extremely well with that of descendant ^{257}No but is the only concordant daughter comparison event of the entire 112 set.

Reference 96La01 reports its $^{273}\text{110}$ alpha particle with energy 11.35 MeV (resolution .04–.12 MeV) not reassuringly close enough to this group's 11.08 MeV and with more than triple the delay time of the latter. There are suggestions of other nuclides in reference 96La01 noted with lesser confidence but that nevertheless do not provide further redundancy to these results by Hofmann *et al.*

JWP ASSESSMENT: The results of this study are of characteristically high quality but there is insufficient internal redundancy to warrant conviction at this stage. Confirmation by further results is needed to assign priority of discovery to this collaboration.

112; 08 The collaborations by Marinov *et al.*, 97Ma01, 98Ma01

These two papers continue to press arguments for the existence of very long-lived isomeric states of actinides and transactinides and of very high fusion cross-sections for their formation, each several orders of magnitude beyond current understanding. These extraordinary phenomena are, in part, necessary for the acceptance of the collaborations' interpretations. The JWP remained unmoved.

COMMENTS

Again in reference to the criteria previously established, the Transfermium Working Group recognized that there could be a situation in which an early paper did not, at the time, carry conviction of discovery, but that was later recognized to have reported correctly signals from the new element in question. The existence of the element in question is then definitely established by subsequent work following the lead of the early paper. The TWG felt it would clearly be wrong to assign absolute priority to that early paper, but that it would be appropriate to recognize its seminal importance. Note that both the early and later papers referred to could be from the same group, laboratory, or with other possibilities of common authorship. Any future decision motivated by new results should keep this in mind. The joint working party encourages the laboratories to continue to pursue the production and characterization of new elements with the vigor and skill evident in its efforts to date.

The joint working party agreed that it would not be much swayed by arguments that depend to a large extent on statistics of speculative interpretations; that is, in the absence or near absence of identifying properties, if the data are not characterized by quality, clarity, and redundancy, conjectures supported mainly by dismissal of alternatives are not sufficient.

SUMMARY OF JWP99 CONCLUSIONS

The IUPAC/IUPAP joint working party performed a critical review of the various claims to discovery of elements 110, 111, and 112. Experimental techniques involving heavy-ion fusion, fusion product separation with magnetic fields, and position-sensitive alpha measurements are proving highly selective in characterizing extremely rare events. In concordance with the criteria previously established for validating claims, the joint working party has agreed that the priority of the Hofmann *et al.* collaboration's discovery of element 110 at GSI is acknowledged. There has been no intent by the JWP to convey invalidation of any of the results. Resourceful work done at Berkeley is certainly consistent with possible discovery of element 110, but nevertheless is unable to fulfill the criteria sufficiently. The uncontested

claim by collaborations using the GSI facility for discovering element 111 and their claim for discovering element 112, although designed to yield the characteristically high-quality results, are not yet sufficient in themselves to be deemed certain owing to still-needed redundancy. Despite the additional papers submitted to us, we endorse the conclusion of the Transfermium Working Group that a persistent competing claim for element 112 from the Marinov *et al.* collaboration, (involving secondary reactions following proton spallation of high-*Z* targets) remains unconvincing.

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