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ON THE CLAIMS FOR DISCOVERY OF ELEMENTS 110, 111, 112, 114, 116, AND 118*

(IUPAC Technical Report)

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*Elements 111, 112, 114, 116 and 118 under the IUPAC systematic (provisional) naming system, are referred to as unununium, ununbium, ununquadium, ununhexium, and ununoctium, respectively. Element 110 has recently been given the name and symbol "darmstadtium, Ds".
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On the claims for discovery of elements 110, 111, 112, 114, 116, and 118

(IUPAC Technical Report)

Abstract: The IUPAC/IUPAP Joint Working Party on the priority of claims to the discovery of new elements has reviewed the relevant literature pertaining to several claims. In accordance with the criteria for the discovery of elements, previously established by the 1992 IUPAC/IUPAP Transfermium Working Group, and reinforced by the 1999 IUPAC/IUPAP Joint Working Party, it was determined that the claim by the Hofmann et al. research collaboration for the discovery of element 111 at Gesellschaft für Schwerionenforschung (GSI) has fulfilled those criteria. For elements 112, 114, and 116, the collaborations of Hofmann et al. and of Oganessian et al. produced high-quality data with plausible interpretations. However, confirmation by further results is needed to assign priority of discovery for these elements. The working party was not persuaded that other collaborations have satisfied the discovery criteria.

INTRODUCTION

The working party of four independent experts drawn from IUPAC and IUPAP that assigned priority of claims to the discovery of elements 110, 111, and 112 was asked to continue its efforts by addressing more recent results. As done in the previous review, the three laboratories primarily involved in the studies were contacted in August 2001 requesting papers relevant to the discoveries with the understanding that a working party will have been formed to carefully review those materials. The deadline was set as December 2001 for material submitted by claimants.

The task of the working party was to review documentation, to make judgments on the priority claims, and to report to the two Unions through Prof. John Corish, former President of IUPAC's Inorganic Division.

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

CRITERIA USED

The article "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) continued as a guide. Sections particularly relevant to our deliberations on elements 110–112 are partially reproduced here for the readers' convenience. The final sentence, balancing a sensibly conservative stance with the need for reasonable flexibility, continues to be 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...
> 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.*"

As an outcome of previous deliberations, the Joint Working Party agreed that it would not be much swayed by arguments that depend to a fragile extent on statistics of speculative interpretations; that is, in the absence or near absence of unambiguous identifying properties, if the data are not characterized by quality, clarity, and redundancy, conjectures supported mainly by dismissal of alternatives are not sufficient.

DISCOVERY PROFILES

We continue to follow the 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 December 2001 by research groups and laboratories in response to formal solicitation by IUPAC and also other relevant publications routinely available in research libraries or through modern electronic search techniques. These include three bibliography entries dated early 2002. As is customary in scientific analysis, considerably 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 and 01JWP content, if any. The element atomic number is in boldface followed by enumerated comment labels.

Element 110, darmstadtium, Ds*

110; *01 (01JWP)*

Prior JWP conclusion: "Element 110 has been discovered by the collaboration of Hofmann et al., 95Ho01."

110; *02 The Hofmann et al. collaboration, 95Ho01*

The 2001 Joint Working Party summarized the effort with the following description: "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}Ds + n. The heavy residue is separated from non-fusion 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

*The name darmstadtium and symbol Ds for the element with atomic number 110 were approved by the IUPAC Council during its session on 16 August 2003. The official IUPAC recommendations are published in *Pure Appl. Chem.* **75** (10), 1613–1615 (2003).

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."

110; *03 Hofmann et al. (02Ho01*)*

In a similar, but more recent collaboration working on elements 111 and 112, Hofmann et al. noted that one of the four $Z = 110$ chains, the second, could not be reconstructed from the original data. The extant chains remain persuasive.

JWP ASSESSMENT: Priority of element 110 discovery by this collaboration has not changed.

110; *04 The Lazarev et al. collaboration, 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 which was assigned to the product ^{273}Ds. The evaporation residues were separated in flight by a recoil separator. The investigators interpret their results as a sequence of five alpha-emitting nuclides beginning with ^{273}Ds. However, only three of the alpha particles are observed. A number of additional incomplete chains and one incomplete 5-α chain are also noted, but with less confidence. There remains the question as to whether subsequent studies might confirm the observed steps in these incomplete chains. Although the referenced chain's ^{273}Ds alpha energy agrees with that seen in the recent Hofmann et al. collaboration (02Ho01), the subsequent two observed alphas have delay times at sharp variance with literature values for ^{265}Sg and ^{257}No.

Element 111

111; *01 (01JWP)*

Prior JWP assessment: "The results of the collaboration of Hofmann et al. 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."

111; *02 The Hofmann et al. collaboration, 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 the evaluation 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.

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.

111; *03 The Hofmann et al. collaboration, 02Ho01*

The Hofmann et al. collaboration reports* on three new chains originating from ^{272}111 in the ^{64}Ni + ^{209}Bi reaction. This brings the number of events to six, three of which proceed through the known descendants ^{260}Db and ^{256}Lr with decay property agreement. The latter in toto provide a strong linkage for the third, fourth, and sixth chains despite some scatter in ^{264}Bh alpha energies and an incomplete alpha determination from ^{272}111 in the fourth chain.

JWP ASSESSMENT: Priority of discovery of element 111 by the Hofmann et al. collaboration in 95Ho03 has now been confirmed owing to the additional convincing observations in 02Ho01.

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*02Ho01 was received as a refereed preprint for publication in 2001.

Element 112

112; 01 (01JWP)

Prior JWP assessment of 96Ho01: "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; 02 The Hofmann et al. collaboration, 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. (The first chain has subsequently been eliminated from contention following re-analysis 02Ho01; see below.) The observed alpha leads to the known isotope ^{265}Sg, but then reports a ^{261}Rf alpha energy that is in significant disagreement with known energies, posing uncertainty with the assignment. The last alpha in the chain 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}Ds alpha particle with energy 11.35 MeV in agreement with this Hofmann et al. group's 11.45 MeV when resolution is taken into account. There are other chains in reference 96La01 noted with less confidence, but that nevertheless do not provide further redundancy to these results by Hofmann et al.

112; 03-04 The Hofmann et al. collaboration, 02Ho01

The Hofmann et al. collaboration 02Ho01 reports on one new chain originating from ^{278}112 in the ^{70}Zn + ^{208}Pb reaction. However, re-analysis of the previous 96Ho01 two chains was unable to confirm the original results for the first chain as noted above. Hofmann et al. thus have a total of two alpha and lifetime chains of events originating with the new isotope ^{277}112 leading to two alpha and lifetime events of ^{273}Ds, in good agreement with a single observation assigned to ^{273}Ds by Lazarev et al. 96La01 in their tenuous chain.

The chains continue to the previously unknown ^{269}Hs for which alpha energies and lifetimes are similar in both events. The Hofmann et al. chains continue through ^{265}Sg, but with no alpha energies. The lifetimes seen are consistent with 8 s from a previous study of ^{265}Sg but not with 158 s found in 96La01. Only the first chain of Hofmann et al. resumes, producing ^{261}Rf with an 8.52 MeV alpha and 4.7 s lifetime. The alpha energy is in agreement with the literature value, but the lifetime is not. This situation is also somewhat confounded by the study of ^{261}Rf in 00La01. Here, 8.30 MeV (but no 8.52 MeV) alpha particles are observed for which the lifetime is acknowledged to be 78 s. As mentioned previously, the first ^{277}112 chain ultimately decays through an alpha and lifetime in very good agreement with that for the known isotope ^{257}No.

In summary, though, there are only two chains, and neither is completely characterized on its own merit. Supportive, independent results on intermediates remain less than completely compelling at this stage.

112; 05 (99Og01 and 99Og02)

The collaborations of Oganessian et al. used the reactions ^{48}Ca + ^{242}Pu and ^{48}Ca + ^{244}Pu, respectively, each to make one observed chain originating with ^{287}114 and ^{289}114, respectively, which pass through unknown intermediates terminating in spontaneous fission.

112; 06 (99Og03)

The Dubna collaboration of Oganessian et al. used the reaction ^{48}Ca + ^{238}U to make ^{283}112 in which the two events decay by spontaneous fission with a lifetime of ~2 min. An independent repeat of the same experiment, however, did not lead to any events (02Lo01) nor did another independent attempt to follow the eka-Hg chemistry of element 112 produced by the same path lead to any events (01Ya01).

112; *07 The Oganessian et al. collaborations, 00Og01 and 00Og02*

The Oganessian et al. collaborations, in which ^{244}Pu + ^{48}Ca in the first and ^{248}Cm + ^{48}Ca in the second case are used to produce decay chains commencing with ^{288}114 or ^{292}116, respectively, followed by a pair of well-reproduced α-decays assigned to the otherwise unknown ^{284}112 and terminating in spontaneous fission of previously unknown ^{280}Ds. The decay energies and lifetimes of three events for ^{284}112 are internally redundant, but no link to recognized nuclei occurs. In the ^{244}Pu + ^{48}Ca study, another event originating with ^{289}114 followed by a chain observed through ^{285}112 and ^{281}Ds terminates with spontaneous fission at ^{277}108, all previously unknown. The experiment in 00Og02 is discussed further in 01Og01 and 01Og02.

JWP ASSESSMENT: The Dubna collaborations have performed careful, high-quality studies whose acknowledgement as discoveries is unfortunately not yet warranted because of unsecured connection to known descendents and unobserved elemental signatures (such as well-resolved X-ray energies).

112; *08 The Marinov et al. collaborations, 96Ma01, 96Ma02, 97Ma01, 98Ma01, 99Ma01, 00Ma01, 01Ma01, 01Ma02, and 01Ma03*

This collaboration continues to press arguments favoring their discovery of element 112 through the existence of very long-lived isomeric states of actinides and transactinides, of very high fusion cross-sections for their formation, each several orders of magnitude beyond traditional understanding. As indirect evidence, their discovery of long-lived ^{236}Bk and ^{236}Am more than a decade ago is frequently cited in their papers, yet the several existing compendia of isotopes do not acknowledge the existence of these species. The collaboration results include mention of observing long-lived proton-decay, of deformed spallation products undergoing secondary fusion reactions, and of hyperdeformed shapes, any of which significant topics by themselves should have attracted studies by other groups years ago. Yet this has not occurred. The collaboration's arguable use of forceful expressions such as "overwhelming evidence", "clear and proven", and "impossible to refute" is neither convincing nor swaying. Extraordinary intriguing phenomena, not much selective in their measured character, are, in part, necessary for the acceptance of the collaborations' interpretations of their data. The JWP needs much more to be able to relinquish its deeply felt unease that the tautological rationalization of the Marinov et al. measurements remains inadequate.

JWP ASSESSMENT: The JWP is unconvinced.

112; *09*

For completeness, we take note of the Berkeley collaboration study of ^{86}Kr + ^{208}Pb by Ninov et al. 99Ni01. Three chains terminating in ^{269}Sg enabled observation of intermediate ^{281}112 in 99Ni01. Retraction of results appear in 02Ni01.

Element 114

114; *01 (99Og01 and 99Og02)*

The Oganessian et al. collaboration used the reactions ^{48}Ca + ^{242}Pu and ^{48}Ca + ^{244}Pu to make one observed chain, respectively, each originating with ^{287}114 and ^{289}114, respectively, which pass through unknown intermediates and terminate in spontaneous fission.

114; *02 The Oganessian et al. collaboration, 00Og01 and 00Og02*

From ^{48}Ca + ^{244}Pu, two chains originating with ^{288}114 end in spontaneous fission of ^{280}Ds after passing through observed intermediate ^{284}112. Additionally, from ^{48}Ca + ^{248}Cm, one chain ending in spontaneous fission of ^{280}Ds passes through observed intermediate ^{288}114.

***114;** 03 The Oganessian et al. collaboration, 00Og01 and 00Og02*

The Oganessian et al. collaborations in which ^{248}Cm or ^{244}Pu + ^{48}Ca are used to produce decay chains commencing with ^{292}116 or ^{288}114, respectively, followed by a pair of well-reproduced α-decays assigned to the otherwise unknown ^{288}114 and ^{284}112 and terminating in spontaneous fission of previously unknown ^{280}Ds. The decay energies and lifetimes of three events for ^{288}114 are internally redundant, but no link to recognized nuclei occurs. In the ^{244}Pu + ^{48}Ca study, one chain originating with ^{289}114 followed by a chain observed through ^{285}112 and ^{281}Ds terminates with spontaneous fission at ^{277}Hs, all previously unknown.

JWP ASSESSMENT: The Dubna collaborations have performed careful, high-quality studies whose acknowledgement as discoveries is unfortunately not yet warranted because of unsecured connection to known descendents or of no observed elemental signatures (such as well-resolved X-ray energies).

***114;** 04*

For completeness, we note the Ninov et al. Berkeley collaboration report 99Ni01 on the reaction ^{86}Kr + ^{208}Pb leading to observation of three chains terminating in ^{269}Sg that pass through observed intermediate ^{285}114. Retraction of results appear in 02Ni01.

Element 116

***116;** 01 The Oganessian et al. collaborations, 00Og01, 00Og02, and 01Og02*

The Oganessian et al. collaborations all report on one event in which ^{248}Cm + ^{48}Ca is used to produce a decay chain commencing with ^{292}116, followed by well-reproduced α-decay sequence assigned to the otherwise unknown ^{288}114 and ^{284}112 and terminating in spontaneous fission of previously unknown ^{280}Ds. The decay energies and lifetimes of three events for ^{288}114 are internally redundant, but no link to recognized nuclei occurs.

JWP ASSESSMENT: The Dubna collaborations have performed careful, high-quality studies whose acknowledgment as discoveries is unfortunately not yet warranted because of unsecured connection to known descendents or of no observed elemental signatures (such as well-resolved X-ray energies).

***116;** 02*

For completeness, we note the Ninov et al. Berkeley collaboration report 99Ni01 on the reaction ^{86}Kr + ^{208}Pb leading to the observation of three chains terminating in ^{269}Sg and passing through observed intermediate ^{289}116. Retraction of results appears in 02Ni01.

Element 118

***118;** 01*

For completeness, the Ninov et al. Berkeley collaboration report on the reaction ^{86}Kr + ^{208}Pb in which three chains are observed to commence with ^{293}118. Retraction of results appears in 02Ni01.

COMMENTS

Elements 110 and 111 have been reviewed and assigned priority of discovery despite lack of independent confirmation of the isotopes observed. These decisions are justified not only on the basis of the quality of the work and reproducibility, but especially on the fact that previously characterized nuclides were identified as part of the detection sequence(s). In the absence of such cross-checks, as happens with more isolated regions of the nuclide chart reviewed in much of the current round of results, the JWP feels the conservative stance of waiting for independent experimental confirmation is both prudent and defendable as experience has shown.

The JWP is also very aware that the highly likely existence, if not prevalence, of isomeric states confounds the ease of reproducibility. Lack of redundancy, nevertheless, is often consistent with explanations that incorporate reasonable alternative decay details. However, the JWP remains convinced that consistency—a sound pretext for lack of replication—is presumptuous and inadequate as support of discovery.

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. Overlap with the prior results or fully characterizing the identity of a descendent in a chain are among the types of coparticipation that would need to be carefully taken into account. The TWG felt it would clearly be wrong to assign absolute priority to that early paper, but that it would be appropriate to weigh 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 JWP 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.

SUMMARY OF JWP01 CONCLUSIONS

The IUPAC/IUPAP Joint Working Party performed a delicate critical review of the various claims to discovery of elements 111 and above and revisited, by necessity, and reaffirmed the previously accepted issue of element 110. Experimental techniques involving heavy-ion fusion, fusion product separation, and position-sensitive alpha-particle measurements continue to improve in their selectivity for these extremely rare events. In concordance with the criteria established for validating claims, the JWP has agreed that the priority of the Hofmann et al. collaboration's discovery of element 111 at GSI is acknowledged. Evidence for the production of elements 112, 114, and 116 is very encouraging but presently unconfirmed. As before, these decisions are of a delicate nature. Observation of element 118 has been retracted by the original investigators. It must be recognized that there is no intent by the JWP to suggest invalidation of any of the other results. Also, despite efforts by the Marinov et al. collaboration using atypical studies in conjunction with provisional theory to reinforce their claim to element 112, we conclude that the results of secondary interactions involving hyperdeformed products of long lifetime and high production probability remain unconvincing, all aspects of which warrant more selective investigation.

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