INTERNATIONAL UNION OF PURE AND APPLIED CHEMISTRY

in conjunction with

INTERNATIONAL UNION OF PURE AND APPLIED PHYSICS

Criteria that must be satisfied for the discovery of a new chemical element to be recognized*

DISCOVERY OF THE TRANSFERMIUM ELEMENTS

PART II: INTRODUCTION TO DISCOVERY PROFILES
PART III: DISCOVERY PROFILES OF THE TRANSFERMIUM ELEMENTS

being the report on the concluding phase (phase ii) of the operations of the TRANSFERMIUM WORKING GROUP OF IUPAC AND IUPAP

Membership of the Transfermium Working Group was as follows:

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^{*}For Part I see Pure & Appl. Chem., Vol. 63, No. 6, pp. 879-886, 1991

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Discovery of the transfermium elements

Abstract

In 1985 IUPAP and IUPAC decided to establish a Transfermium Working Group to consider questions of priority in the discovery of elements with nuclear charge number Z>100. The membership of the Group was determined by the Unions in 1987. The Group met seven times for approximately one week each, three of the meetings being in the Laboratories of chief concern namely those at Berkeley, Darmstadt and Dubna. The work of the Group was carried out in two phases. Phase (i), the establishment of criteria that must be satisfied for the discovery of an element to be recognized, did not concern itself with individual cases or with priorities; it was carried out in close consultation with the Laboratories. The report on Phase (i), Part I of the present paper, was accepted by IUPAP and by IUPAC in 1990 and published separately (Pure and Appl. Chem. 63 (1991)879-886). The report on Phase (ii), the judgemental phase of the work of the Group, was accepted by the IUPAP Council in Madrid, Sept. 1991, and approved for publication by the IUPAC Bureau in Hamburg, Aug. 1991 and forms Parts II and III of the present paper. It completes the work of the Group by applying the criteria of Part I on an element-by-element basis; it considers and analyzes all the pertinent literature and discusses in chronological and critical detail those papers considered important for the building up of confidence that each element had been put in evidence. This delineation of discovery profiles results, in some cases, in a sharing of the credit for discovery.

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PART II: INTRODUCTION TO DISCOVERY PROFILES

II.1 INTRODUCTION

In part I of this report, being an account of Phase (i) of our operations *), we have set out and discussed criteria, and the interplay between those criteria, of relevance for the discovery of new elements. We have also explained in section I.11, that such a discovery is not always a single, simply identifiable event or even the culmination of a series of researches in a single institution, but may rather be the product of several series of investigations, perhaps in several institutions, perhaps over several years, that has cumulatively brought the scientific community to the belief that the formation of a new element had indeed been established. However, since different sections of the scientific community may have different views as to the importance and reliability of interpretation of different sorts of scientific evidence, the bringing into that belief of these different sections of the community may well occur at different times and at different stages of the accumulation of the evidence. Where, then, does discovery lie?

We must, indeed, recognize that the very sense of the word "discovery" depends on the context in which it arises and, furthermore, that different persons may attach different significance to it in the same context. This is certainly true for our present study of the discovery of new heavy elements as we now illustrate by some fictional examples supplementing those already given in Section I.12 of Part I.

The problem of discovery is complicated by the fact that confidence in the interpretability of certain sorts of data or certain forms of evidence, itself changes with time, either as experience grows or as theoretical understanding deepens. For example, evidence adduced by Group A may not, at that time, have been considered compelling in respect of the formation of the new element and may have been overtaken by later evidence of a different sort, adduced by Group B, that was considered to be immediately compelling so that it was then said that Group B had discovered the element; but at a yet later time understanding of the science lying behind the work of Group A had improved to the degree that, retrospectively, the claim of Group A could be accepted with full confidence, neither the evidence nor its interpretation having changed but only our confidence ¹). Where then does "discovery" lie? Does it remain with Group B or is it transferred to Group A?

A more extreme case is that of a Group X who made a certain observation, with great detail and accuracy, but completely misinterpreted it, claiming that it proved the formation of element E, with which, in fact, it had nothing to do. But later, another Group proved the formation of element F with properties such that it was clear that it was that element that Group X had seen. Again, where then does discovery lie? There is no doubt that Group X had "discovered" element F in the sense of having synthesized it and correctly determined its properties, but had not only not claimed that discovery but had, indeed, falsely claimed a different discovery. (We note that in the year of publication of this Report we shall be celebrating the 500th anniversary of the "discovery" of North America by Christopher Columbus.) An intermediate case is that of Group Y which made an observation that they neither understood nor made the basis of any claim but which was later regarded, with certainty, as the signal of a new element - do we then say that Group Y had discovered that element?

A further consideration of relevance to the assignment of credit concerns the suggestion of an appropriate method and its actual development: Group M proposes a new method for the synthesis of heavy elements and demonstrates its utility in known cases; Group N then applies this method to the discovery of new elements. Should the credit for those discoveries then lie wholly with Group N; or should some share of the credit attach to Group M whose method had made them possible?

We pose these questions, supplementing that raised in I.12, to illustrate the many ambiguities that surround the use of the word "discovery" and to reiterate our conviction, expressed in I.11 and I.12, that the correct procedure is the careful and critical delineation of discovery *profiles*, accompanying

^{*)} Published in Pure and Appl. Chem. 63(1991)879

¹) We emphasize that this scenario, and those that follow, are not to be read as historical cases, with the identity of actual groups hidden behind the letters, but rather as fictional illustrations of the range of problems that might arise in the allocation of "credit" for discovery.

them with some opinion of our own as to the relative importance of the various steps along the way and therefore, by implication, as to the appropriate apportionment of credit in the final "discovery".

In these discovery profiles we will give an historical account of relevant publications on each element and give our opinion as to the value of the evidence that they present, on the basis of the criteria discussed in Part I. When this approach leads, in our view, to a clear priority, we shall say so, but that will not always be the case.

In our assessment and conclusions we will take into account that, as we mentioned in I.8, in earlier times the discovery of an element was sometimes proposed and accepted on the basis of evidence that would not fully meet the criteria of today. It goes without saying that such data, in order now to be accepted as of continuing validity, must not disagree unreasonably with data found in later work now judged to be more dependable.

We must also consider the fact that some of the first descriptions of work on new elements were not presented in refereed journals of international standing, for which, as stated in I.II.8, we have a strong preference, but in laboratory reports and such. The situation could be especially critical for conference proceedings and for the Dubna JINR reports. In cases where this factor is of importance, we will discuss the consequences.

Our procedure is the following. We will discuss, for each element under consideration, all relevant papers at least up to the time when, in our view, "full" confidence was reached. For each paper (or group of connected papers), we will first give a factual description of the relevant experimental data and of the conclusions drawn by the authors. Factual comments based on later knowledge will then be added. Finally, in the TWG Assessment, we will give our estimate of the significance of the paper, and our reasons for that estimate.

After consideration of all relevant papers for each element we will give our overall Conclusion in which we delineate the key steps on the road to discovery and indicate our view as to their relative importance. We realize that, as we have indeed insisted, there can be no complete objectivity in these matters and that both our Assessment of the individual papers and, most particularly, our overall Conclusion can be only our opinions. They are, however, opinions that have been arrived at with great concern and after protracted consideration. As we reported in I.5, our work for Part I occupied five meetings of several days each, the series beginning and ending with a completely "private" meeting, and with the three meetings in between in the laboratories of chief concern. Since the acceptance of Part I by IUPAP and IUPAC, we have held two further "private" meetings:

3-7 December 1990 Amsterdam(Netherlands) 1-5 July 1991 Krakow (Poland).

Our work in Phase (i), reported above as Part I, was carried out in intimate consultation with the laboratories of chief concern and our Part I report was finalized by us only after its submission to those laboratories, solicitation of their comments and receipt of their essential agreement to its chief tenets. Our work in Phase (ii), here reported as Parts II and III, in contrast, has had no element of iteration with the chief laboratories and our Assessments and Conclusions have not been discussed with them in any form or manner. Our sole contact with those laboratories in Phase (ii) has been in our addressing to them certain factual questions and in seeking elucidations. We have also, in both phases, consulted external experts to aid our understanding at a few points.

We have carried out what we believe is an exhaustive study of the relevant primary literature, but give below only condensed accounts. More elaborate expositions about the new elements are given for elements 101-104 in 89F199 and 91Zv99, for 104-105 in 87Hy99, for 107-109 in 89Ar99 and 88Mu99 and for all in 87F199. The TWG emphasizes that, especially where its assessments deviate from those given there, and in the many other review papers by various workers in the field, the other arguments have been considered very carefully.

¹⁾ We recognize that there are those whose demand for certainty goes beyond the "reasonable doubt" with which we shall concern ourselves here and who may require, for example, the sure identification and measurement of the energy of X-rays before granting the accolade of discovery. We do not take this extreme position but, of course, welcome the confirmation that such an observation, the final "nail in the coffin", would bring and shall note it, where it exists, in our profiles.

We reiterate the hope expressed in I.9 that our work will lead to the submission to IUPAC, by those most directly involved, of proposals, joint ones where appropriate, for a name for each element for which no name has yet been recommended by the IUPAC Commission II.2 on Nomenclature of Inorganic Chemistry.

In concluding its task, the TWG would like to pay tribute to the tremendous dedication and personal commitment of so many people to the quest for the very heavy elements and it also wishes to recognize and to applaud the substantial material investment that institutions have made to the furthering of that quest. Over the many years that our survey has covered, approaches to the quest have changed as it moved to heavier and heavier elements: from neutron capture, via heavy ion reactions with the heaviest targets, to cold fusion reactions with the most stable targets. And the technologies have also changed with the times from primarily chemical methods to, latterly, primarily physical uses of kinematical filters and separators. But through it all the human dimension has remained invariant: we have been immensely impressed by the integrity and devotion that we have found wherever our meetings and discussions have taken us. Inevitably, as whenever science moves into new territory, where new grounds can be won only by the most painful and protracted effort, many attempts have proved disappointing, inconclusive or fruitless and we have been acutely conscious of the ever-present temptation, in such cases, to wring more from such hardly-gained data than those data are truly able to yield. It is indeed the case that, in this field, it is often not so much the facts that are in dispute and that we have had critically to examine, as the interpretation that is placed upon those facts; our task in these circumstances has been not so much to weigh the facts as to weigh their interpretation. The following pages record in detail how that weighing has been carried out.

We recorded in Part I our appreciation of the warm and open discussions that we have enjoyed in all three laboratories of chief concern. We here wish additionally to thank those laboratories for their welcome and hospitality. We also thank those institutions that have been generous in their support of our three "private" meetings subsequent to the completion of our visits to the laboratories: the Charles University in Prague, the Foundation for Fundamental Research of Matter in Utrecht for the meeting in the Amsterdam National Institute for Nuclear and High Energy Physics NIKHEF, and the Henryk Niewodniczanski Institute of Nuclear Physics in Krakow.

II.2 SOME SCIENTIFIC CONSIDERATIONS

Before presenting our discovery profiles, it may be helpful to make a few comments upon certain possibly less-familiar scientific considerations that are important for an understanding of the subsequent discussions.

II.2A Half-life determination

As we discussed in Section IV of Part I, half-life is only very rarely a positive assignment property, although it is often useful in limiting the range of possibilities for the assignment of the decaying body. However, in the context of our discovery profiles we shall often have to ask the question as to whether a certain reported half-life value can be or cannot be admitted as consistent with another. The half-lives in question may have been measured following different reactions, perhaps by observation of different decay modes and perhaps under conditions in which effects of a chemical or mechanical nature might additionally have supervened in a manner leading to a change in one or both of the apparent half-lives. These problems are the most severe when, as is often the case with work in the very heavy elements, the statistics are poor, when other, possibly unknown activities may have been present and when the background is poorly determined, if at all. We find that authors are sometimes over-optimistic in the accuracy that they attach to measurements made in such multiply-adverse circumstances. In such cases our policy is to be as liberal as seems warranted and not to restrict our judgements by the errors quoted by the authors themselves if we feel that there is good reason to believe that effects of the kind that we have just indicated might have been at work and have not been adequately considered. Of course, with this liberal attitude must also go very critical consideration of the provenance of the reported signals in the sense of our being sure that they were not due entirely to background or to some unidentified and irrelevant substance. In addition to our sensitivity to the above effects we have carefully studied the many statistical approaches to the extraction of half-lives from small numbers of events and to the testing of given event samples for consistency or otherwise with a prescribed half-life.

II.2B Backgrounds due to target impurities

Owing to competition with direct fission, the cross sections for the formation of transfermium elements are low. Therefore, even minute quantities of impurity can lead to the production of unwanted backgrounds of radioactive nuclides.

Detection of transfermium isotopes is made exclusively by measuring fragments from spontaneously fissioning nuclides with short half-lives or by α -particles of relatively high energy and, for these energies, relatively long half-lives.

Spontaneously fissioning nuclides are not formed in bombardments with relatively light ions (up to Ne) on possible contaminants (up to ²⁰⁹Bi and Pb) in actinide targets. But in the attempt to make very high Z elements in the bombardment of Th or U with ⁴⁰Ar or ⁴⁸Ca ions, lead or bismuth impurities could lead to the formation of short-lived SF isomers. Such isomers can, however, also be formed by massive transfer reactions on the U or Th nuclei.

Originally, it was thought that detection of α particles would also have a unique signature for transfermium nuclides. The α -particle energies expected for them are high, and the resulting half-lives far longer than those for relatively unhindered α -decays of lighter elements, with correspondingly smaller Coulomb barriers. Already in early Moscow experiments on bombardment of plutonium with 16 O ions, quite high energy α -particles were found connected with relatively long half-lives due to interactions with Pb impurities (58Fl99) (for references see Bibliography), and a study of these activities was made. High spin isomers with highly hindered α -transitions were found to occur: 25 s 211m Po (E_{α} 8885 keV, 7%, 7275 keV 91% and some weaker branches) and 45 s 212m Po (11635 keV,97%, 9098 keV,1.4%, 8523 keV, 1.9%), both formed by massive transfer on Pb isotopes.

In addition, some short-lived high-energy α -particle emitting nuclides can occur with much larger apparent half-lives because they are in radioactive equilibrium with longer-lived ancestors. Examples are 2 μ s ²¹⁴At (E_{α} =8802 keV) following a 66 s isomer ^{222m}Ac formed in (Cm+Pb) + ¹⁸O bombardments (73Si40) and, rather surprisingly, 20 ms ²¹³Rn (E_{α} =8090 keV) following a small (0.55%) electron-capture branch of 34.7 s ²¹³Fr formed in (Am+Pb) + ¹⁵N bombardments (77Be09).

It is evidently necessary to be critically sensitive to the possible influence of such impurities.

II.2C Quantitative cross section comparisons

In looking for consistency between cross sections for the same reaction, under similar conditions of bombardment, reported by different groups, we recognize the very considerable difficulties that attend the making of such comparisons. For example, the reaction yield may be measured by one group under the tight geometry, extended over several metres, of a magnetic/electrostatic maze following a thin target bombarded by particles of well defined energies, and by another group using purely geometrical baffles with substantial solid angles, over only a few centimetres, behind a thick target thus causing a spread of energies of the reacting particles. To compare such yields is fraught with many difficulties to do with poorly known angular distributions, acceptances and yield-curve widths, even leaving out of account those associated with bombarded targets and particle currents. It is notoriously difficult to establish reliably small cross sections under heavy ion bombardment and extreme geometries; we need not, accordingly, necessarily attach significance to apparent discrepancies in such cases perhaps even of about an order of magnitude, although all such discounting must be made consciously and with care.

II.2D Cold Fusion

It might be supposed that the best way to approach a very heavy element is to start with a target of as high a Z_t -value as can be secured in adequate quantity and to bombard it with an ion of sufficiently high z-value to achieve the aim (using Z_t and z as defined in Section III of Part I.) This method was indeed used with success in the synthesis of elements up to Z=106 (see below.) However, as one moves towards the heaviest elements of concern in our present study, this approach takes us into regions of formed compound nuclei of so high an excitation energy that many particles, including charged ones, tend to be evaporated before the final nuclei are reached so that the chance of reaching the desired aim becomes very small, and also tends to become obscured by a large quantity of lighter heavy nuclei produced. The solution to this problem, cold fusion, was first suggested by Dubna workers (see 75Og01 and earlier work referred to in that paper).

Cold fusion exploits the fact that, if one bombards Pb or Bi nuclei (having large binding energies due to closed nuclear shells) with ions of z>16, preferably also near closed nuclear shells, at energies just above the Coulomb barrier, then the excitation energies of the compound nuclei are much lower than those of the same compound nuclei produced in the bombardment of targets of much higher Z_t by lighter ions. As a result, the probability of fission especially is very much reduced in the former case and, under sufficiently fine-tuned circumstances, neutron-only emission will dominate over other or additional light particle emission particularly if the further kinematical constraint of near-forward detection of the produced heavy nuclei is applied.

The Dubna group which invented the method also tested and proved it in experiments on several fermium and transfermium isotopes. As will be recounted below, it was used in the discovery of the elements 107, 108 and 109. The task of the TWG is to determine discovery on a Z-by-Z basis, as we do in the profiles that follow. But we wish to say that, were we rather to allocate credit on a global basis, having regard to those contributions that have led to the most significant advances in our knowledge of the transfermium elements taken as a whole, then very substantial credit would be allocated to Dubna for its concept and development of cold fusion.

II.2E Reaction Mechanisms

As we have indicated in Part I, inference based upon assumptions as to reaction mechanisms, by which we mean arguments relating to angular distributions, excitation functions and the relative probabilities of competing processes, cannot be absolute although in certain cases it can be quite highly suggestive. Suppose that: (i) the relative probability of certain sorts of cross section; (ii) the course of certain excitation functions; (iii) the nature of certain angular distributions, have all been well and systematically established within a certain range of relevant parameters of: (i) ion species including most particularly of Z_t+z ; (ii) bombarding energy; (iii)excitation. Suppose further that no significant anomalies have been found within those ranges; then we may admit that another reaction carried out within the boundary conditions so established may be assumed to obey the same systematics and that confident inference may be drawn: this is interpolation. If, however, the reaction of concern lies outside those boundary conditions, we are involved in extrapolation: the confidence attaching to inference must then be correspondingly reduced, the more so the further we move from the established boundaries. This is particularly so if that move takes us into a region of the periodic table where on account of, for example, possible shell properties, we might have to anticipate significant changes in the nuclear structure.

An illustration of importance to us here concerns cold fusion. It is well established, as we have noted in section II.2D above, that this reaction mechanism in many cases leads effectively to the emission of neutrons-only, particularly if moderated by kinematical selection (forward emission) of the heavier reaction products, so that the certain observation of a remote descendant of the formed nucleus may be taken to signal the formation of an isotope of the element with $Z=Z_1+z$. We are convinced that this is frequently the case and we find it the more convincing if the excitation function of the putative descendant of the putative element Z_{t+2} follows the systematics established for other reactions within the boundary conditions as defined above. If, however, the novel case takes us outside the established boundary conditions and, particularly, if adequately-detailed information as to excitation functions and/or angular distributions is not available, we are in the realm of extrapolation and hold that information as to the formation of element $Z=Z_1+z$ cannot be drawn with sufficient confidence, suggestive as the evidence may be. We are, therefore, not inclined to award the accolade of "beyond reasonable doubt" to any experiment that has not recorded a signal directly from the claimed element of Z_t+z although we hold that inference based upon the reaction mechanism, particularly in the case of cold fusion, can be suggestive, perhaps highly suggestive, but short of compelling. In saying this, we grant that there are indeed cases where the assumption of neutrons-only emission in unknown territory and with very small cross sections is seen to be correct, or largely correct, but only in retrospect and by comparison with cross sections determined by another method with greater certainty - see e.g. the Comment in Section 109; 02 (b. 1792).

PART III: DISCOVERY PROFILES OF THE TRANSFERMIUM ELEMENTS

Presentation of the profiles takes the following form for each element Z:

- (i) Numbered paragraphs **Z**; 01, **Z**; 02 etc. discuss those papers ¹), or group of papers, considered by the TWG to be of importance in the building up of the discovery profile to the point where essentially full confidence has been reached.
- (ii) The first section under that paper or group of papers presents a factual account ²) of the relevant content of that paper or group of papers and of the authors' conclusions.
- (iii) The second section (Comment) gives factual comments placing the paper or papers under consideration in the context of our present knowledge.
- (iv) The third section (TWG Assessment) gives our estimate of the significance of the paper or papers.

After consideration of all papers relevant to the establishment of confidence in the formation of the new element, a final section presents the TWG Conclusion as to the credit for discovery.

For each element, the presentation is completed by a summary listing of all papers relevant to the discussion, including the operative date of their receipt for publication and a very brief "telegraphic style" account of their salient features and the criteria that they involve. As operative date we feel obliged to adopt the date of receipt by the editors, which may happen to precede the year of publication. At the end of the report, a Bibliography 1) is added of all papers on the subject that have come to the attention of the TWG.

ELEMENT Z=101

101; 01 The Berkeley paper 55Gh99.

In this paper the production of element Z=101 was announced. A tiny amount (about 10⁹ atoms) of 20 d ²⁵³Es electroplated on the back of a thin gold foil was bombarded with α -particles. The reaction recoils were caught on another Au foil. This foil was then treated chemically by ion exchange techniques; ²⁵³Es and ²⁴⁶Cf were added as calibrants. Spontaneous fission (SF) with a half-life of roughly 3.5 h was observed at two positions interpreted as the Fm (Z=100) and element Z=101 positions, as decided by comparison with the calibrants.

Considering that SF for odd Z elements was earlier found to be severely hindered, "it is tempting" to make the following mass number assignment: the Z=101 isotope is formed in the reaction 253 Es(α ,n) 256 101 and decays by electron-capture "with a half-life of the order of a half hour" to 256 Fm, which then decays with the 3.5 h half-life by SF.

The proof that element 101 was observed is based on the following arguments. The short SF half-life proves that "one of the very heaviest elements" is formed. The method of formation excludes production of an element with Z>101. Finally, "the elution position immediately ahead of element 100 shows that the chemical properties are those of an element heavier than 100."

COMMENT. Additional evidence that the then unknown isotope ²⁵⁶Fm is a 3 h SF activity was obtained, as described in the paper 55Ch30 immediately following 55Gh99, by studying multiple neutron-capture in ²⁵³Es. Later, its half-life was found to be 2.63 h and it decays 92% by SF. The parent ²⁵⁶101 has a half-life of 75 min and decays 90% by electron-capture (71Ho16).

TWG ASSESSMENT. In hindsight, not much doubt exists that the indicated isotopes were indeed observed. The (much later) discovery of SF isomers with Z-values around 94, however, weakens one of the arguments on which the assignment was based.

¹) The papers will be indicated by Nuclear Data Sheet type reference keys, starting with the year of publication and the first two letters of the name of the first-named author, followed by two more characters explained in the Bibliography.

²) If, in this section, a phrase is given in quotation marks, it is a literal quotation from the paper under consideration. Such quotations from papers originally written in Russian are taken from available translations (see the Bibliography); they have been checked by members of the TWG who understand Russian.

101; 02 The Berkeley paper 58Ph40.

In this paper, essentially the same technique was used but with several orders of magnitude more target material. Again, SF was detected in both the Fm and element 101 positions. The half-life in the Fm fraction was now found to be 160±10 m. For the electron-capture decay of ²⁵⁶101, "a revised half-life of 1.5 h was measured."

Furthermore, in measuring the α -activities present, a 7340 keV group with a half-life of 3/4 h was found in the Z=101 fraction. Another group at 7080 keV with a half-life of 21 h was found both in the Fm and element 101 fractions. A renewed chemical separation of the Z=101 fraction showed this α -ray again to be present in both fractions, demonstrating that it must be due to 255 Fm (already reported before, 56Jo09) formed by decay of a Z=101 parent with a half-life of about 1/2 h (not essentially different from that of the 7340 keV α -ray) itself produced in the reaction 253 Es(α ,2n) 255 101. Rough yield-curve data are given in a table.

COMMENT. Best data now: 255 Fm half-life 20.1 h, E_{α} =7015.8 keV; 255 101 $T_{\frac{1}{2}}$ =27±2 m, E_{α} =7326±5 keV. The agreement is sufficient.

TWG ASSESSMENT. The combination of the two Berkeley papers, but especially the A=255 parent-daughter relation found in the latter, provide sufficient confidence in the existence of element Z=101.

101; 03 TWG CONCLUSION. Element 101 was discovered by the Berkeley group - with certainty in 1958 (58Ph40) following strong indications in 1955 (55Gh9).

Z=101: SUMMARY OF PAPERS, DATES AND CRITERIA

In the summaries, α -particle energies given without mentioning the unit are given in keV. The criteria used in the papers are indicated by the symbols defined in Part I above, except that Cs is replaced by σ and Ei, E α by E_i , E_{α} .

Z Ref. Date Place 101 55Gh99 550418 (Berkeley) 58Ph40 580821 (Berkeley)

55Gh99 253 Es(α ,n) 256 101 E_i Ci SF G1 T

Method. Ion exchange chemistry of reaction recoils. Found SF time dependence interpreted as $T=\approx 30 \text{ min}^{256}101 \text{ decaying to 3 h SF}^{256}\text{Fm}$ (genetic relation not demonstrated).

Proposes the name mendelevium, symbol Mv; name accepted by the IUPAC Commission on Nomenclature of Inorganic Chemistry, who changed symbol to Md.

58Ph40 253 Es(α ,2n) 255 101 Ey σ Ci G1(Tc) E_{α} T 253 Es(α ,n) 256 101 T

Method. As 55Gh99. Parent-daughter relation of \approx 30 m $^{255}101$ to 21 h E_{α} =7080 255 Fm proved by repeated chemistry (milking.)

ELEMENT Z=102

102; 01 The Argonne-Harwell-Stockholm paper 57Fi99.

This paper claims the discovery of element 102. With the Stockholm cyclotron, one of the first accelerators to produce heavy ions, a 1 mg/cm² Cm target was repeatedly bombarded with about 1 μ A of ¹³C⁴⁺ ions with energy ranging between 65 and 100 MeV. After each 30 min bombardment, the target was treated chemically using ion exchange techniques. In about 12 out of 50 cases, 8.5 ±0.1 MeV α -particles were observed in samples prepared from drops emerging somewhat earlier than Fm calibrants (Z=100), and much earlier than those of Cf (Z= 98). They appeared in experiments with 3 out of 6 targets, during the first two weeks after their preparation. The half-life was approximately 10 m. The activity was assigned to the isotope ^{2510r253}102 though the possibility was not excluded that the α -particles were due to its electron-capture Z=101 daughter, presumed to be rather shorter lived.

COMMENT. The Berkeley paper 58Gh40 describes a repetition of the experiments, using monoenergetic ¹²C and ¹³C ions from their linear accelerator and with virtually the same target material as used in Stockholm. Though they produced orders of magnitude more ²⁴⁶Cf demonstrating that their intensity was much higher, they found only very few counts in the 8.5 MeV region which, moreover, must be assigned to background.

In the subsequent Argonne-Harwell-Stockholm paper 59Fi99, an attempt was made to explain the difference, maintaining that element 102 was indeed seen. However, in all later work, no trace was found of any z=102 isotope with a half-life longer than 3 m, for A<259 (the maximum that could have

been formed). But an ancestor of 4.2 μ s ²¹³Po, with an α -energy of 8375 keV, and a half-life of roughly 10 m, could have been formed in the Stockholm bombardments. It has been reported to the TWG that, in somewhat similar bombardments, 8 m ²²⁵Th is formed prolifically, and also that it would not have been removed by the chemical treatment.

The first Berkeley chemical investigations on element 102 (68Ma99) showed that, unexpectedly, the divalent state is more stable than the trivalent one. This undermines the chemical argument in the Argonne-Stockholm paper for assignment to element 102.

TWG ASSESSMENT. The paper 59Fi99 defending the Stockholm work fails to convince, certainly in the light of later work. It must be considered highly unlikely that an isotope of element 102 was seen in 57Fi99.

102; 02 The Berkeley paper 58Gh41.

The discovery of element 102 was then claimed in this paper. The authors tried to find Z=100 daughters from Z=102 α -decays by a double recoil method. A thin target of Cm (95% ²⁴⁴Cm, 5% ²⁴⁶Cm) was bombarded with ¹²C ions of which the energies were varied from 60 to 100 MeV. The reaction recoils were caught and decelerated in He gas and then deposited, with the help of an electric field, on a moving belt passing under a catcher foil for the expected secondary α -recoils. A ²⁵⁰Fm activity was indeed found on the catcher, as found from the observed α -particle energy of 7430 keV, and in an experiment in which the whole catcher was analyzed with ion exchange techniques. In other experiments, in which the catcher was divided into 5 parts corresponding to different time delays, the half-life of the parent was found to be about 3 s. "The excitation function for its production ... was found to peak sharply at 70±5 MeV ... in accordance with" .. its assignment to ²⁵⁴102 formed by the reaction (12 C,4n) on the lower abundant Cm isotope present in the target, and an available theory for excitation curves. The cross section was said to be a few microbarns.

COMMENT. In Dubna work, see 102; 06, ²⁵⁴102 was reported to be found in essentially the same way. They found, however, that the intensity of the ²⁵⁰Fm recoils corresponded with a parent half-life of 50±10 s. This result has been confirmed fully (see 102; 07).

The Berkeley group, see 67Gh01, tried to explain this discrepancy by assuming that the activity found in the chemical experiments was in fact 250 Fm, but that seen in the half-life determinations was, instead, 244 Cf (T=20m, E_{α} =7220 keV), granddaughter of 252 102 formed by the same reaction as mentioned above but on the more abundant 244 Cm isotope. The latter Z=102 isotope was indeed found later to have a half-life of 2.3 s (77Be09). The difference in energy from the 250 Fm 7430 keV α -particles was explained as due to "resolution and drift problems".

It should be noted that much later Berkeley experiments, reported in 73Gh03, showed quite unexpectedly, that 250 Fm recoils can be caused by the isomeric decay of a 1.8 s isomer in this nuclide. This isomer could have been formed abundantly by the reaction 244 Cm(12 C, α 2n) at about the same 12 C energy.

TWG ASSESSMENT. The interpretation of the observed α -particles as being due to a Z=102 ancestor must be considered as probably wrong: assignment to 250m Fm recoils is more probable. No resolution and drift problems had been mentioned before, and they should also have influenced the measured energy values of, among others, 246 Cf and 250 Fm.

102; 03 The Berkeley paper 59Gh99.

In this paper a report is given of a continuation of the above experiments. The Berkeley group attempted to find α -particles due to the supposed 3 s Z=102 isotope. For this purpose, the reaction recoils were deposited on a thin foil drawn into a grid ion chamber after a short irradiation. Indeed, 8.3 MeV α -particles were found with a half-life of about 3 s. Also, spontaneous fission events were found and interpreted as being due to a 30% SF branch. It was also stated that "due to the difficulty and the complexity of the conditions ... we cannot be sure that this activity was actually caused by element 102"

COMMENT. Later, see e.g. 67Gh01, ²⁵²102 (but not ²⁵⁴102) proved to have the properties reported in this work.

TWG ASSESSMENT. It seems quite likely that an isotope of element 102 was seen here for the first time, but its assignment was not even claimed to be convincing.

102; 04 The Moscow papers 58Fl42, 58Fl99 and 60Fl01.

In these papers, photographic emulsions for the detection of α -particles were used. The first paper states that thin layers of the isotopes ²³⁹Pu and ²⁴¹Pu were bombarded with ¹⁶O⁵⁺ ions of energies up to 100 MeV. The recoils were caught on a collector periodically moved to the thick photographic emulsion. In this first paper, 18 and 8 tracks respectively were found for the two target isotopes mentioned in the region above 8.5 MeV. The effect was assigned to one of the isotopes ^{251,252,253}102. The paper states that production of the 8.5 MeV α -emitter from Pb or Bi impurities in the target cannot be excluded. Formation of Z=101 isotopes with this α -energy in a (¹⁶O,pxn) reaction is excluded only because the α -particle energies for them are expected to be rather lower.

Somewhat later work with the same technique reported in a review paper 58Fl99 shows that bombardments of Hg, Tl, Pb and Bi with 16 O did indeed yield "unknown isotopes" which emit α -particles of energies 8-9 MeV and 11-12 MeV. Yet, "repeated control experiments ... confirm the (earlier) deduction that by irradiation of 241 Pu with 16 O atoms, one can observe the formation of the isotope of element 102". The half-life is given as shorter than about 30 s, the α -particle energy as 8.8 ± 0.5 MeV. The background due to isotopes formed from Pb impurities, as estimated from the ratio to 7.4 MeV α -particles, is estimated to be only 24%. It is mentioned that the reported α -particle energy may be somewhat low.

Another series of these experiments is described in 60Fl01. The influence of the possibly unequal distribution of the Pb impurities in the target is investigated by comparing the yield of the 9 MeV α -rays to that of those of 7430 keV known to be due to ²¹¹Po in equilibrium with its parent 16 h ²¹¹Rn.

The intensity ratio was found to be different in bombardments of the target with 12 C and 16 O, which was taken as an indication that Pb was a surface contaminant. Indeed, for Pb targets of different thicknesses, the yield ratios were also found to be different in a way that could be explained as due to the different ranges of 12 C and 16 O recoil products. The energy of the supposed Z=102 isotope is now given as 8900 ± 400 keV and the half-life as between 2 and 40 s. A rough yield-curve experiment was taken as an indication that the activity was due to the reaction 241 Pu(16 O,4n) 253 102, though the production of 254 102 was not excluded. Again, (16 O,pxn) and (16 O,oxn) reactions are excluded only because the resulting α -particle energies are expected to be significantly smaller.

Repetition of the bombardments of ²³⁹Pu led to the conclusion that the 58Fl42 result for it mentioned above "was entirely due to a background effect."

COMMENT. It is now known that 95±10 s 253 102 has E_{α} =8010±30 keV (67Mi03), whereas 53±3 s (79Ni01) 254 102 has E_{α} =8086±20 keV (85He22). In a later review paper, 67Fl99, the earlier Moscow energy value was lowered to 8600±400 keV based on lower energies 11650 (instead of 12000) keV for 4.5 s 212m Po, and 7270 keV for 25 s 211m Po (also emitting the 13-times weaker 8885 keV peak) instead of 7430 keV for 0.5 s 211 Po (which, notwithstanding its short half-life, could be present e.g. as EC daughter of 211 At). The resulting energy value suggests that it might still agree with the mentioned Z=102 α -energies. But, using in addition for calibration the 211m Po 8885 keV line in the spectrum from the Pb bombardments as shown in Fig. 4 of 60Fl01, one must conclude that the ranges for these α -particles should be 40 and 41 μ m respectively, well below the "ranges in the interval 45-51 μ m of emulsion" shown in their Fig. 5 and discussed in the paper.

TWG ASSESSMENT. We agree with the statement in the papers 70Fl98 and 87Fl99 by the Dubna group itself, that these results cannot be considered reliable.

102; 05 The Berkeley paper 61Gh03.

In this paper the discovery of element Z=103 is claimed (see 103; 01). In addition, an α -line with an energy of 8.2 MeV with a half-life of 15 s was assigned to element Z=102 based on the fact that its intensity in a bombardment of Cf with ¹²C (assigned to a ($^{12}C,\alpha xn$) reaction) was higher than in ones with ^{10,11}B. Without giving reasons for the mass number assignments, it was "thought to be mostly $^{255}102$."

COMMENT. Half-life and energy do not agree with later values for $^{257}102$. They do agree with those for $^{257}102$ ($T=23\pm2$ s, $E_{\alpha}=8250\pm20$ keV for the average of two slightly different branches, see 67Gh01), and the agreement is even better for the value 8240 keV obtained in a recalibration of the α -particle energies in this work (see 103; 02)

TWG ASSESSMENT. Although it seems highly likely that the isotope ²⁵⁷102 was seen in this experiment, this paper cannot be accepted as (and was not intended to be) a convincing demonstration of the existence of element 102.

102; 06 The Dubna papers 64Do10 (=63Do99) and 66Do04.

These papers describe experiments to detect α -decay daughters of Z=102 isotopes, by a method closely similar to that used in the Berkeley paper 58Gh41. The internal Ne beams of the Dubna cyclotron were made to impinge on a thin ²³⁸U target. The recoils, slowed down in a gas, were deposited through diffusion onto an annular recess in a rotating disk and carried along a silver catcher foil. The deposit on this foil was purified chemically, by ion exchange techniques, and the Fm fraction counted in a grid ion chamber. The α -energies and the half-life values agree with the values known for 23 h ²⁵²Fm and 30 min ²⁵⁰Fm.

In the first series of experiments, the efficiency of the collection method was checked by comparing the 250 Fm activity produced directly in the reaction 238 U(20 Ne, α 4n) 250 Fm with the amount of its daughter 246 Cf collected on the Ag catcher. The yield curves determined in both ways agreed quite well and showed that the overall collection efficiency was about 15%, in agreement with the calculated value. In contrast, the collection efficiency in a case of electron-capture, produced in the reaction 140 Ce(16 O,7n) 149 Dy(ϵ) 149 Tb, was only 1%.

The double recoil yield of 252 Fm, interpreted as indicating the existence of its α -decay parent $^{256}102$ formed in the reaction 238 U(22 Ne,4n), showed a shape as expected for this reaction with a maximum cross section of 45 nb at 112 MeV.

It is mentioned that a large amount of ²⁵²Fm is formed directly in the reaction ²³⁸U(²²Ne,α4n)²⁵²Fm which has a maximum cross section of 250 nb. Therefore, special attention was paid to preventing this reaction product from reaching the catcher foil directly.

In order to determine the half-life of the parent, the experiment was done at different rotating speeds. For each of them, the catcher foil was cut into two pieces and the ratio of the relevant activities was determined. "The distribution of the activity of ²⁴⁶Cf as daughter of ²⁵⁰Fm (*T*=30 m) along the collector was found to be independent of the speed at which the disk was rotated (in the interval to 1 rpm)" which emphasizes the importance of the fact that the intensity of ²⁵⁰Fm itself decreased with decreasing rotation speed. An analysis yielded the result that "The half-life was established as close to 8 s."

In 66Do04 the same method was applied to ²⁵⁰Fm as supposed daughter of the isotope formed in the reactions ²⁴³Am(¹⁵N,4n)²⁵⁴102 and ²³⁸U(²²Ne,6n)²⁵⁴102. The excitation curves showed a neutron evaporation character with (both) maxima of about 50 nb, at 82 MeV and 125 MeV respectively. A half-life determination made with the first reaction showed that it was certainly much longer than the 3 s mentioned in **102**; 02. The other reaction yielded the value 50±10 s.

COMMENT. At the time of publication, no explanation could be given for the difference between the new result on the half-life of ²⁵⁴102 and the earlier Berkeley one: the Dubna paper states that "Additional studies are clearly required to explain the enormous discrepancy between our value for the half-life of ²⁵⁴102 and that given by" .. the Berkeley group, see **102**; 02.

After 1971 (71Gh03), the following explanation could be given. As stated in 102; 02, the recoils in the Berkeley work were probably due to the 2 s 250m Fm isomer. The intensity of this isomer in the Dubna work was weaker owing to the following circumstances. The energy of the impinging 12 C ions in Berkeley was selected to be optimum for a (12 C,4n) reaction on the less abundant 246 Cf isotope in the target, but is then also advantageous for the (12 C, α 2n) reaction on the 20 times more abundant 244 Cm. Also, the ratio of (HI,xn) to (HI, α xn) reactions is expected to be higher for HI= 12 C than for HI= 22 Ne. The relative amount of 250m Fm was therefore smaller in the Dubna experiments.

TWG ASSESSMENT. The Dubna results discussed here were obtained in essentially the same way as the Berkeley one. The lack of understanding of the difference between the Dubna and Berkeley results for ²⁵⁴102, at the time, prevented attaching more convinction to these Dubna results alone than to the earlier Berkeley one. In retrospect, the Dubna results must be considered to be quite probably correct, even though the reported half-life value for ²⁵⁶102 is somewhat high (a value 3.2±0.2 s is reported in 67Gh01.)

102; 07 The Dubna paper 66Za04.

This paper describes an attempt to measure the α -particles of $^{254}102$ produced in the same reaction as used by 66Do04 (which two papers were received by the publisher on the same date.) Reaction recoils from thin targets were caught in a He gas stream that was swept thereafter continuously through a 0.5 mm orifice. The gas jet then deposited the formed activity onto a wheel that could be turned stepwise to semi-conductor α -particle detectors. By shielding these detectors after some time, the α -activities from the Z=100 daughter deposited on them by α -recoil could also be studied. An α -ray of 8100±50 keV was observed to decay with a half-life between 30 and 40 s, with a production cross section (100 nb) comparable with that (200 nb) of the production of 248 Fm in the parallel reaction 237 Np(15 N,4n) 248 Fm.

Since the half-life and the α -energy are similar to those of 213 Rn formed as granddaughter of 34.6 s 213 Fr, they studied the α -decay of the recoils on the detector as a result of the α -decay of the formed activity. They indeed detected 250 Fm (identified by half-life and α -energy) in the right intensity ratio with the 30 s activity. Study of its intensity as a function of time indicated that its parent indeed had a half-life much longer than the 3 s reported earlier by Berkeley. Increasing the bombarding energy from 80 to 90 MeV strongly reduced the intensity of both the 30 s activity and of the 250 Fm recoils, showing that "only part of the observed effect could be due to impurities in the target."

COMMENT. This Dubna result was fully confirmed later, among others by the Berkeley report 67Gh01.

TWG ASSESSMENT. This work is considered fully convincing.

102; 08 The Dubna papers 64Dr99 and 66Ku15.

In these papers, the spontaneous fission activity following possible production of $^{256}102$ was studied. The first one mentions a neutron-emitting 4.15 s ^{17}N background causing n-induced fission. They saw in addition a 10 s activity formed with a cross section of 300 pb; by comparison with the 45 nb cross section reported by 63Do99 (=64Do10) for production of the 8 s α -emitting activity formed in the same reaction $^{238}U + ^{22}Ne$, they derive a value 1/150 for the fraction decaying by SF. The yield curve agrees with a 4n emision process. The activity is not seen in ^{16}O and ^{20}Ne bombardments on ^{238}U .

A similar investigation reported in 66Ku15 found a cross section of 200-300 pb in the same reaction, and appreciably more in $^{242}Pu + ^{18}O$: a maximum of 700 nb for a yield curve with a shape corresponding with a 4n evaporation. The paper is devoted especially to measurement of the half-life which was found to be 8.2 ± 1.0 s.

COMMENT. The only other mention of a SF branching in ²⁵⁶102 we could find is 67Gh01 who sees "what could be a SF branching by the 3-sec ²⁵⁶102 at a level approximately a factor 2 below that found in" ... the above papers.

The difference between the $^{256}102$ half-life value used in 64Dr99 and measured in 66Ku15 and the value 3.2 ± 0.2 s first obtained in 67Gh01 and confirmed by the value 3.7 ± 0.5 s of Dubna's 68Fl05 is somewhat difficult to explain, even taking into account the known difficulties in half-life determinations.

TWG ASSESSMENT. These experiments cannot be accepted as proof of the existence of element 102.

102; 09 The Oak Ridge paper 71Di03.

These experiments on the detection of X-rays following the α -decay of ²⁵⁵102 clearly showed the presence of Z=100 K X-rays, in about 150 events.

COMMENT. This case is mentioned here only as example of the use of this incontrovertible criterion.

TWG ASSESSMENT. This observation is a very convincing confirmation of the assignment to the element z=102.

102; 10 TWG CONCLUSION. The two 1966 (simultaneously published) Dubna results 66Do04 and, especially, 66Za04, both submitted in 1965, give conclusive evidence that element 102 had been produced.

Z=102: SUMMARY OF PAPERS, DATES AND CRITERIA

Z	Ref.	Date	Place	Remark
102	57Fi99	570719	(Stockholm-Harwell-Argonne)	
	58F142	580228	(Dubna)	see 60Fl01
	58Gh40	580606	(Berkeley)	
	58Gh41	580606	(Berkeley)	
	58F199	580901	(Dubna)	see 60Fi01
	59Fi99	590128	(Stockholm-Argonne)	
	59Gh99	590521	(Berkeley)	similar to 58Gh41
	60F101	590801	(Dubna)	
	61Gh03	610413	(Berkeley)	
	63Do99	630803	(Dubna)	=64Do10 *)
	64Do10	631118	(Dubna)	
	64Dr99	640228	(Dubna)	
	65Do98	650413	(Dubna)	=65Do09
	65Do09	650419	(Dubna)	
	66Do.A	651126	(Dubna)	=66Do04
	66Do04	651215	(Dubna)	
	66Za04	651215	(Dubna)	
	66Ku15	660122	(Dubna)	
	67Gh01	670209	(Berkeley)	
	71Di03	710310	(Oak Ridge)	

*) Papers so indicated are given only to show that the information provided in the second paper was available earlier.

57Fi99 246 Cm(13 C,5n) 254 102 $T E_i E_{\alpha}$ Ci G1

Method. Target 95% 244, 1% 245, 4% 246. Reaction recoils caught on foil. Ion exchange chemistry is reported to show the known isotopes 250 Fm and 246 Cf, considered to be descendants of the new Z=102 nuclide, itself eluting at the place expected from extrapolation. Proposes the name Nobelium (see 67Gh99 below).

58Gh41 246Cm(12C,4n)254102 Ey T G1(Tc) 240Pu(12C,4n)248Fm Ei G1(Tc)

Method. Target 95%244, 4.5%246. Reaction recoils deposited on moving belt carried along catcher foil for α -recoils. On catcher ²⁵⁰Fm is found. From cutting the catcher in strips and counting α -rays, T is found. Check: the experiment on a Pu target yielded α -recoils to ²⁴⁴Cf.

59Gh99 246 Cm(12 C,4n) 254 102, $T E_{\alpha}$ Br 30%SF

Method. Reaction recoils deposited on thin foil, drawn into grid ion chamber. Target rigorously purified to suppress backgrounds due to Pb impurities. Data presented as preliminary.

67Gh99 Review. Proposes to retain the name Nobelium (symbol No) though the supposed discovery by 57Fi99 is not accepted. This name was eventually recommended by the IUPAC Commission on Nomenclature of Inorganic Chemistry.

58F142 239 Pu(16 O,4n) 251 102 E_i E_{α} 241 Pu(16 O,4-6n) $^{251-3}$ 102 E_i E_{α}

Method. Reaction recoil collector transferred to photographic plate. Yield reported to increase up to E_i =100 MeV.

58F199 same, $T << 4m E_{\alpha}$

60F101 241 Pu(16 O,4n) 253 102 T=2-40 s E_{α}

Method. As 58F142. With 239 Pu target little found. Investigates influence of possible impurities with separate bombardments on Hg, Tl, Pb, Bi. Thus discovers 211m Po E_{α} =9.0 MeV 212m Po E_{α} =12 MeV (now 25 s 8885, 45 s 11635 keV)

67F199 Reanalysis of the above.

Revises E_{α} based on better values for the ^{212m}Po and ^{211m}Po lines, and then identifies with ²⁵²102.

61Gh03 250 Cf(12 C, α 3n) 255 102 E_i Cb TE_{α} 250 Cf(11 B,p5n) 255 102

Method. See 61Gh03(Z=103). Target 3% 249, 33% 250, 12% 251, 51% 252. This result is a byproduct of an experiment designed to discover element Z=103.

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64Do10 <sup>238</sup>U(<sup>22</sup>Ne,4n)<sup>256</sup>102 Ey T G1(Tc)
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Method. Internal beam. Reaction recoils slowed down in gas, deposited through diffusion on an annular recess in a rotating disc and thus carried along a catcher foil for α recoils. The catcher is treated chemically, the collected ²⁵²Fm activity analyzed as function of the position on the catcher to determine the half-life of the parent. Background through direct production in ²³⁸U(²²Ne, α 4n)²⁵²Fm avoided through leak proofing. Reports T=8 s; comment of 68Fl05: "The experimental accuracy ... was at best \approx 40%."

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65Do09 <sup>238</sup>U(<sup>22</sup>Ne,4-6n)<sup>256-4</sup>102 Ey σ G1
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Same method, ^{252–250}Fm daughters observed. No mention of half-lives, only yield curves given.

66Do04 243 Am(15 N,4n) 254 102 Ey σ Cb T G1(Tc)

²³⁸U(²²Ne,6n)²⁵⁴102 same

Method: Daughter ²⁵⁰Fm chemically isolated by same procedure as in 64Do10.

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64Dr99 ^{238}U(^{22}Ne,4n)^{256}102 SF \sigma T \approx 10 s
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Method. Target after irradiation moved to electronic detectors.

66Ku15 238 U(22 Ne,4n) 256 102 SF σ $T \approx 10$ s

 242 Pu(18 O,4n) 256 102 SF $\sigma T 8.2\pm1.0$ s

²⁴²Pu(¹⁸O,p3n)²⁵⁶101 T long SF

Method. Reaction recoils caught on belt moving along glass fission-track detector, slightly modified version of that in 64FI04, Z=104.

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66Za04 ^{243}Am(^{15}N,4n)^{254}102 Ey Cb TE_{\alpha} G1(Tc,Ic) ^{237}Np(^{15}N,4n)^{248}Fm Ey
```

Method. Focussed external beam to target. Reaction recoils caught in He, gas jet through 0.5 mm orifice on metal catchers on end of arms of cross, flipped to Si detector. Detection of daughter activities put by α -recoil on detectors after placing absorber in front of them. Parent of 250 Fm; from collection, T(parent) >> 3s. Much less activity with 14 N bombardment.

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67Gh01 ^{244,246,248}Cm(^{12-13}C,3-5n)^{251-257}102 Ey Cb TE_{\alpha}
```

Method. Reaction recoils stopped in gas, stream through 0.2 mm orifice deposits on rim of wheel, rotated digitally in 50 degree steps to α detectors.

68Si99 244,246,248 Cm(12,13 C,3-5n) $^{251-257}$ 102 Ey Cb $^{T}E_{\alpha}$

More extensive report on the results reported in 67Gh01. Gives yield curves. Mentions 214 Ra(2.6 s) 214 Fr(4 ms E_{α} =8430) background.

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71Di03 ^{249}Cf(^{12}C,\alpha2n)^{255}102 TE_{\alpha}(C)E_{x}
```

Method. He jet (see 67Gh01) deposition on foil pneumatically transported to α -X-ray coincidence counting assembly. Very clear Z=100 K X-ray spectrum detected in coincidence with α -spectrum assigned already earlier to ²⁵⁵102. Most convincing assignment to Z=102.

ELEMENT Z=103

103; 01 The Berkeley paper 58Gh41.

Already in this paper on element 102, reference is made to an attempt to produce an isotope of element 103 in the reaction between the same Cm target used there and ^{14}N ions, this time looking for long range α -particles with nuclear emulsions. They found 18 tracks with an energy of 9±1 MeV and a half-life of approximately 1/4 s. "These tracks could be due to an isotope of element 103 but from these crude preliminary experiments it is of course not possible to rule out" .. other possibilities.

COMMENT. The reported data can be said to agree reasonably with the 8870 keV α -particle energy and 0.6 s half-life of $^{257}103$ (71Es01) formed in the reaction ($^{14}N,3n$) on the 5% ^{246}Cm in the target.

TWG ASSESSMENT. The indications obtained are far too weak to merit credit for the discovery of element 103.

103; 02 The Berkeley paper 61Gh03.

The first substantive paper concerning element 103 is this one. A thin Cf target (3% 249, 33% 250, 12% 251, 51% 252) was bombarded with ¹⁰B, ¹¹B and ¹²C ions. The reaction recoils were collected in a He gas stream which was then swept through a 0.05 inch orifice. The He jet, impinging on a Cu

carrier tape, deposited there the activities formed. The tape carried them to solid state detectors to measure the resulting α -particle energies.

In bombardments with both ^{10}B and ^{11}B , an activity with a half-life of 8 ± 2 s and an α -energy of 8.6 MeV was found, in addition to previously unknown peaks at 8.4 and 8.2 MeV both with an apparent half-life of about 15 s. The yield curves for the 8 s activity "were, of necessity, very broad because the same activity could be produced by" .. reactions on several isotopes in the target. For this reason the Z=103 isotope is thought to be $^{257}103$, formed in the reactions $^{252-x}Cf(^{11}B,(6-x)n)^{257}103$ and $^{252-x}Cf(^{10}B,(5-x)n)^{257}103$.

Indication that this activity may have been due to element 103, and not to Z=102 or 101 formed in (B,pxn) or (B,oxn) reactions, was obtained in the following way. The ¹²C bombardments are expected to yield Z=102 through (C,oxn) reactions at considerably higher intensity, compared with Z=103 or 101 through (C,pxn) and (C,opxn) reactions. Indeed, due to the change from B to C bombardments "It was found that the 8.6 MeV (8 s) activity was decreased by more than a factor 2, and the (15 s) 8.2 MeV activity was increased by a factor of about 20" (about the latter see 102; 05). Furthermore, "Possible light isotopes of (element 101) that could be produced and conceivably might emit α -particles in the 8.2-8.6 MeV region were ruled out by bombardments of ²⁴³Am with ¹²C ions."

COMMENTS. Later work, firstly from Dubna (68Fl01), showed that $^{257}103$ did *not* have the assigned properties. The mass assignment was changed by Berkeley (67Se99, 68Le99) to $^{258or259}103$. Indeed the properties for $^{258}103$ (a 4.2 ± 0.6 s emitter of 8620 ± 20 keV α -rays) were found to agree reasonably with those reported above.

The change was criticized on several counts by Dubna workers.

- a. The possibility that the formed activity is a higher isomer of an isotope of Z=101 was not excluded (68Do19, 68Do99).
- b. If isotopes of element 103 had been present, α -rays of the isotope $^{256}103$ ($T=26\pm2$ s, E_{α} : 20% 8520, 13% 8480, 34% 8430 ±20 keV and some weaker branches, see 71Es01) should have been seen too (70Dr08).
- c. The ratio of 8610 to 8450 keV α -particles assigned to $^{258}103$ and $^{259}103$ respectively in production with 78-88 Mev ^{15}N particles on ^{248}Cm , see Fig. 8 of 71Es01, is about 1.6. But in 61Gh03 (see Fig. 2 there) the ratio of the 8.6 to 8.4 MeV peaks is at least about 5, after production in $^{250-252}Cf+^{11}B$, though the excitation energy of the compound nucleus formed must be about the same. Moreover, the 8.2 MeV line there is not seen at all in 71Es01, confirming that the results given were not reliable (recent addition to 89F199 of Donetz and Shchegolev.)
- d. The yield in (¹⁰B,3n) is expected to be much smaller than that in (¹⁰B, 4n or 5n) reactions, moreover ²⁵²Cf is the most abundant isotope in the target. Therefore, in the ¹⁰B bombardments, ²⁵⁸103 must have been produced almost exclusively from the reaction ²⁵²Cf(¹⁰B,4n) and then the yield curve would not have been "very broad" (68Do19, 68Do99).

Criticism a, seems to have overlooked the reported ²⁴³Am+1²C experiment reported in 61Gh03.

An answer to criticism b. is obtained by recalibrating the observed α -spectrum (Fig. 2 in 61Gh03) with help of modern α - energy values for some impurities present. The α -energy for the 8 s peak becomes 8590 keV with an error of say 30 keV, in quite good agreement with the best value 8600±15 keV for the average of the two strongest peaks in 6.6±0.6 s 258 103 (76Be.A). The fact that the reported half-life is somewhat longer is not difficult to understand in view of the low statistics. The 8.2 MeV activity, which is now associated with 257 102, is discussed in 102; 05. The reported 8.4 MeV peak in reality shows three sub-peaks corresponding to energy values 8530, 8450 and 8370 keV. These values agree almost surprisingly well with the energy values mentioned for the three strongest lines in 256 103. Also, this peak is not reported to be stronger in the 12 C bombardments. Thus, no conclusion can be based on the non-occurrence of 256 102.

As to critizism c. it must be pointed out that the excitation curve Fig. 6 in 71Es01 shows that the ²⁵⁸103/²⁵⁹103 ratio is a very sensitive function of the excitation energy of the compound nucleus formed. The bombardment energy in 61Gh01 (not given) must have been chosen to get an optimum yield for the 8.6 MeV peak in the spectrum Fig. 2, but then, according to the yield curve mentioned, the yield of the 8.4 MeV peak was rather less. As to the non-occurrence of the 8.2 MeV ²⁵⁷102 peak in 71Es01, it is a fact that all spectra produced after ²⁴⁸Cm+¹⁵N bombardments show element 102 peaks, but none after ²⁴⁹Cf+¹¹B reactions. The production of element 102 evidently does not occur via compound nucleus

formation and can therefore depend sensitively on the bombarding particles.

It must be conceded that criticism d. has not been answered in a fully satisfactory fashion. Private information from Oak Ridge indeed indicates that the cross sections for ²⁴⁹Cf(^{10,11}B,4n)^{258,257}103 are the same, but those for 3n evaporation (but also for 5n) are about a factor 5 smaller.

TWG ASSESSMENT. At the time of the appearance of the Berkeley paper, it was felt to be a quite convincing demonstration that an isotope of the element 103 had been observed. The mass assignment cannot be considered fully convincing and, indeed, was later proved to be wrong, but the arguments for assigning this activity to element 103 (relative intensities in bombardments of Cf+B, Cf+C and Am+C) do not depend on the mass number assignment. Of the later arguments against accepting the evidence presented, only the one based on the width of the cross section curve must still be considered valid. It is possible that the difference between the reported "very broad" yield curve and the sharper one that it is now clear should have been found is due to the poor statistics involved, but in view of the regrettable lack of proper reporting of numerical yield curve data at the time of publication and the lack of later confirmatory data on this subject, this must remain a point of uncertainty.

The reported half-life is a little on the high side, but in view of the low statistics the TWG does not consider the difference significant.

103; 03 The Dubna paper 65Do10.

This first Dubna paper on element 103 followed the method already described above, see 102; 06. The recoils deposited on the rotating wheel were obtained from the reaction 243 Am+ 18 O which, by emission of 5 or 4 neutrons, produces the isotopes 256or257 103. The decay of 256 103 proceeds by electron-capture to "8 s" 256 102 and/or by α -decay to "8 m" 252 101, which in their turn may both decay to 252 Fm. Indeed, atoms of 252 Fm were found on the collector foil as proved by chemistry and from half-life (25 h) and α -particle energy (7040 keV). The half-life of the ancestor "proved to be close to 45 s", which showed that it was neither 252 102 nor 252 101. Also, the excitation curve is narrow, with a maximum of 60 nb at 96 MeV, as expected for the reaction 243 Am(18 O,5n) 256 103, whereas that for the reaction 243 Am(18 O, α 5n) 252 101 should be broader and have its maximum at higher energy. Moreover, the efficiency for recoil in electron-capture decay, necessary to explain the result as consequence of the last reaction, is found to be small.

The possibility that Fm atoms directly formed in the reaction might be deposited was said to be "eliminated since the shielding system reliably (separated) the target region from the region of daughter product collection." Also, "the quantity of ²⁵²Fm on the collector was 20% of the entire amount of ²⁵²Fm nuclei produced as result of irradiation involving the synthesis of ²⁵⁶103". And a separate experiment showed "that less than 0.1% of all ²⁵⁰Fm atoms produced in the reaction ²³⁸U(¹⁸O,6n)²⁵⁰Fm reached the collector."

COMMENT. The reported half-life value of 45 ± 10 s is somewhat higher than the later values 31 ± 3 s (71Es01) and 25.9 ± 1.7 s (77Be36).

TWG ASSESSMENT. The difference with the later half-life value can be explained by a rather limited amount of background and is not considered to be serious. It seems probable that an effect due to the isotope ²⁵⁶103 was in evidence. Nevertheless this experiment alone cannot be considered as a proof that element 103 was in evidence (see 102; 06.)

103; 04 The Dubna paper 68Fl01.

In this work the gas jet technique (see 102; 07) was applied to the reaction 243 Am+ 18 O. The α -spectrum showed several peaks in the region 8350-8600 keV decaying with a half-life of about 35 s. Yield curve measurements showed a difference between the parts 8350-8500 keV and 8500-8600 keV which, on this basis, were assigned to the two isotopes 256 103 and 257 103 respectively. The maximum cross section for the lower energy part is estimated to be 30 nb at 95 MeV. No attempt was made to find the α -decaying 252 Fm granddaughter activity. Notwithstanding repeated attempts, the result of 61Gh03 assigned to 257 103 could not be reproduced.

COMMENT. In later Berkeley work (71Es01), both parts were assigned to ²⁵⁶103 alone. The cross

section reported in this Dubna work is only half as large as that reported in the preceeding paragraph, but since it applies only to the lower half of the α -spectrum of the mass 256 isotope, it can be concluded that sufficient agreement exists.

The isotope ²⁵⁷103 was reported in 71Es01 to have none of the properties assigned by the earlier workers but to have a half-life of 0.7 s, as also confirmed in later work (76Be.A).

TWG ASSESSMENT. This result, especially if combined with that discussed in section 103; 03, provides good but not complete confidence that the isotope ²⁵⁶103 was produced.

103; 05 The Dubna chemical paper 69Ch99.

This paper reports chemical work on the ²⁵⁶103 isotope prepared as described in paragraphs 103; 03 and 103; 04. The technique used is gas chromatography as described in 104; 02. "The reported experiments ... do not provide full chemical characterization of the new elements." They, (that is 102 and 103) "are not to any extent similar to the elements of Group IV or beyond and ... must be related to Group III of the Periodic system."

TWG ASSESSMENT. The results, though interesting, do not show a conclusive difference from the preceding actinides.

103; 06 The Berkeley paper 70Si98.

In this paper, a description is given of solvent extraction chemistry using ²⁵⁶103 prepared by bombardment of ²⁴⁹Cf with ¹¹B. The percentage extraction was given as function of the pH. The results overlap curves measured for Am, Cm, Cf and Fm.

TWG ASSESSMENT. The result shows no difference from other actinides and is not claimed to prove that element 103 was indeed present.

103; 07 The Dubna paper 70Dr08.

This paper briefly mentions experimental work in Dubna that yielded a 20 s α -emitter with E_{α} =8380 keV assigned to ²⁵⁵103 on the basis of an excitation curve, for which no further data are given. The assignment to Z=103 is reinforced by the result of yield comparisons through different collimators (see 105; 02).

COMMENT. The assignment was fully confirmed later.

TWG ASSESSMENT. The assignment is strongly suggestive but not completely compelling, being based on extrapolations of reaction properties.

103; 08 The Berkeley paper 71Es01.

This paper (preceded by the one 70Gh02 giving already part of the results) describes gas jet technique work as used before (67Gh01; see also 104; 04) and similar to that used in some Dubna work (see 102; 07). Recoils from thin targets bombarded with ions of several isotopes of B, C, N and O were stopped in a gas stream eventually directed through a narrow orifice to a wheel, there depositing the formed activities. The wheel was rotated in 39° digital steps to a series of 4-detector stations. Two of the detectors at each station alternately faced the activity and one of the two other detectors, the latter in order to detect the α -activities of the daughters deposited on them by recoil from the preceding α -emission.

In this way, results were found for the six Z=103 isotopes with mass numbers A=255-260.

COMMENT. All results reported in this work have been confirmed later. They provide adequate confidence that element 103 had indeed been observed. The results agree with findings in the previous Berkeley work on ²⁵⁸103 (but see **103**; 01, COMMENT) and in the Dubna work on ²⁵⁶103 (but not on ²⁵⁷103, see **103**; 02).

103: 09. The Oak Ridge papers 76Be.A and 77Be36.

Both experiments describe attempts to detect L X-rays following α -decay, of $^{258}103$ and $^{260}105$ respectively. For the latter paper see 105; 06; the results confirm the assignments to both elements 103 (for the daughter 25.9 ± 1.7 s $^{256}103$) and 105. Of the first experiment we found details only in an Annual Report. The 600 or so L X-rays events shown there again provide proof that the activity mentioned belongs to element 103.

COMMENT. The case is mentioned only as example of the method. More events are necessary to identify L X-rays than K, but the numbers registered suffice.

TWG ASSESSMENT. The assignments leave very little doubt.

103; 10 TWG CONCLUSION. An important step on the way to discovery of element 103 was made in 1961 by the Berkeley group (61Gh03) although evidence fell short of full conviction, particularly in relation to the cross section curve. The Dubna papers 65Do10 and 68Fl01 taken together with 70Dr08 approached effective certainty; but it was not until the Berkeley paper of 1971 (71Es01), which confirmed earlier Berkeley and Dubna work, that it could be said that all reasonable doubt had been dispelled.

In the complicated situation presented by element 103, with several papers of varying degrees of completeness and conviction, none conclusive, and referring to several isotopes, it is impossible to say other than that full confidence was built up over a decade with credit attaching to work in both Berkeley and Dubna.

Z=103:	SUMMARY	OF PAPERS.	DATES AND	CRITERIA
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Z	Ref.	Date	Place	Remark
103	61Gh03	610413	(Berkeley)	
	65Do99	650414	(Dubna)	=65Do10
	65Do10	650420	(Dubna)	
	67Se99	670302	(Berkeley)	
	67F106	670331	(Dubna)	=68Fl01
	68Do19	68xxxx	(Dubna)	
	68F101	670810	(Dubna)	
	67F195	671019	(Dubna)	=68F108
	68F108	671105	(Dubna)	
	68Do99	680405	(Dubna)	=68Do19
	68Do19	68xxxx	(Dubna)	review
	68Le99	68	(Berkeley	Table of Isotopes
	69Ch99	690115	(Dubna)	chemistry
	70Dr08	691117	(Dubna)	·
	70F199	700218	(Dubna)	
	70Si98	700313	(Berkeley)	chemistry
	70Gh02	700417	(Berkeley)	=71Es01 for 103
	71Es01	710317	(Berkeley)	
	76Be.A	76	(Oak Ridge)	

61Gh03 $Cf(^{10,11}B,xn)^{257}103$ Cb Ey $T E_{\alpha}$ $Cf(^{12}C,\alpha xn)^{255}102$ Ey $T E_{\alpha}$

Method. Target 3%249, 33%250, 12%251, 51%252. Reaction recoils in He gas stream, through 0.05 inch orifice deposited on Cu conveyor tape, carried to solid state detectors. Presumed Z=103 activity found considerably weaker in 12 C bombardment, presumed Z=102 activity 20 times stronger. An $E_{\alpha}=8.4$ MeV group, also having $T\approx 15$ s, remains unassigned.

The three α -groups are not seen in bombardments with B on Bi, 240 Pu and 241 Am; and with 12 C on 243 Am, the latter being interpreted to show that they are not due to possible Z=101 isotopes.

The name Lawrencium (symbol Lw) is proposed for the new element, and this is the name recommended by the IUPAC Commission on Nomenclature of Inorganic Chemistry, who, however, changed the symbol to Lr. 67Se99,68Le99 Interpretation change, see also 70Gh02

The assignment of the 8(2) s E_{α} =8.6MeV isotope is shifted from ²⁵⁷ 103 to ^{258or 259} 101.

70Gh02 Narrows this down to $^{258}103~TE_{\alpha}$. And assigns 15 s E_{α} =8.4 MeV to $^{257}102$.

68Do19, 68Do99,70Dr08 Criticize this new Z=103 assignment.

```
65Do10 ^{243}Am(^{18}O,5n)^{256}103 Ey T G2 ^{243}Am(^{18}O,\alpha3n)^{254}101 T > 2 min G1 ^{238}U(^{19}F,5n)^{252}101 G1
```

Method. As in 64Do10(Z=102), the observed granddaughter is 252 Fm. The authors also see 254 Fm and assign this to 254 101 electron-capture. The collection efficiency for catching recoils of the capture decay is much lower than for α -decays.

```
68F101 ^{243} Am(^{18}O,5n)^{256}103 Ey T \approx 35 s E_{\alpha} = 8350-8500 ^{243} Am(^{18}O,4n)^{257}103 Ey T \approx 35 s E_{\alpha} = 8500-8600
```

Method. As in 66Za04(Z=102).

68Fl08 243 Am(18 O,5n) 256 103 Ey T=0.4 m E_{α} σ

Method. See 68Fl08(Z=102). One α -branch (E_{α} =8420(30)) may be of electron-capture daughter ²⁵⁶102.

70Dr08 243 Am(16 O,4n) 255 103 TE_{α}

Method. Same. Discusses the new results and emphasizes the incorrectness of 61Gh03's results.

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69Ch99 ^{243}Am(^{18}O,5n)^{256}103 Cg ^{238}U(^{22}Ne,4n)^{256}102 Cg
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Method. See 66Zv01(Z=104). The two isotopes behave like actinides, homologues of Tb and Lu.

Mentions that the name Joliotium (symbol Jo) is proposed for element Z=102. See, however, the remark at 61Gh03 above.

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70Gh02 ^{249}Cf(^{15}N,\alpha2n)^{258}103 Ey ^{TE}_{\alpha} ^{249}Cf(^{15}N,\alpha3n)^{257}103 Ey ^{TE}_{a} ^{260}105(\alpha)^{256}103 ^{TE}_{\alpha}
```

Method. See 70Gh02(Z=105). Fuller report in next item.

71Es01 Many reactions to $^{255-260}103$ Ey Cb TE_{α} G1(Tc,Ic)

Method. As 69Gh01 (see Z=104). Many details given. The results confirm the earlier results published in Dubna on the isotopes 255 (70Dr08) and 256 (65Do10+67Fl06) and corrects those on $^{257}102$ (now T=0.7 s, 8 E_{α} =8870, 2 E_{α} =8820).

```
70Si98 ^{249}Cf(^{11}B,4n)^{256}103 T=35s E_{\alpha} Ch
```

Method. He gas jet deposit. Solvent extraction chemistry. No great difference from Cf and Fm (otherwise than for last lanthanide and for Z=102 which extracts at pH values much nearer to those for Ra and Ba.)

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76Be.A ^{249}Cf(^{15}N,\alpha2n)^{258}103 ^{7}E_{\alpha} ^{249}Cf(^{11}B,^{4}n)^{256}103 ^{7}E_{\alpha}CE_{X}
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Method. See 71Di03(Z=102). Clear Z=103 L X-rays following α-spectrum assigned already to ²⁵⁸103.

ELEMENT Z=104

104; 01 The Dubna papers 64F199, 64F198 and 64F104.

The first work concerning Z=104 is described in these very similar papers. A plutonium target (composition 97% 242, 1.5% 240, 1.5% 238) was bombarded with 100-130 MeV ²²Ne ions in the internal beam of the Dubna cyclotron. The reaction recoils were caught on an endless Ni belt moved continuously past glass fission-track detectors, allowing a half-life determination of formed spontaneously fissioning nuclides. Three activities were observed, with half-lives of 13 ms, 0.3 ± 0.1 s and about 8 s. On the basis of yield curves, they were assigned to the spontaneously fissioning isomer ^{242mf}Am (formed in high yield, up to tens of nb), to ²⁶⁰104 and to ²⁵⁶102 (cross section up to 0.4 nb) respectively. The yield curve of the 0.3 s activity was found to be similar to that of ²³⁸U(²²Ne,4n)²⁵⁶102. Cross bombardments ²³⁸U + ²²Ne and ²⁴²Pu + ¹⁸O did not yield the 0.3 s activity (cross section limit 20 pb compared with a maximum of 150 pb in the above bombardment.) The possibility of an interpretation ²⁴²Pu(²²Ne,p3n)²⁶⁰103(EC)²⁶⁰102 leading to then unknown nuclides was excluded by investigating the parallel reaction sequence on ²³⁸U: the cross section for (²²Ne,p3n) was found to be 3% of that for (²²Ne,4n), and the maximum was there found to be 10-12 MeV higher. Also, a half-life as

short as 0.3 s is not acceptable for electron-capture of ²⁶⁰103.

Though it is said that "it is possible that the excitation curve for the reaction 242 Pu(22 Ne, α 2n) 258 102 is similar to that for the emission of four neutrons", assignment to 258 102 is thought to be excluded because of the low value of the ratio of its half-life and that of 256 102.

COMMENT. In the Dubna paper 68Do16, an attempt was made to find the possible α -decay granddaughter ²⁵²Fm of ²⁶⁰104 produced in the same reaction, using the same method as 64Do11, see 102; 06. The cross section for this branch was found to be smaller than 0.2 nb. This paper contains the suggestion that the 0.3 s nuclide could be ²⁵⁹104.

In the Dubna paper 70Og05, ²⁴²Pu + ²²Ne reactions yielding SF activities were reinvestigated using collimators with different collimating angles in order to distinguish between fusion/evaporation and transfer reactions. Furthermore, these experiments were made with an external beam, allowing cleaner conditions. SF half-lives were found of 15 ms and 0.1±0.05 s. The dependence on collimation agreed with assignment of the 15 ms activity to ^{242m/}Am and of the 0.1 s activity to a fusion/evaporation reaction. The latter activity was assumed to be identical with the earlier 0.3 s one. The difference was explained as due to a longlived background in the earlier experiments.

Later work in Berkeley (see 104; 03) and in Dubna (85Te99) that could lead to 260 104 only yielded an even shorter half-life ($T \approx 20$ ms). In the latter paper, the occurrence of the longer half-life is explained as due to the presence of He gas coolant around the target and the detectors. As suggested earlier by 76Gh99, this gas may carry some of the abundantly generated 3 h 252 Fm to the nearest detectors and deposit it there.

In later Dubna work (see 104; 06) it is claimed that the reported nuclide of half-life about 8 s is due to the formation of 259 104. The best present value for its half-life is 3.2 ± 0.8 s and it indeed decays for about 10 % by SF (73Dr10, 69Gh01). A later Dubna (71Fl99, Fig. 5) extension of its excitation curve (half-life reported there 4.5 s) beyond energies used in 64Fl04 shows that (evidently at variance with expectation there) the yield drops sharply, in better agreement with that for a (22 Ne,5n) reaction than with (22 Ne, 22 An).

TWG ASSESSMENT. The paper did not prove that element 104 was formed, but gave the impetus for the Dubna chemistry work discussed in 104; 02.

104; 02 The Dubna papers 66Zv99 = 66Zv01, 67Zv99, 69Zv98 and 68Zv99 = 69Zv99 (which appeared before the Berkeley work 69Gh01), and 69Zv96 = 70Zv98 = 70Zv99 (which appeared slightly later).

The physics paper 64Fl04 just discussed concludes that, "taking into consideration difficulties in the synthesis of transfermium elements, the authors believe it quite desirable to conduct chemical experiments for additional identification". Such work (which had to be quite sophisticated in view of the reported short half-life) is described in the above-mentioned papers. The paper 69Zv98 shows that "under the conditions selected for the study of element 104, the adsorption behavior of chlorides of known elements (is) in unequivocal correspondence to the heat of evaporation of these compounds." The paper 66Zv01 is "A brief communication on these experiments ... published previously". The quotations are from 69Zv99 which we will now study.

The authors used the internal ²²Ne beam of the Dubna cyclotron to bombard the same target material as used in 64Fl04. They then use a method of high speed continuous separation of gaseous chlorides developed earlier. "Recoil products emitted by a ... thin target are chlorinated by interaction with gaseous NbCl₅ or ZrCl₄. ... Analysis is continuous, and takes place in a fraction of a second from the instant of formation of the atom." The formed chlorides were then carried outside the cyclotron by a stream of N₂ gas through a 4 m long tube with "inserts of various materials" and then through a filter to a flat long chamber provided on both sides with mica detectors to detect fission fragments and their spatial distribution. Tube, filter and chamber were heated to the same temperature. The time schedule was such that the carrier gas passed the 4 m long chromatography column in 0.3 s, and the detector chamber in the succeeding 0.7 s (in two cases, 1.2 s). The authors checked carefully the functioning of the mica detectors under these circumstances, and the possible backgrounds.

Four series of experiments are described. In three series (differing in the wall material in the chromotography tube and in the composition of the chlorinating agent) a temperature of 250°C was

maintained. Only few SF events were observed: 2, 1 and 1 in series 1, 2 and 3 respectively. The bombarding energy was 114 MeV.

In the fourth series, the temperature was raised to 300° C. Ten events were then found with an integrated flux of 10×10^{17} ²²Ne particles, compared to a total of 43×10^{17} in the first three runs. In one run, the bombarding energy was raised to 128 MeV. Then, with an integrated flux of 3.2×10^{17} , no event was found. This negative result is interpreted, by comparison with the yield curves in 64Fl04, as indicating that, of the three activities reported there, only that of 0.3×10^{17} s plays a role here.

The position (and thus, presumably, time delay) distribution of the 14 observed events (see their Fig. 2) was interpreted as confirming the 0.3 s period: "it shows positively that the effect was not caused to an appreciable extent by the decay of nuclides undergoing spontaneous fission with half-lives of 0.014 and 3.7 sec."

The temperature result shows that this activity is due to an element with a chloride slightly less volatile than that of Hf but far more so than those of actinides (Z=89-103). Since no element with Z>104 could be formed by reaction of 94Pu with 10Ne, it must be 104.

These experiments were repeated and expanded in the work reported in the paper 70Zv99 which, according to a footnote, was presented at a meeting in San Francisco in April 1968. (The Berkeley paper discussed in 104; 03 appeared in the the period between this meeting and the date of receipt of the JINR preprint 69Zv96 of 70Zv99.) Now, the external beam of the Dubna U-300 cyclotron is used, offering the possibility of a revised and, probably, cleaner experimental arrangement. After chlorination, the activity is carried by N₂ gas through a 65 cm long chromatography column (transit time for carrier gas 0.065 s) into the chamber containing the mica detectors. The target material was slightly less enriched ²⁴²Pu. The transit time of the carrier gas through the detector chamber was 1.5 s. However, "It is possible that the molecules of the (104) chloride pass through the chamber more slowly than the carrier gas. If so, the experimental half-life might be lower than the true one." Experiments at 300° and 350° C gave essentially the same number of fission events per incident ²²Ne ion as in 69Zv99 but, since the irradiation doses were larger, more than 60 SF events were recorded. A time analysis of 44 of them (from the most abundant runs) gave a half-life of "about 0.5 s", but in view of the possibility mentioned, the real half-life could be longer. Cross bombardments of ²⁴²Pu and ²⁴³Am with ¹⁸O ions yielded very few SF events. Measurement of α-activity of the mica detectors after completion of the experiment showed that the Cm, Cf and Fm activities formed, and the ²⁴²Pu coming from the target, contributed at most one SF event of the 44 mentioned.

Notwithstanding the low volatility of Cm, Cf and Fm chlorides, atoms of these substances were transported through the column during the rather long course of the experiments, as measured after completion of the SF counting. The fraction in the detector chamber was 3.5% for Cm and Cf but 6.5% for a Fm activity which, based on half-life and α -energy, could be 252 Fm, 255 Fm or a mixture of both. Probably this excess was due to decay of a 260 104 or 259 104 ancestor.

The similar distributions of the (3%) Sc activity (due to bombardment of some Al in the target) and the 15% Pu over the detectors in the detector chambers were clearly *steeper* than that of the SF-counts, which makes it difficult to explain the latter as being due to a Pu SF isomer.

A last contribution to the assignment of the SF activity to element 104 was an auxilliary experiment in which 1/3 of the length of the column was replaced by a capillary of KCl which is known to bind tetravalent molecules much more efficiently than tri- or penta-valent ones. Then, "the retention of Am, Cm and Cf only slightly increased (the transfer through the column diminished to 1%)" but "no fission tracks were detected."

COMMENT. All these results must be reanalyzed in view of the now generally accepted fact that no 0.3 s SF isotope of $Z\approx104$ exists. The authors proposed (71Zv01) that the observed effects were caused instead by the 7% SF branch of 3.2 ± 0.8 s $^{259}104$ (see 73Dr10).

The quoted statement on the half-lives in 69Zv99 has been withdrawn, and not only with regard to the just-mentioned possible shift reported in 70Zv99. In an unpublished document in the possession of the TWG, Zvara calculates with Monte Carlo methods the average decay time in the 0.7 s counting interval for the case that the half-life is 4 s. From comparison with the average decay time actually found, he concludes that the latter agrees with the result of the calculation within 2 σ , and that therefore the exclusion of a 3.7 s half-life was a mistake as demonstrated from the empirical data in the paper itself.

After consulting an expert on statistical methods, the TWG made its own analysis. As to the half-life determination in 70Zv99 it is noted that it rests on the assumption that the recorded activity has died out completely at the end of the detector chamber. A better way of analysis is to consider the integrated number of counts as a function of time and to compare them with the expected function $N(1-2^{-t/T})$ to find the best value of T. For the paper 70Zv99 this procedure yields a best value T=2.1 s with an error value that generously overlaps the known half-life value of $^{259}104$. For the paper 69Zv99, a calculation of the probability that the observed SF counts agree with a half-life of more than 2 s yields a value of only a few %, but a shift in the proposed direction entirely compatible with the result found in 70Zv99 drastically increases this value.

The interpretation of the non-occurrence of SF counts at 128 MeV in 69Zv99 is based on extrapolation of the yield data of 64Fl04. If the "about 8 s" activity is due to ²⁵⁹104, the yield must drop beyond 120 MeV (the highest point measured by 64Fl04), as was indeed found to be the case later (see 71Fl99). At 128 MeV, it is only about a factor 2 lower than at 114 MeV. The statistics are so low, however, that the negative result cannot really be considered as a proof that the effect is not due to ²⁵⁹104, but it does disprove assignment to ²⁵⁶102.

The number of tracks found is said to agree with the cross section for the presumed 0.3 s activity as found by 64Fl04. Calculations by Zvara and a member of the TWG show that the SF intensities found in 64Fl04, 69Zv99, 70Zv99 and the undisputed paper 71Zv99 also agree reasonably if calculated with the excitation curve of 71Fl99 for formation of a 3.5 s activity.

The authors therefore nowadays maintain that careful study of the data in these papers demonstrates that an activity with a half-life in the range 0.3 to a few s belongs to element 104.

TWG ASSESSMENT. The Dubna chemical method depends on the dramatic increase in volatility of chlorides which is anticipated to begin with element 104. Thus, the significant point is not the determination of a half-life, but rather the transmission of an SF activity in situations where Z=104 could be formed and the absence of it when only Z<104 products are possible.

The TWG is convinced that the isotope ²⁵⁹104 indeed caused the observed effect, which was made probable in the papers before and including 69Zv99 and proved in 70Zv99.

104; 03 The Berkeley papers 69Gh98, 69Gh01 and 69Gh99; SF detection.

The first reported Berkeley work on Z=104 is described in these papers. The reaction 249 Cf + 12 C was investigated. SF decay was looked for using "an apparatus consisting of a drum rotating next to the target to catch the transmutation recoils directly". "The drum goes back and forth along its axis so that any long lived backgrounds are spread out over a very large area". Mica and lexan detectors were used to record fission fragments. An 11 ± 2 ms activity was found. Yield curves (not given) and yield (about 10 nb) were reported to agree with 249 Cf(12 C,3n) and 249 Cf(13 C,4n) reactions leading to 258 104. Arguments, though judged by the authors not fully convincing, are given that it is not 13 ms 242mf Am.

A special attempt was made to find the ²⁶⁰104 activity with a reported half-life of 0.3 or 0.1 s. It was not found in ²⁴⁸Cm(¹⁶O,4n): cross section limit a factor 4 below the calculated value. Instead, a 10-30 ms SF activity was found, which could be ²⁶⁰104.

COMMENT. Later work confirmed the assignments to ²⁵⁸104 and to ²⁶⁰104. (Best present half-life value for the latter 21±1 ms, see 85So03).

TWG ASSESSMENT. As the authors themselves indicate, this part of the work by itself does not establish with confidence the existence of element 104; see, however, next section.

104; 04 The Berkeley papers 69Gh98, 69Gh01 and 69Gh99; α detection.

In the same papers, a study is made of α -decay of possible element 104 activities. Isotopically pure ²⁴⁹Cf was bombarded with 60-100 MeV ¹²C ions. "The transmuted atoms recoiling from the target were swept by He gas from the target region ... through an orifice ... to be deposited on the periphery of a ... wheel." This wheel was rotated digitally through 39° steps (so that only after 240 steps was the same spot on the wheel rim used again) to four α -particle counting positions provided with semiconductor detectors. Stepping with time intervals comparable with the half-life, the latter could be determined.

The α -spectrum showed a number of lines with a half-life of 4.5 s, with a yield curve suggesting a 249 Cf(12 C,4n) 257 104 reaction.

For investigating the parent-daughter relation with 105 s 253 102, the four detectors were shuttled, after 200 s counting periods, to positions opposite to four others, in order to see, in a following 200 s period, the activities due to α -recoils deposited onto them. The 253 102 activity was indeed found, in intensities agreeing with the proposed parent-daughter relation.

The primary spectrum showed lines of Rn isotopes, evidently produced from a slight Pb impurity in the target. The recoil spectrum in the first counting position shows a strong ²⁵⁴102 activity, the occurrence of which could not be explained.

COMMENT. Later, the authors discovered (see 73Gh03 but also 71Gh03) that the nuclides ²⁵⁰Fm and ²⁵⁴102 possess isomeric states with half-lives of 3 and 0.2 s - quite unexpected for nuclei with even mass and charge numbers. Recoil of ²⁵⁴102 from decay to its ground state by isomeric transition explains the occurrence of its activity in the first detector.

Several results in 69Gh01 were discussed in the Dubna publications 70Dr98, 70Ak03 and 70Fl98, which claimed that they disagreed with known facts. They compared the result of a Pb+12C bombardment "under similar conditions" with the results discussed above. An experiment was also made with the collector wheel kept fixed; then "practically the same spectrum" was found, explained as due to a "gas penetration mechanism". The authors then notice that several lines found in these experiments are also found in the paper 69Gh01, and are not discussed there. Some other lines show decay characteristics which do not agree with expectation based on rather evident assignments. All of them show half-lives similar to those reported here for 257104 and 259104. The Dubna authors therefore conclude that the results for the latter cannot be trusted either.

On the basis of present day knowledge, these criticisms must be rejected, as was first discussed in the paper 71Gh03. This paper reports the results of a Pb+C irradiation under identical conditions to those in 69Gh01. They now vary the electrical bias on the wheel and the gas pressure, in order to suppress recoils due to isomeric transitions. The result is that all deviations have a now quite understandable cause.

TWG ASSESSMENT. Some perhaps at first puzzling decay characteristics of background lines have been adequately explained. The reported parent-daughter relation stays valid and is a very strong argument indeed for assigning the new activity to ²⁵⁷104. Thus, the assignments in the paper 69Gh01 are convincing.

104; 05 The Oak Ridge work 73Be33.

These experiments on the detection of X-rays following the α -decay of $^{257}104$ showed the presence of Z=102 K X-rays, caused by γ -decay of a 31 μ s delayed 300 keV level in $^{253}102$.

COMMENT. This case is mentioned here only as an example of the use of this incontrovertible criterion.

TWG ASSESSMENT. The observed 13 events are quite sufficient to demonstrate the presence of the K X-rays. This result is a very convincing confirmation of the assignment to the element Z=104.

104; 06 TWG CONCLUSION. The chemical experiments in Dubna (69Zv99 with 70Zv99) and the Berkeley experiments (69Gh01) were essentially contemporaneous and each show that element 104 had been produced. Credit should be shared.

Z=104: SUMMARY OF PAPERS, DATES AND CRITERIA

Z	Ref.	Date	Place	Comment
104	64F199	640702	(Dubna)	see 64F104
	64F198	640829	(Dubna)	=64F104
	64F104	641008	(Dubna)	
	66Zv99	660423	(Dubna)	=66Zv01
	66Zv01	660518	(Dubna)	
	67Zv99	661013	(Dubna)	Review
	68 Z v99	680401	(Dubna)	=69Zv99
	68Do16	680419	(Dubna)	
	69Zv98	680917	(Dubna)	
	69Zv99	681014	(Dubna)	
	69Gh98	690430	(Berkeley)	=69Gh01
	69Gh01	690505	(Berkeley)	
	69 Z v96	690625	(Dubna)	preprint of 70Zv98
	70 Z v98	690703	(Dubna)	=70Zv99
	70Dr98	690715	(Dubna)	comments on 69Gh01
	70Zv99	690818	(Dubna)	
	69Gh99	691117	(Berkeley)	Review
	69Zv97	691117	(Dubna)	Review
	69Ak99	691104	(Dubna)	=70Ak03
	70Ak03	691117	(Dubna)	same as 69Dr98
	70Og05	691218	(Dubna)	
	70F198	7001	(Dubna)	Review
	70Gh01	700428	(Berkeley)	
	70Si99	700805	(Berkeley)	Chemistry
	71Gh03	700915	(Berkeley)	
	71Zv99	710604	(Dubna)	preprint of 71Zv01
	71Zv01	710706	(Dubna)	Ct=temp. gradient meth.
	71F199	710711	(Dubna)	
	73Be33	730625	(OakRidge)	
	76Dr06	760225	(Dubna)	
	77Dr10	770408	(Dubna)	
	85Te99	850821	(Dubna)	

```
64F104 ^{242}Pu(^{22}Ne,4n)^{260}104 Ey Cb \sigma SF _{7}=0.3±0.1 s ^{242}Pu(^{22}Ne,\alpha4n)^{256}102 SF _{7}≈ 8 s
```

Method. Internal beam. Target 97% 242, 1.5% 240, 1.5% 238. Reaction recoils caught on endless Ni belt, carried along glass fission-track detectors. Not found in 238 U+ 22 Ne and 242 Pu+ 20 Ne. Checked for 242 Pu(22 Ne,p3n) 260 103, not seen. Later, the 8 s activity is identified with 259 104.

Method. As 66Ku15(Z=102) with added angular selection diaphragms (see 70Dr94). Tried out on the Z=102 activities, and others. Shorter half-life than 64F104's explained by long-lived background there "due to input beam".

```
71Fl99 ^{242}Pu(^{22}Ne,4n)^{260}104 Ey SF T=0.1 s ^{242}Pu(^{22}Ne,5n)^{259}104 SF T=4.5±1.0 s
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Method. See 70Og05. Gives yield curves.

66Zv01 242Pu(22Ne,4n)260104 SF Cg T

Method. "High speed continuous separation of gaseous chlorides" through long column. SF activity detected with mica detectors. The found half-life agrees with 64F104. 12 fission-tracks seen in a position expected for Eka-Hf, not for an actinide, therefore assigned Z=104.

67Zv99 Review of the above. First copy of time distribution as discussed in 69Zv99.

In 67Zv99 as well as in 67Fl97 preceeding it in the same conference proceedings, the name kurchatovium (symbol Ku) is proposed for this element (but see 69Gh99 below).

69Zv99 ²⁴²Pu(²²Ne,4n)²⁶⁰104 SF Cg

Method. Elaborate description of the 66Zv01 work.

70Zv99 242 Pu(22 Ne,4-5n) $^{260-259}$ 104 SF Cg $T \approx 0.5$ s

Method. Essentially same experiment but on external beam, higher statistics (65 events) and several new check experiments.

69Zv97 Comment on the same result.

"We deal with a Ku activity (or activities) with a half-life of about 1 s or more and also with the tail of ... 0.1 s Ku". (for 0.1 s see 70Og05 above.)

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71Zv01 <sup>242</sup>Pu(<sup>22</sup>Ne,5n)<sup>259</sup>104 SF Cg
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Method. Introduces a new and powerful technique: "adsorption of gaseous chlorides from flowing carrier gas on the surface of a glass tube, over which a temperature gradient was established." This technique has already been mentioned earlier in 70Zv97(Z=105.)

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69Gh01 ^{249}Cf(^{12}C,4n)^{257}104 Ey Cb _{\alpha} ^{T} P1(Tc,Ic) ^{249}Cf(^{13}C,3n)^{259}104 Ey _{\alpha} ^{T} P1(Tc,Ic) ^{248}Cm(^{16}O,6n)^{258}104 Ey SF ^{T} ^{249}Cf(^{12,13}C,2-3n)^{257}and ^{259}104 no SF seen
```

Method. Reaction recoils caught in He, streaming through 0.4 mm orifice in a low-pressure region, there deposited on outer rim of wheel, digitally rotated to stations with solid state detectors. For studying daughters, detectors (with α -recoils) periodically shuttled to position opposite to other detectors For SF: reaction recoils to fast rotating drum carried along mica fission-track detectors.

69Gh99 248 Cm(16 O,4n) 260 104 SF 10ms<T<30ms

Method. As above. The Berkeley authors do not find the 0.3 s (or 0.1 s) SF activity reported in earlier Dubna work. The authors therefore claim priority and suggest the name rutherfordium (symbol Ru) for Z=104. This proposal, like that of kurchatovium (see 67Zv99 above), has not been adopted by the IUPAC Commission on Nomenclature of Inorganic Chemistry.

69Dr98, 70Ak03, 70F199 Disputing the 69Gh01 discovery of ²⁵⁷104 and ²⁵⁹104.

Method. Comparing the result of a Pb+¹²C bombardment "under similar conditions" with 69Gh01's Cf+¹²C, in which a Pb impurity must have been present. Used methods: for 69Dr98: 4 s 70Mev ¹²C - recoil onto filter paper - transfer in unknown time to Si counter, counted 4 s. For 70Ak03: collector wheel method of 68Fl08(Z=102), time of irradiation and of measurement 3.2 s, transfer time on wheel to counter 0.2 s.

71Gh03 Pb+C, under conditions identical to those in 69Gh01.

Method. Same as 69Gh01. Shows that background lines not labeled in 69Gh01 were no problem. Also repeats experiment Cf+C. Here, electrical bias on wheel and gas pressure were varied to study recoil from isomeric transitions.

```
70Gh01 ^{248}Cm(^{18}O,5n)^{261}104 Ey \sigma Cb E_{\alpha} T P1(Tc,Ic)
```

Method. Same as 69Gh01.

70Si99 248 Cm(18 O,5n) 261 104 E_{α} T Ch

Method. As 70Si98(Z=103) but (cation) exchange column. Result: "entirely different from trivalent or divalent actinide element but" ... similar to Hf and Zr.

```
73Be33 ^{249}Cf(^{12}C,4n)^{257}104 E_i E_{\alpha} X
```

Method. Looks for X-rays in coincidence with selected α -rays. Finds 13 coincident K X-rays from Z=102 following α -decay from 57 104.

76Dr06 246 Cm(18 O,4n) 260 104 Ey SF As T=80±20 ms 248 Cm(16 O,4n) 260 104 Ey SF As T

 248 Cm(18 O,xn) $^{266-x}$ 104 Ey SF As no T=80 ms!

Method. As 700g05 but with very long collector tape to avoid background of 2.7 h ²⁵⁶Fm.

77Dr10 249 Cf(15 N,4n) 260 104 SF T=76.0±8.1 ms

Method. As 76Dr06

85Te99 249 Bk(15 N,4n) 260 104 As SF $T=28^{+6}_{-5}$ ms

Method. Wheel with catchers rotating along track detectors. Explains the earlier

0.3→0.1→0.08 s Dubna results as due to the He gas used there between target and catchers.

ELEMENT Z=105

105; 01 The Dubna paper 68Fl09.

This paper is the first report claiming the production of element 105, except for a short note at a Tokyo conference (68Fl04). The authors bombarded ²⁴³Am with a deflected but unfocussed beam of 123 MeV ²²Ne particles from the Dubna cyclotron. Reaction recoils were caught in He gas and deposited through an orifice on the rim of a wheel, which was rotated by a Maltese cross mechanism over 88° 48' to a semiconductor detector (see also 68Fl08 on Z=102). Two α -ray lines were observed, of $9400\pm100 \text{ keV}$ with a half-life of 0.1-3 s, and of $9700\pm100 \text{ keV}$ with a half-life of >0.05 s.

After the arrival of pulses due to these α -rays, the irradiation was interrupted for 160 s in order to wait for the possible recording of α -particles of daughter products in the energy region 8.35-8.6 MeV with half-lives of about 35 s, reported for $^{256}103$ and $^{257}103$. These were indeed observed in delayed coincidence with both lines mentioned above; in contrast with the results with a 10100 keV line also observed.

The cross section for the 9400 keV line was about 0.2 nb, for that of 9700 keV 0.1 nb, rather less than the expected value of about 1 nb. This may be explained by the possibility that the most intense part of the α -spectrum could have been concealed under the rather high background at somewhat lower energies due to reactions with slight Pb impurities (see 71Dr01).

In view of the above results and the energies expected for the Z=105 isotopes, the 9400 keV and 9700 keV lines were assigned to $^{261}105$ and $^{260}105$ respectively.

COMMENT. Since the half-life of $^{257}103$ is now known to be as short as 0.7 s and its α energies above 8800 keV, the possible Z=105 mother activity can only be assigned to $^{260}105$. But later experiments (see 105;06) showed for 1.5 s $^{260}105$ only lines in the 9030-9120 keV region (see 105; 06), and only 8930 keV for 1.8 s $^{261}103$ (71Gh01).

TWG ASSESSMENT. The reported evidence cannot be accepted as an indication for the existence of element 105.

105; 02 The Dubna papers 70Fl99, 70Fl10, 70Fl11, 70Fl16, 70Fl04 and 71Fl02.

The first paper describes a continuation of the investigation of the same reaction, at 119 MeV, now looking for fission fragments of Z=105 isotopes. Such activities were found, with half-lives of 14 ms (interpreted as 242mf Am) and 2.2 ± 0.5 s. In order to investigate whether the latter activity, in contrast with the shorter-lived one, was not due to a transfer reaction, the yields were compared behind two collimators with different angular selections (as in 70Og05 see 104; 01.) In agreement with expectation, the yield ratio for the 2.2 s activity, 2.2 ±0.5 , was rather smaller than that 5.0 ±0.5 for the 14 ms one.

In order to answer the question whether this activity might be due to a $(^{22}\text{Ne},\alpha\text{xn})$ reaction, irradiations of the same target material were made with ^{18}O . The possible products $^{256}103$ and $^{256}103$ showed very little SF activity. Based on systematic trends, the then unknown Z=103 isotopes with mass numbers 258 and 259 (formed in $(^{22}\text{Ne},\alpha\text{xn})$) reactions with x=3 and 2, which should have much larger cross sections than those with x=4 and 5) are expected to show even less SF.

The cross section ratios for (22 Ne,pxn) and (22 Ne,axn) reactions (not necessarily the same x) are known to be very small, and in addition "the known 0.1 s SF is not found".

Thus, it was concluded, the 2.2 s nuclide must be an isotope of element 105.

In the paper 70Fl10, that appeared after the Berkeley report analyzed in 105; 03 (and in the succeeding reports 70Fl11, 70Fl04, 70Fl16 and 71Fl02 on the same experiments reported in this paper and the preceeding one), the yield-curve for the 2.2 s (with a maximum cross section of 0.5 nb at 117 MeV) is found to resemble closely that for the ²³⁵U(²²Ne,5n)²⁵²102 reaction with a maximum at a slightly higher energy. The conclusion is therefore drawn that this nuclide is formed in a ²⁴³Am(²²Ne,4n)²⁶¹105 reaction, though the formation of ²⁶⁰105 is not quite excluded.

COMMENT. In later work, ²⁶¹ 105 was indeed found to have about 30% SF (see 79Dr07.) As observed in Oak Ridge (see 77Be36), ²⁶⁰ 105 has a similar half-life, and only 9.6±0.6% SF.

TWG ASSESSMENT. Two factors influence the weight to be given to the first of these reports. First, one must consider application of the criteria used here for assigning the charge (and mass) numbers to SF activities. The angular selection and yield curve results look convincing, but one should not forget that this seemed also to be the case for the 0.1 s (originally 0.3 s) activity first assigned to Z=104 but later shown to be probably an artifact (see 104; 01). The arguments used to exclude Z=103 are suggestive but cannot be considered to have been fully convincing at the time of receipt of this paper.

The second factor is that 70Fl99 is a JINR report, not one in a journal of "internationally recognized" standing. It is dated 2 months before the "regularly" published Berkeley paper to be discussed in the next section. This particular JINR report has not even the status of a preprint, in contrast to the somewhat later JINR reports 70Fl11 and 70Fl04. It might be argued that this special paper is not automatically acceptable as dated evidence.

The TWG is of opinion that the latter factor is unimportant here, in view of the fact that even the (virtually identical) "fully acceptable" papers 70Fl16 and 71Fl02 (received only about 4 months later by the editors) must be given the same characterization, namely suggestive but not fully convincing. The TWG therefore accepts in this case that the paper 70Fl99 is a positive but certainly not final step on the road to discovery of element 105.

105; 03 The Berkeley paper 70Gh02.

Using the method described in 104; 03, the Berkeley scientists found in bombardments of a 249 Cf target with about 85 MeV 15 N ions a 1.6±0.3 s activity with three α -particle groups of about 9100 keV. Cross bombardments with 14 N, and with 15 N on Hg and Pb, failed to show this activity. A parent-daughter relation was found with 30 s 256 103. Checks were made to exclude the possibility of recoil through isomeric transitions. The new activity was therefore assigned to 260 105.

The yield curve showed a maximum of about 3 nb at 85 MeV and was considered to confirm this assignment.

The results mentioned in 105; 01 are discussed in this paper which reports that they cannot find any evidence for 9400 and 9700 keV lines.

COMMENT. This result is fully confirmed, see 105; 05 and 06.

TWG ASSESSMENT. This paper leaves very little doubt about the existence of the element with z=105.

105; 04 The Dubna chemistry paper 70Zv97

This first paper on the application of the thermal gradient version of the gas-chromatography method tries to demonstrate that the chloride of the SF activity discussed in 105; 02 has a volatility which nearly agrees with that of NbCl₅ rather than HfCl₄ (and thus element 104), and which therefore strongly points to an assignment to element 105. The method has already been mentioned in 104; 06.

The locus of deposition was a somewhat diffuse area coinciding with that of Hf isotopes. It was to be expected, though, that the supposed 2 s activity tends to be deposited earlier than more long-lived isotopes of the same element, that is at positions of higher temperatures. The corrected position must be in more reasonable agreement with that for Nb activities.

A further analysis is given e.g. in 74Zv98, where the half-life shift is studied with 3.3 min and 12 h isotopes of the element Hf. The result is used to estimate the shift for the even shorter lived SF nuclide, which then indeed agrees with the expected position for an eka-Nb nuclide.

Experiments using bromides of the same nuclide are reported in 76Zv99. About the shift correction, here also necessary, it is mentioned that "A calculation of the shift ... is hindered by the lack of a satisfactory theory of the process."

COMMENT. In much later work (85Zv99) a theory for the half-life shift was developed. The result confirmed that the shifts for Hf and element 105 for the same half-lives were equal, so that the applied correction was correct.

TWG ASSESSMENT. Without the latter information, one could not exclude the possibility that the observed effect (with low statistics: only 18 events) was due to a mixture of isotopes of element 104.

105; 05 The Dubna paper 71Dr01.

This paper describes an improvement on the work reported in 105; 01. A 243 Am target was used with 100 times less Pb impurity, and the intensities of the transfer reaction products were further reduced by use of a collimator before the catcher. The reaction recoils were caught in a gas subsquently pumped through collectors which were, after an irradiation period, transported to semiconductor detectors. Later, the latter were also analyzed for α -decay daughters deposited on them by α -recoil. A 9.1 MeV α -peak with a half-life of $1.4^{+0.6}_{-0.4}$ s was found in "delayed" coincidence with α -particles of 8.3-8.6 MeV with a 35 s half-life, assigned to $^{256\alpha r 257}_{-0.4}$ 103, and their parent therefore to $^{260\alpha r 261}_{-0.4}$ 105. In addition, a 8.9 MeV peak was assigned to a Z=105 isotope.

COMMENT. As concluded almost simultaneously in 70Gh02, the 9.1 MeV α -particles belong to 260 105.

TWG ASSESSMENT. This work shows convincingly the existence of at least one isotope of element 105, and agrees in this respect with the Berkeley work 70Gh02.

105; 06 The Oak Ridge paper 77Be36.

This paper describes the detection of X-rays following the α -decay of $^{260}105$ (α -energies 9031, 9074 and 9120±14 keV). It showed Z=103 L X-rays in coincidence with the α -particles.

TWG ASSESSMENT. An L X-ray pattern is rather more complex than a K. Nevertheless the observed 151 events suffice to show most convincingly that the X-rays belong to Z=103, and therefore that the parent had Z=105.

105; 07 TWG CONCLUSION. Independent work reported in 1970 from Berkeley (70Gh02) and from Dubna (71Dr01) was essentially contemporaneous and equally convincing. Credit for the discovery should be shared.

Z=105: SUMMARY OF PAPERS, DATES AND CRITERIA

Z	Ref.	Date	Place	Remark
105	68F104	670713	(Dubna)	short note
	68F109	680411	(Dubna)	
	70F199	700218	(Dubna)	early version of 70F116
	70Gh02	700417	(Berkeley)	•
	70F110	700508	(Dubna)	early version of 70F116
	70Zv97	700515	(Dubna)	•
	70F111	700608	(Dubna)	preprint of 70F116
	71Dr01	700615	(Dubna)	• •
	70F116	700630	(Dubna)	see 71Fl02
	70F104	700713	(Dubna)	preprint of 71F102
	71F102	700728	(Dubna)	• •

Z	Ref.	Date	Place	Remark
	73Zv99	731113	(Dubna)	preprint of 74Zv98
	74Zv98	7312	(Dubna)	
	74Zv99	740402	(Dubna)	review, including 70Zv97
	76Zv99	750416	(Dubna)	•
	77Be36	770509	(OakRidge)	

68Fl09 243 Am(22 Ne,4n) 261 105 E_i G1(Tc,Ic) E_{α} σ T 243 Am(22 Ne,5n) 260 105 E_i G(Tc,Ic) E_{α} σ T

Method. Apparatus as 68F108(Z=102). Unexpectedly low cross sections, possibly due to the main α -spectrum being hidden below lower energy backgrounds.

71Dr01 243 Am(22 Ne,5-4n) $^{260-1}$ 105 As TE_{α} MeV G1(Tc,Ic)

Method. An Am sample with a considerably reduced Pb contamination. A parent-daughter relation was found for both α -rays with a 35 s daughter with 8.35-8.6 MeV α rays.

70Fl99 243 Am(22 Ne,4n) 261 105 SF σ As T 243 Am(18 O,xn) $^{261-x}$ 103 no SF seen

Method. See 64Fl04(Z=104). The reported half-life is that of a Z=105 isotope proper, not of a daughter.

70F110 243 Am(22 Ne,4n) 261 105 SF Ey σ T 235 U(22 Ne,5n) 252 102 SF Ey Cb

Method. Same as above. Yield curves have been compared for the two reactions.

Method. Combination of the two preceding sets of data. Intensity ratio with two collimators now 0.18(.02) for transfer reaction to 242mf Am, 0.47(0.10) to 2s SF strongly points to a compound nucleus reaction.

The authors propose for this element the name nielsbohrium, as we found first mentioned in the review paper 71Fl07 (but see 70Gh02 below).

70Zv97. 243 Am(22 Ne,4n)261 105 SF Cg

Method. As in 71Zv01(Z=104), using chlorides. A γ -spectrometer is used for comparison nuclides. Mica fission-track detectors. The resulting track distribution agrees with Hf (as for Z=104) but must be corrected for a position shift due to the short half-life.

74Zv98. 243 Am(22Ne,4n)261 105 SF Cg

Method. Further analysis of the results in 70Zv97. The half-life shift is studied with two Hf isotopes with different half-lives. It is concluded that the shifted position agrees with Nb and not with Hf.

76Zv99. 243 Am(22Ne,4n)261 105 SF Cg

Method. As in 71Zv01(Z=104) but using bromides. Corrections for half-life shift as in 73Zv99. Found volatility much higher than for actinides and even, after the correction, than for Hf (or, element 104).

70Gh02 249 Cf(15 N,4n) 260 105 Ey Cb T) s E_{α} G1(Tc,Ic)

Method. See 69Gh01 (Z=104). Daughter: $^{256}103$ (T=30 m, $E_{\alpha}=8400$). This paper reports being unable to confirm the 68Fl09 etc. results, therefore claims discovery. Suggested name for the element: hahnium, element symbol Ha. Neither this name, nor the name nielsbohrium (71Fl02 above) has been adopted by the IUPAC Commission on Nomenclature of Inorganic Chemistry.

77Be36 249 Cf(15 N,4n) 260 105 E_i T SF E_{α} X

Method. As 71Di03(Z=102). Observes 150 counts in Z=103 L X-ray region in coincidence with α -rays. Best possible assignment to Z=105.

ELEMENT Z=106

106; 01 The Dubna papers 74Og99 and 74Og04.

The paper 74Og04, preceded by the JINR report 74Og99, describes the first attempt to produce previously unknown elements by the "cold fusion" method (see II.2D). Exploratory experiments were interpreted as showing that "the background due to SF of the heavy elements is practically eliminated, as is the background due to the SF isomers in the region from U to Cf". Also, "a large number of control experiments in which various isotopes of Pb and Bi were bombarded with 51 V and 52 Cr ions" showed that the background from reactions of the type (HI,pxn) and (HI, α xn) (x=0, 1, 2) "is negligibly small".

In bombardments of 208 Pb and 207 Pb with 54 Cr ions, 20 and 31 SF events respectively were found with a half-life between 4 and 10 ms. Such an activity was not found in Pb + 52 Cr and Bi + 51 V, which is taken as an indication that this activity is not due to a transfer reaction leading to nuclides with Z substantially less than 106. In view of the results mentioned above, the authors "are therefore inclined to assume that the 50 events observed in the experiment are due to SF of nuclei with Z=106". The specific isotope is "assumed" to be $^{259}106$ on the basis of "the yield of this emitter in experiments with different Pb isotopes, and also on the basis of the new ideas concerning the systematics of the SF".

COMMENT. The conclusion that SF backgrounds are insignificant has later been fully confirmed. The earlier-dated JINR report 74Og99 gives even more information than 74Og04. Significant for the absence of SF isomers in this type of work is that neither in ²⁰⁶Pb+⁵⁴Cr nor in ²⁰⁸Pb+⁵²Cr were indications found for any production of SF nuclides.

On the other hand, the partial half-life of $^{259}106$ was found to be larger than 100 ms in later Dubna work (84De07), and Darmstadt (85Mu11) reported a half-life of 0.5 s and less than 20% SF, whereas $^{260}106$ has a half-life of 2.5±1.5 ms (84De07) or 3.6 $^{+0.8}_{-0.6}$ ms (85Mu11) and decays by α -decay to 7.4±0.8 ms $^{256}104$ (which itself decays almost exclusively by SF) and for $50^{+30}_{-20}\%$ (85Mu11) or <20% (84De07, as found by measuring a growth-and-decay curve) by direct SF. The combined decay curve for $^{260}106$ and its daughter does not disagree with the half-life estimate given by 74Og04.

The conclusions of this work on the production of $^{256}104$ and $^{255}104$ were confirmed later, with the possible comment that the 4 s half-life reported for the latter is somewhat larger than the later value 1.5 ± 0.2 s (86He08).

TWG ASSESSMENT. It seems likely that, in the work reported in 74Og04, ²⁶⁰106 was formed, observed and assigned to the right element. The TWG concludes, however, that this paper, lacking as it is in yield curves and angular selection results, does not yield sufficient confidence that the isotope ²⁶⁰106 was formed.

106; 02 The Berkeley-Livermore paper 74Gh04.

This paper describes the results of the investigation of the reaction products in the bombardment of a 249 Cf target with 18 O ions, using equipment which is similar to that used before on element 104 (see 104; 04, 69Gh01). The reaction recoils were caught in a He gas stream and deposited on the rim of a wheel rotating them to detector sets so arranged that correlations with daughter products could be studied. By adding NaCl aerosol to the He gas, causing the active deposit to be embedded in a thin NaCl layer, care was taken that recoils from the deposit to the detectors could not be caused by isomeric transitions, but only from α -decay. As an extra feature, stopping the wheel after detection of a possible daughter allowed granddaughters to be seen.

The authors found 87 α -events in two groups, one at 9060 keV and (according to their Fig. 2) a 10 times weaker one at 9250 keV; of these, only 14 ± 3 could be assigned to a background. The remainder decayed with a half-life of 0.9 ± 0.2 s. The parent-daughter analysis yielded α -particle energies and a half-life agreeing with known data for $^{259}104$ in approximately the right intensity. The granddaughter $^{255}102$ was also found in the right intensity. (The annual report note 75Gh.A gives detailed data on the genetic relations.) The production cross section for 95 MeV ^{18}O ions was 0.3 nb, and little was found at 91 and 100 MeV, in agreement with expectation for the reaction $^{249}Cf(^{18}O,4n)^{263}106$.

COMMENT. These results are confirmed by those reported in 75Be.A and 79Dr07.

TWG ASSESSMENT. The assignment to the decay chain $^{263}106(\alpha)^{259}104(\alpha)^{255}102$ is very convincing.

106; 03 TWG CONCLUSION. Independent work reported in 1974 from Berkeley-Livermore (75Gh04) and from Dubna (74Og04) was essentially contemporaneous. The Dubna work is highly important for later developments but does not demonstrate the formation of a new element with adequate conviction, whereas that from Berkeley-Livermore does.

Z=106: SUMMARY OF PAPERS, DATES AND CRITERIA

Z	Ref.	Date	Place	Remark
106	740g99	740711	(Dubna)	=74Og04
	74Gh04	740909	(Berkeley-Livermore)	
	74Og04	741020	(Dubna)	
	75Be.A	7509xx	(Oak Ridge)	
	79Dr07	781212	(Dubna)	

740g04

```
^{208}Pb(^{54}Cr,3n)^{259}106 E_i Cb \sigma SF T ^{207}Pb(^{54}Cr,2n)^{259}106 E_i Cb \sigma SF T ^{206}Pb(^{54}Cr,n)^{259}106 E_i Cb \sigma SF T ^{208}Pb(^{50}Ti,2n)^{256}104 E_i \sigma SF T ^{207}Pb(^{50}Ti,n)^{255}104 E_i \sigma SF T=4 s
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Method. No description given of experimental arrangement (but see 76Og02,Z=107). Z=106 mass assignment based on production ratio on different Pb isotopes. Assignment to Z<106 eliminated on basis of cross bombardments; not in Pb+52Cr and 209Bi+51V.

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74Gh04 ^{249}Cf(^{18}O,4n)^{263}106 Ey \sigma T E_{\alpha} G1,2(Tc,Ic)
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Method. As in 69Gh01. Recoils carried through 4.8 m capillary to counting wheel set up. Wheel digitally rotated every second in 45° steps, with a 1.5° degree offset every 30 min to avoid long-lived background build up. T=3.0 s daughter detected (a) by decay from the same source position (not necessarily same detector), (b) from recoils captured by wheel monitoring detectors by shuttling them every 6 s to shielded position facing other detectors. T=3 min granddaughter detected through not returning wheel monitoring detectors from off-wheel position for 10 min after detection of a $^{259}104 \, \alpha$ -ray.

Method. Recoil nuclei collected on tape carried along glass or lavsan fission track detectors. Comparison of σ with 74Gh04 would lead to assigning 70% SF to 263 106.

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75Be.A ^{249}Cf(^{18}O,4n)^{263}106 Ey TE_{\alpha} Method. As 71Di03(Z=102). Looked for X-rays in coincidence with \alpha-rays; not found.
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ELEMENT Z=107

107; 01 The papers 76Og97, 76Og01, 76Og02.

These papers, the first ones on attempts to produce this element, describe Dubna work with cold fusion. A thin target layer on a rotating wheel was bombarded at a shallow angle, so that fission fragments from spontaneously fissioning nuclides formed could escape this layer and be detected in a number of fission track detectors surrounding the wheel. In many bombardments with ions from ⁴⁵Sc to ⁵⁴Cr on several isotopes of Tl, Pb and Bi, no SF isomer was observed. Thus it is concluded, that any new SF activity found must be due to an evaporation reaction (emission of a few neutrons).

Very little SF was observed in bombardments that could produce element 103. On the other hand, the combinations 209 Bi + 50 Ti, 208 Pb + 51 V and 205 Tl + 54 Cr (all combining to Z=105) produced a $5.0^{+1.7}_{-1.1}$ s activity, and 207 Pb + 51 V one with a half-life of $1.2^{+0.5}_{-0.3}$ s. "By comparing the calculated cross sections with the one found for SF one can conclude that the SF branch (in the 5 s activity) is about 20%."

The mass assignment for the 5 s isotope to A=257 is based on comparison of the cross section ratio 5.2 ± 1.5 for production of the 5 s activity in the reactions Bi+Ti and Pb+V with the theoretical values 4.5 and 1.5 calculated for 2n and 1n evaporation respectively. It then follows that the 1.2 s isotope is probably $^{256}105$. "Its identification, however, requires additional experiments."

Finally, then, experiments were made for producing element 107 by bombarding 209 Bi with 54 Cr and 208 Pb with 58 Mn. Two activities were found, one with a half-life "estimated to lie between 1 and 2 ms", the other one being 5^{+4}_{-2} s, both with essentially the same (partial) cross section of 90 pb. Of each, several dozens of events were seen.

In view of the low cross sections expected for (HI,pxn) and (HI, α xn) reactions, the two activities are thought to be due neither to element 106, nor to direct production of element 105. The 5 s activity is therefore thought to be identical with the $^{257}105$ activity as identified above but here due to the formation of $^{261}107$ decaying to it with a short half-life. This assignment is thought to be confirmed by the experimental ratio 3±1 of its production cross sections in the reactions Bi+Cr and Pb+Mn, agreeing with the theoretical ratio 2.8 for 2n evaporation processes.

The 1-2 ms activity is thought not to be an SF isomer since the cross sections for massive transfer reactions at the bombarding energies employed are expected to be small; also, known SF isomers like 14 ms 242mf Am were not seen. The ratio of the intensity of the 1-2 ms activity to that of 5 s was the same in all experiments. The authors therefore conclude that both activities are due to the formation of the isotope 261 107 with a half-life of 1-2 ms, decaying for the greater part by α -decay to 5 s 257 105, but partly, 20%, by SF since the latter as mentioned above also decays for 20% by SF.

COMMENT. Later, instead, the isotopes $^{257}105$ and $^{258}105$ were found to have half-lives of $1.4^{+0.6}_{-0.5}$ s and $4.4^{+0.6}_{-0.6}$ s respectively. The first one and its descendants show little SF decay, but $^{258}105$ decays $33^{+2.6}_{-0.5}$ % to the purely SF isotope 12 ms $^{258}104$ (85He22). An additional reason for identifying the Dubna 5 s Z=105 isotope with $^{258}105$ is that the Darmstadt cross section for its production via its parent agrees with the Dubna "257" one. In Dubna paper 83Og99 the assignment of the daughter is indeed revised to A=258. Nothing is said there about the half-life of the Z=107 parent, but a Dubna review paper (87Fl99) gives an increased value $2.8^{+2.1}_{-2.1}$ ms (referring to a report at a 1985 conference which, however, has not been published). Even this value is rather shorter than the Darmstadt half-lives for $^{261}107$ and for both isomers of $^{262}107$ (see 107; 02 COMMENT). And the Darmstadt cross section limit for Z=107 SF production, 14 pb, is rather smaller than the value proposed by Dubna.

TWG ASSESSMENT. The part of this work on element 105 can be accepted if the A-assignment for the 5 s isotope is corrected to A=262. The assignment of the 1-2 (or 2.8) ms SF events to one of the two isotopes of Z=107 must be considered unconvincing. The conclusion that the observed 5 s Z=105 SF events in the ⁵⁴Cr bombardments is a decay product of a short lived Z=107 parent is plausible but not proved in this early work.

107; 02 The Darmstadt paper 81Mu06.

This paper describes experiments on the same reaction but using a different and very sophisticated technique. The 54 Cr beam is made to impinge on a thin 209 Bi foil. The reaction recoils are separated in flight from the incoming beam and from products of transfer reactions by SHIP, a velocity filter consisting of a combination of magnetic and electric fields. They are then implanted in position-sensitive solid state detectors. These allow the observation of energies of α -particles (in the slightly over 50 % of the cases where they do not leave the detector) or SF events, in position- and time-correlation with one another and with the time of implantation. Also, combination of the observed energy of the impinging ions with the time of flight can be used to obtain an estimate of the masses of these particles (see 109; 01.) Thus, very selective 'fingerprints' can be obtained.

In this first Darmstadt publication on element 107 results of irradiations at two energies, 252 and 257 MeV, were described. The authors observed 5 decay chains initiated by a 10376 keV α -transition with a half-life of $4.7^{+2.3}_{-1.6}$ ms, and one with an energy of 9704 keV occurring after 165 ms. Three of these showed a granddaughter with a half-life and an α -particle energy that could be identified with ²⁵⁴103, and two and one respectively with further descendants assigned in the same way to ²⁵⁰Fm and ²⁵⁰101. It was therefore decided that with high probability the original parent was ²⁶²107.

The α -decay of the direct descendant $^{258}105$ of this nuclide had not been reported before. Therefore, experiments were made in which 209 Bi was bombarded with 50 Ti ions of 238 MeV. They showed a $4.0^{+1.8}_{-1.6}$ s nuclide with two α -particle energies, 9189 ± 35 and 9066 ± 35 keV, assigned to $^{258}105$ on the basis of its observed decay to $^{254}103$ and its successors. (Also, an SF activity with a half-life of $1.6^{+0.5}_{-0.5}$ s was observed. At higher energies, 243 and 248 MeV, these activities disapppeared but "decay chains of $^{257}105$ ending in the sequence 249 Md- 245 Es appeared".)

These data agree with the $1.8^{+1.1}_{-0.7}$ s half-life and 9181 ± 35 and 9104 ± 50 keV α particle energies found in the ⁵⁴Cr bombardments.

COMMENT. In later Darmstadt work 89Mu09 the following values were obtained for the half-lives: for the $^{262}107$ isomers 8.0 ± 2.1 and 102 ± 26 ms and for $^{261}107$ $11.8^{+5.3}_{-2.8}$ ms.

TWG ASSESSMENT. This work is considered sufficiently convincing, and was confirmed in 1989 experiments (89Mu09).

107; 03 TWG CONCLUSION. The Darmstadt work 81Mu06 provides convincing evidence for the formation of element 107. The preceding Dubna work (76Og01 and 02) reports events with Z=105 which quite probably were due to the formation of element Z=107; yet, taken alone, this work does not give sufficient confidence that element Z=107 was in evidence.

Z=107: SUMMARY OF PAPERS, DATES AND CRITERIA

Z	Ref.	Date	Place	Remark
107	76Og97	760129	(Dubna)	=76Og01
	76Og01	760130	(Dubna)	see 76Og02
	76Og02	760809	(Dubna)	
	79Bu99	79	(Dubna)	
	81Mu06	810403	(Darmstadt)	
	83Bu99	83	(Dubna)	

76Og02

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<sup>209</sup>Bi(^{54}Cr,2n)<sup>261</sup>107 E_i SF T σG1(Tc) Br(80% α)
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Method. Ions impinging under small angle on rotating 10 cm wheel covered with thin target layer, surrounded with mica fission-track detectors. The 1-2 ms activity and its daughter were not found with ²⁰⁹Bi(⁵³Cr,2n).

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79Bu99 ^{249}Bk(^{22}Ne,4n)^{267}107 E_i SF As T \sigma
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Method. Extracted beam, recoils to running tape, see 76Dr06, 77Dr10, 79Dr07. Background ²⁵⁶Fm reduced factor 7 by collimation, remaining effect twice background. (²²Ne,α1-3n) not excluded.

83Bu99 Same experiment

Method. Same. Extensive discussion, conclusion: "Z=105-107 can be excluded".

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81Mu06 ^{209}Bi(^{54}Cr,n)^{262}107 E_i G123(Tc,Ic,Pc) E_{\alpha} Tf As T
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Method. SHIP facility: from recoils from thin target, evaporation residues are selected in flight by a Wien filter, and by time of flight low resolution mass spectrometry from products of more complicated reactions and implanted in position-sensitive semiconductor detectors. Combined measurement of elaborate α -decay chains. Z and A assignment through decay to 250 Fm and 246 Cf.

ELEMENT Z=108

108; 01 The Dubna paper 83Og99.

This report on the production of element Z=108 describes Dubna work, in a note in conference proceedings. This work is included in the reports discussed in the next paragraph 108; 02.

The method used was identical to that used before, see 76Og02(Z=107). In bombardments of ²⁰⁹Bi with 305 MeV ⁵⁵Mn ions, 20 SF tracks were found of which half showed a time distribution corresponding to a half-life of 1.9½% s (the other half were observed in another run in a time interval too short compared with this half-life). They were explained as due to the granddaughter ²⁵⁵104 of the nuclide ²⁶³108, formed with a cross section of about 4 pb (reduced in the later reports to 2 pb) and, together with its direct daughter, having a half-life somewhat shorter than 1.5 s.

COMMENT. No later report on $^{263}108$ is known. A later Darmstadt paper 85Mu11 assigns a half-life of 0.5 s and >80% α -decay to the $^{259}106$ daughter, and the reported half-life value of $^{255}104$ agrees with later values (85He06).

TWG ASSESSMENT. The data reported in this work are suggestive but not sufficient to give much confidence that effects necessarily due to the formation of element 108 were observed. The assignment of a 1 s SF activity to ²⁵⁵104 can not be considered as unique.

 $^{^{208}}$ Pb(55 Mn,2n) 261 107 E_i Same σ , T

 $^{^{209}}$ Bi(50 Ti,2n) 257 105 E_i SF T σ

108; 02 The papers 84De99 and 84Og02.

These papers, reporting similar experiments with 58 Fe ions impinging on 208 Pb and 207 Pb, showed 13 and 8 SF events respectively, with half-lifes of 8^{+20}_{-4} ms and 6^{+5}_{-2} ms, formed with cross sections of 2 pb and 5 pb, and initially assigned to the formation of 264 108, though assignment to 265 108 was not excluded (84De99), and later clearly assigned to 264 108 (84Og02).

Also, a completely different way of trying to demonstrate the formation of a new element is introduced in these papers (see also 83Kh99, Z=109 SUMMARY). One looks for a descendant, which can be separated chemically and determined efficiently, after production of its ancestor belonging to the new element in a reaction that, on good grounds, is thought not to produce any other of its ancestors. This method allows use of bombardments much longer than the half-life of the new ancestor, and therefore promises useful results even if the production cross section is very small. It is, moreover, not dependent on the shortness of that half-life.

For studying $^{265}108$ in this way, 3.0 d ^{253}Fm or 20 d ^{253}Es are suitable choices. Indeed, after bombardments of ^{208}Pb targets with ^{58}Fe , ^{54}Cr and ^{50}Ti , the authors found 3 and 34 α -events of ^{253}Es and 72 of ^{253}Fm respectively. In deriving cross section values, it is assumed that the $^{257}104$ nuclide decays about 35% to ^{253}Es .

COMMENT. The observed SF is related to the formation of $^{264}108$ and occurs in $^{260}106$ (3 ms) following a 1 ms α -decay.

The decay path of ²⁵⁷104 and the percentage mentioned are acceptable, even though no decay data on the link involving the nuclide ²⁵³101 are known. (The mentioned Dubna result indicates that it must decay, with a half-life of less than 3 days, by electron-capture to ²⁵³Fm.)

TWG ASSESSMENT. Experiments by the latter method, even if it is accepted that the activities found must belong to somewhat remote descendants of a Z=108 activity, only demonstrate that the latter is an α -emitter with a half-life shorter than several hours, formed with a certain cross section. No data are known on yield curve or angular selection. Also, in view of the very small cross section for the assumed process, it is difficult to exclude other processes that could produce lower-Z ancestors of the observed activities. It is noticed, on the other hand, that cross section values reported by Dubna are, in fact, even somewhat *smaller* than those found in Darmstadt. It can therefore be said, in retrospect, that very probably element 108 played a role in the Dubna experiment.

108; 03 The Darmstadt papers 84Mu17 and 87Mu15.

Using the equipment described in 107; 03, the Darmstadt group found three relevant decay chains in bombardments of 208 Pb with 58 Fe. The $1.8^{+2.7}_{-0.7}$ ms parent decays to a 110^{+140}_{-40} ms nuclide of which one track has an energy of 9570 keV. (In the other two cases, the α -particles evidently leave the detector before expending their full energy.) These values should be compared with the half-life 260^{+110}_{-80} ms and the energy 9560 ± 30 keV found in 54 Cr bombardments of 208 Pb and "unambiguously identified" as due to 261 106. Also, the α -energies and the $6.0^{+2.7}_{-2.1}$ s half-life for the granddaughter in the abovementioned three decay chains agree with the values for the daughter 257 104 of 261 106. It is therefore concluded that the three decay chains belong to the isotope 265 108. It is then formed with a cross section of 19^{+18}_{-11} pb.

TWG ASSESSMENT. The Darmstadt work in itself is sufficiently convincing to be accepted as a discovery.

108; 04 TWG CONCLUSION. The formation of element 108 was established by simultaneous and independent work in Darmstadt (84Mu17) and Dubna (84Og02). The work in Darmstadt is more detailed and, of itself, carries conviction. In retrospect, the Dubna work taken together with cross section measurements in Darmstadt leaves little doubt that element 108 was produced and was inferred correctly through identified descendants following a satisfactorily-understood decay path. The major credit should be accorded to Darmstadt.

Z=108: SUMMARY OF PAPERS, DATES AND CRITERIA

Z	Ref.	Date	Place	Remark
108	83Og99	830421	(Dubna)	
	84De99	840410	(Dubna)	precursor 84Og02
	84Mu17	840419	(Darmstadt	see 87Mu15
	84Og02	840614	(Dubna)	
	87Mu15	870529	(Darmstadt)	

83Og99 ²⁰⁹Bi(⁵⁵Mn,n)²⁶³108 SF T σ

Method. As 76 Og 02 (Z=107). Ten SF tracks (half-life roughly 1 s) and ten others without time information are assigned to the $^{255} 104$ granddaughter. Then either cross section unexpectedly low or high SF/ α ratio of grandparent with half-life too small to be seen in this experiment.

84Og02 209 Bi(55 Mn,n) 263 108 E_i SF σ 208 Pb(58 Fe,2n) 264 108 E_i SF σ 207 Pb(58 Fe,n) 264 108 E_i SF σ 7 208 Pb(58 Fe,n) 265 108 E_i G6 σ

Method. As above and as in 83Kh99(Z=109). Bi+Mn experiment probably same data as 83Og99. The 20 ²⁶⁴108 SF tracks are assigned as possibly from the isotope itself but more probably belonging to the descendant ²⁵⁶104. The descendant seen in the G6 relation is ²⁵³Es.

87Mu15 208 Pb(58 Fe,n) 265 108 E_i G1,2(Tc,Ic,Pc) Tf As σ T Method. As 81Mu06(Z=107). Z and A assignment through α -decay chain to 253 102.

ELEMENT Z=109

109; 01 The Darmstadt papers 82Mu15 and 84Mu07.

In bombardments of 209 Bi with 58 Fe ions discussed in these papers, only one decay chain supposed to start with a Z=109 isotope was found. Though the only valuable data on the daughters were the time delays between production and α - and SF decay respectively of the daughter and the granddaughter, their tracks were found to be position-correlated. The SF event was, moreover, the only one seen in the whole experiment.

The time of flight measurement combined with a determination in the counter of the energy of the implanted ion causing this decay chain yielded a value $A=264\pm13$ for its mass number; it was one out of only 3 events with mass number above 250. Also, it was one out of only 5 events with initial α -particle energy in the range 9.5-13 MeV and half-life in the ms range (as expected for Z=109), and the only one which combined the two characteristics. The decay chain of the daughter can easily be identified with a known decay branch of $^{262}107$, and not with anything else. It is concluded, with a very high degree of certainty, that this event must be due to an atom of $^{266}109$. The cross section for its production is then about 10 pb.

COMMENT. In a later Darmstadt paper 88Mu15, two more events were found. One must be assigned to a known longer decay branch of $^{262}107$ ending with an event assigned to the α -decay of $^{254}102$. The other event could, in principle, be due to a (58 Fe, α n) reaction, but this is thought quite improbable since observed relative cross sections for direct α -emision are small and, in addition, SHIP (see 107; 02) supresses arrival of such products on the counters by an order of magnitude. In combination with the above-mentioned case, the cross section is found to be 10^{+30}_{-3} pb.

TWG ASSESSMENT. The result is convincing, even though originally only one event was observed.

109; 02 The Dubna paper 85Og99.

Both by searching for SF events and, after radiochemical separation, for the 1.5 day descendant 246 Cf, the authors looked for the production of element 109 in the same reaction. One SF event, possibly due to the same decay chain first mentioned in 109; 01, and 7 α -events with the 246 Cf energy were seen. The resulting cross section for production of 266 109 is 3 pb. In an earlier report 83Kh99 of the Dubna group no events of 246 Cf were seen, leading to an upper limit of about 6 nb.

TWG ASSESSMENT. The results agree reasonably with those of the Darmstadt group and can be considered as a confirmation. Conversely, the lower Dubna cross section value is an indication that very few other ancestors of ²⁴⁶Cf can have been formed in this cold fusion reaction.

109; 03 TWG CONCLUSION. The Darmstadt work (82Mul5) gives confidence that element 109 has been observed.

Z=109: SUMMARY OF PAPERS, DATES AND CRITERIA.

Z	Ref.	Date	Place	Remark
109	82Mu15	821008	(Darmstadt)	see 84Mu07
	83Kh99	830421	(Dubna)	
	84Mu07	830905	(Darmstadt)	
	85Og99	850110	(Dubna)	
	88Mu15	881212	(Darmstadt)	

84Mu07 209 Bi(58 Fe,n) 266 109 E_i Tf As G123(Tc,Ic,Pc) E_{α} σ

Method. See 81Mu06(Z=107); here extensive discussion of dependability. Only one decay chain, ending with SF of ²⁵⁸104.

88Mu15 209 Bi(58 Fe,n) 266 109 E_i G123(Tc,Ic,Pc) E_{α} T σ

Method. Continuation of the above. Two new events; for one of them the first transition is missing. For all three events: $\sigma=10^{+10}_{-10}$ pb.

83Kh99 209 Bi(58 Fe,n) 266 109 E_i G6 σ 209 Bi(55 Mn,2n) 262 108 E_i G5 σ 209 Bi(54 Cr,n) 262 107 E_i G5 σ 208 Pb(55 Mn,n) 262 107 E_i G5 σ 209 Bi(50 Ti,n) 258 105 E_i G4 σ

Method. Chemical separation of 246 Cf descendant (identified by T and E_{α}). No daughter of 266 109 seen in the first reaction, contrary to expectation based on Darmstadt cross section. In the two last reactions, ≈ 100 cases seen. No similar activity is seen in 58 Fe and 55 Mn bombardments on 209 Bi, thus production in all reactions not transfer but neutron evaporation.

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85Og99 ^{208}Pb(^{59}Co,n)^{266}109 E_i G6 \sigma ^{209}Bi(^{58}Fe,n)^{266}109 E_i G6 \sigma
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Method. See 83Kh99, observation of 7 α -tracks of ²⁴⁶Cf assigned to Z=109 decay, as well as one observed SF event, assigned to the descendant ²⁵⁸104 of ²⁶⁶109.

ELEMENT Z=110

110; 01. The Dubna papers 86Og99 and 87Og99.

The cross sections for cold fusion reactions to produce element Z=110 are expected to be extremely small. The Dubna workers therefore tried another method: bombarding Th and U isotopes with Ca and Ar ions. The targets were thin ring-shaped foils rotated for cooling. Behind the irradiated position was another rotating wheel with 26 thin collector foils. Fission-track detectors were mounted on both sides of the collector wheel. Collimators were used to reduce the number of formed SF isomers caught by the collector foils. Their number was further reduced by a judicious choice of the catcher foil thickness. For this purpose, a study was made of the behaviour of the known fission isomers 0.9 ms ^{240mf}Am and 14 ms ^{242mf}Am formed in such bombardments by exchange reactions.

In bombardments of ²³⁶U with 212 MeV ⁴⁰Ar, SF activities were seen with half-lives of 1 and 10 ms. The first one was identified with ^{240mf}Am, but (as calculated from the expected production ratio) only about 20% of the 10 ms activity could be explained as ^{242mf}Am.

Experiments at a few other energies yielded the result that "the excitation function for ... ^{242m}/Am ... and for the activity in question are very different from one another." (The upper limit for the cross section for the latter is about 12 pb.) "However, it is not excluded that with such small cross sections some unforeseen irregularities may manifest themselves in the energy dependence of the cross sections for production of the isomers ^{240m}/Am and ^{242m}/Am."

With several other similar reactions, "it was shown that the contributions coming from (HI,pxn) and (HI, α xn) channels are small compared with the main (HI,xn) channel for all x values from 0 to 3. There are all grounds to believe that" ... this is also true in the above case. The rather peaked excitation curve in the Th experiment, and the strong decrease in the U one for E_i above 215 MeV, then suggest assignment of the 10 ms activity to Z=110 with mass number 272 or, perhaps, 271.

TWG ASSESSMENT. The results are suggestive but not yet sufficient to prove that element 110 has indeed been produced.

110; 02 The Darmstadt paper 90Sc11.

In this work, a search was made for long-lived heavy actinides that could have been produced by decay of Z=110 atoms formed in reactions of ⁴⁰Ar with ²³³U and ²³⁵U. The upper limit for the cross section was found to be 21 pb. Preparations are being made for a renewed Darmstadt (90Ho.A) and a Berkeley (private communication) attempt to find element 110.

110; 03 TWG CONCLUSION. The Dubna experiment (870g99) describes interesting preliminary work but is insufficient to give confidence that element 110 has been produced.

Z=110: SUMMARY OF PAPERS, DATES AND CRITERIA

Z	Ref.	Date	Place
110	86Og99	860402	(Dubna)
	87Og99	870615	(Dubna)
	90Sc11	891125	(Darmstadt)

86Og99 232 Th(44 Ca,4n) 272 110 Ey SF 7 σ = \approx 10pb 232 Th(48 Ca,xn) $^{280-x}$ 110 Ey SF 64 C1 pb 209 Bi(59 Co,n) 267 110 Ey 64 C1 pb

Method. As 76Og02(Z=107). A shortlived (1 ms) background is attributed to ^{240mf} Am, based on the yield curve.

For Bi + Co, no detection method is mentioned.

87Og99 232 Th(44 Ca, 4 n) 272 110 as above 236 U(40 Ar, 4 n) 272 110 Ey As Cb SF 7 232 Th(40 Ar, 272 - 27 108 E_i SF 6 C2pb 235 U(40 Ar, 275 - 275 - 210 E_i SF 6

Method. Reaction recoils to thin collector foils, polycarbonate fission-track detectors. In 238 U(40 Ar,X) only 240mf Am and 242mf Am are seen. In 232 Th(40 Ar,x) no SF is seen (σ <2pb for T=10 ms).

90Sc11 233,235 U(40 Ar,xn) A 110 E_i SF σ <21pb Method. See 84Mu07. Element 110 not found.

ELEMENT Z=111

111; 01. The Dubna paper 86Og99.

In a bombardment of ²⁰⁹Bi with ⁶⁴Ni (method see 86Og99, Z=110), no indication was found for the formation of ²⁷²111 (upper limit for the cross section 4 pb.)

111; 02 TWG CONCLUSION. No data indicating formation of element 111 are available.

ELEMENT Z=112.

112; 01 The Harwell-Jerusalem-CERN paper 84Ma99.

In W targets bombarded for a long time with 24 GeV protons, these workers found an SF activity with a half-life of several weeks in the chemically purified Hg fraction. The energy of some SF fragments was low (about 65 MeV), and coincidences were found between two such fragments. They were interpreted as due to SF into four fragments (a process never observed before) of an isotope of element 112 which is expected to be a heavier homologue of Hg. Mass spectrometry yielded a mass

number in the region A=308-318. The proposed interpretation is that, in the very energetic collision, atoms such as ⁸⁸Sr are formed with energy around the Coulomb barrier which then produce secondary reactions such as ¹⁸⁴W(⁸⁸Sr, γ)²⁷²112. The higher observed mass value is explained as due to formation of molecules by combination with atoms of C, N, O or Cl.

TWG ASSESSMENT. Little is known about the production of heavy ions such as Sr in such bombardments; and even less about their energy distribution. Although it would be surprising if their yield were sufficient in the rather narrow energy band that would be operative for reactions such as that required for the production of element 112, this possibility can not be definitively dismissed and further work is to be hoped for.

112; 02 The Jerusalem-Darmstadt work 90Ma99.

The Jerusalem authors together with (among others) Darmstadt workers here describe experiments on the 88 Sr + 184 W reaction. Using 450 MeV Sr ions, a maximum cross section of 1 nb was found for the production of SF, using track detectors. Observed electromagnetic radiations of 163.7 and 29.1 keV agree with the energies to be expected for z=112 X-rays. Particle-particle coincidence measurements showed a coincidence between a 12.2 MeV α -ray and a 46 MeV fission fragment. The results are thought to indicate the production of a long-lived isomeric state of one of the isotopes $^{271}112$ or $^{272}112$, since the calculated excitation energy of the compound nucleus in this reaction is only 20 MeV so that radiative capture or emission of one neutron is probable.

TWG ASSESSMENT. The reported cross section may be surprisingly large but cannot be said to be impossible. Further work is needed.

112; 03 TWG CONCLUSION. Data reported so far are insufficient to indicate that a new element has been produced.

SUMMARY OF TWG CONCLUSIONS

Element 101

Element 101 was discovered by the Berkeley group - with certainty in 1958 (58Ph40) following strong indications in 1955 (55Gh9).

Element 102

The two 1966 (simultaneously published) Dubna results 66Do04 and, especially, 66Za04, both submitted in 1965, give conclusive evidence that element 102 had been produced.

Element 103

An important step on the way to discovery of element 103 was made in 1961 by the Berkeley group (61Gh03) although evidence fell short of full conviction, particularly in relation to the cross section curve. The Dubna papers 65Do10 and 68Fl01 taken together with 70Dr08 approached effective certainty; but it was not until the Berkeley paper of 1971 (71Es01), which confirmed earlier Berkeley and Dubna work, that it could be said that all reasonable doubt had been dispelled.

In the complicated situation presented by element 103, with several papers of varying degrees of completeness and conviction, none conclusive, and referring to several isotopes, it is impossible to say other than that full confidence was built up over a decade with credit attaching to work in both Berkeley and Dubna.

Element 104

The chemical experiments in Dubna (69Zv99 with 70Zv99) and the Berkeley experiments (69Gh01) were essentially contemporaneous and each show that element 104 had been produced. Credit should be shared.

Element 105

Independent work reported in 1970 from Berkeley (70Gh02) and from Dubna (71Dr01) was essentially contemporaneous and equally convincing. Credit for the discovery should be shared.

Element 106

Independent work reported in 1974 from Berkeley-Livermore (74Gh04) and from Dubna (74Og04) was essentially contemporaneous. The Dubna work is highly important for later developments but does not demonstrate the formation of a new element with adequate conviction, whereas that from Berkeley-Livermore does.

Element 107

The Darmstadt work 81Mu06 provides convincing evidence for the formation of element 107. The preceding Dubna work (76Og01 and 02) reports events with Z=105 which quite probably were due to the formation of element Z=107; yet, taken alone, this work does not give sufficient confidence that element Z=107 was in evidence.

Element 108

The formation of element 108 was established by simultaneous and independent work in Darmstadt (84Mu17) and Dubna (84Og02). The work in Darmstadt is more detailed and, of itself, carries conviction. In retrospect, the Dubna work taken together with cross section measurements in Darmstadt leaves little doubt that element 108 was produced and was inferred with confidence through identified descendants following a satisfactorily-understood decay path. The major credit should be accorded to Darmstadt.

Element 109

The Darmstadt work (82Mu15) gives confidence that element 109 has been observed.

Element 110

The Dubna experiment (870g99) describes interesting preliminary work but is insufficient to give confidence that element 110 has been produced.

Element 111

No data indicating formation of element 111 are available.

Element 112

Data reported so far are insufficient to indicate that a new element has been produced.

BIBLIOGRAPHY on TRANSFERMIUM NUCLIDES

Introduction. This Bibliography gives, in the first place, the papers mentioned in the Discovery Profiles. An attempt has been made, however, to make this bibliography on Transfermium elements reasonably complete. We even mention preprints of papers published in regular journals if provided with dates and with GSI-, JINR-, LBL- and UCRL-numbers, or other publications with such indications, not least since at least some of them are quoted in other literature. Sections of Annual Reports and a few "unorthodox" publications are provided when they contain information not available in better publications; reference keys for them contain a dot followed by a Capital letter instead of the last two digits for more "regular" publications.

Several Russian journals are regularly translated into English; if so, the page number in the translation is also given. In such cases, the titles and the spelling of the author names are taken from the translation, and therefore not necessarily consistent as between publications.

With few exceptions, each paper in this bibliography has been scrutinized by at least one member of the TWG for information that could be of interest for our work.

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