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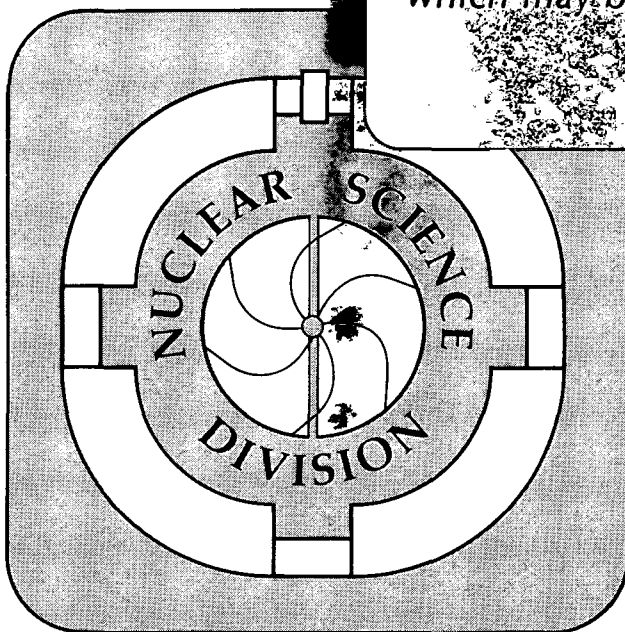
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E.K. Hyde, D.C. Hoffman, and O.L. Keller, Jr.

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A HISTORY AND ANALYSIS OF THE DISCOVERY
OF ELEMENTS 104 and 105*

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Abstract

Although the scientific literature now includes information on ten isotopes of element 104 and 6 isotopes of element 105 as well as on the chemical properties of these two elements, no official ruling has been made by any international body concerning their names nor the assignment of credit for discovery. A controversy has developed between research teams at the Joint Institute for Nuclear Research, an international laboratory at Dubna in the Soviet Union, and research teams at the Lawrence Berkeley Laboratory in the USA concerning the right to be recognized as the discoverers of elements 104 and 105. Both institutions have major facilities including nuclear particle accelerators which were used to synthesize isotopes of the new elements by bombardment of heavy element targets with heavy ions.

This paper reviews the experimental studies carried out at both laboratories during the 1960-1977 period and makes a critical assessment of the physical and chemical evidence relating to the synthesis of elements 104 and 105 and the claims based upon that evidence. Work done at several other institutions is cited when it is pertinent to the claims made by the research groups at Dubna or Berkeley. The authors then present their conclusions regarding the claims of the two laboratories to priority in the discovery of elements 104 and 105 and the right to name these elements.

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I. Introduction

It is the purpose of this paper to make a systematic review of publications concerning the first synthesis and identification of isotopes of elements 104 and 105. The majority of the reports on this subject originates with a group of investigators at the Joint Institute for Nuclear Studies in Dubna, USSR, led by G. N. Flerov and a group at the Lawrence Berkeley Laboratory in the USA led by A. Ghiorso. A controversy has developed between these groups as to which has the right to claim first clear identification (discovery) of element 104 and thus the right to name the element. The Soviet group has suggested the name Kurchatovium (Ku) while the American group has put forth the name Rutherfordium (Rf). Similarly both groups have reported research on the synthesis and identification of element 105. The Soviet team claims priority and has suggested the name Nielsbohrium, symbol, Ns. The American team also claims priority and has suggested the name Hahnium, symbol Ha. All unpublished reports and refereed publications which are important for the competing claims occurred in the period 1960-1977 for element 104 and in the period 1968-1977 for element 105. These reports and publications are extensively cited and reviewed in this article. A few later publications which are significant either because they reinforce the earlier claims or because they contribute data on the other isotopes of these two elements are also cited, but no attempt was made to discuss every publication from 1977 to the present time.

The conflicting claims and suggestions for the names of elements 104 and 105 caused great difficulty for the International Committee on Nomenclature of the International Union of Pure and Applied Chemistry (IUPAC). In 1974 IUPAC in collaboration with its companion agency, the International Union of Pure and Applied Physics, agreed to appoint an international ad-hoc committee of neutral experts including three persons each from the USA and the USSR and three from other countries (including the chairman) "to consider the claims of priority of discovery of elements 104 and 105 and to urge the laboratories at Berkeley (USA) and Dubna (USSR) to exchange representatives and in their presence to repeat the experiments regarding these elements." The three authors of the present article were chosen as the USA members of this ad-hoc committee. None of them had participated in the Berkeley experiments, but all three had had extensive research experience in the study of the nuclear and chemical properties of the transuranium elements. At the time of appointment one of them (D.C.H.) was on the staff of the Los Alamos Scientific Laboratory. This article had its origin in a 1975 request from the chairman of the committee that members of the committee "prepare a draft of the history of the work and respective views in the naming of these elements by the Russian and American parties." The committee carried on some informal activities to define the problem and to get better communication and agreement between the Berkeley and Dubna groups but it never completed its work nor issued a report. The present article, while it was stimulated by the committee, is entirely the responsibility of the three authors. It is their hope that this

review may prove useful to the scientific community interested in the synthetic elements and, in particular, to those who may have the opportunity to participate in an official way in the naming of these elements which are of great interest because they are the first elements beyond the actinide series in the Periodic System.

At the present time (1986) the identity and physical properties of many isotopes of elements 104 and 105 are clearly established and the chemical properties of both elements have been investigated sufficiently to determine beyond doubt that they are distinctly different from those of the actinide elements as would be expected for the first two elements beyond the actinide series. Furthermore, the identity and properties of several isotopes has since been verified by investigators in other laboratories; in particular in the Oak Ridge National Laboratory and the Gesellschaft für Schwerionenforschung (GSI) in the Federal Republic of Germany.

A principal reason that the controversy arose in the first place and has persisted so long is the extreme nuclear instability of these elements and the difficulty of preparing more than a few atoms at a time for physical or chemical measurements of their properties. Modern chemical and physical techniques are so powerful that unequivocal identification could easily be made if samples of a few thousand atoms could be prepared, even if the half life for radioactive decay were as short as a fraction of a second. Unfortunately many of the definitive chemical techniques with which radiochemists are familiar are inapplicable to samples of only a few atoms with very short half lives. Therefore, careful selection,

development, and calibration of methods was required for the study of the chemical properties of elements 104 and 105.

To aid those readers whose research experience lies outside of nuclear chemistry or physics, this report begins with several sections describing the trends in nuclear stability for elements with atomic numbers above 100, the types of nuclear disintegration significant to the investigations, the possible methods of synthesis of elements 104 and 105, the physical methods of detection and atomic number identification, and the problems involved in the chemical identification of a radioactive element on the basis of only a few atoms. A discussion of the crucial question of the criteria to be used in deciding whether a new element has been discovered follows.

A major part of this report is devoted to a chronology of the results and claims presented in all major publications on elements 104 and 105. These results and claims are evaluated in the final sections where the authors of this review present their conclusions on the validity of the claims and counter claims of the two groups of experimentalists.

II. Nuclear Stability Considerations

a) Coulomb forces

Unlimited extension of the periodic system of the elements by synthesis of nuclei with larger and larger numbers of protons is not possible because the total binding energy of a nucleus increases approximately linearly with the number of nucleons, whereas the repulsive Coulombic force between charge centers in the nucleus increases as the square of the atomic number. The liquid drop model of the nucleus predicts that the ultimate limit of stability occurs around atomic number 120. At this limit the model predicts that nuclei cannot be held together longer than the transit time of nucleons crossing nuclear dimensions, which is $\sim 10^{-22}$ seconds. It is known that the liquid drop predictions must be modified to accommodate the stabilizing effects of the quantum states of the last few nucleons. These effects extend the ultimate limit of stability by several units of Z and have important consequences which we shall discuss below, but do not affect the general explanation of why there are only about 100 stable or long lived elements.

b) Alpha decay

Heavy nuclei, even those with atomic number considerably below 120, are quite unstable toward radioactive decay by emission of alpha particles. Uranium, the heaviest of the natural elements available in macro quantities, decays by this process. Alpha decay is a quantum mechanical process first described by Condon and Gurney [1] in 1929. It is intrinsically a slow process and is very strongly influenced by

the size and shape of the potential barrier against alpha emission and by the energy and angular momentum of the alpha particle.

Both the potential barrier and the alpha-particle energy are sensitive functions of the nuclear structure. Collective properties of the nucleus are involved as well as single particle (and associated closed shell) properties. The wave functions of the last few nucleons in the potential well have the most influence on these properties. We can get some idea of the trends in alpha decay instability in the elements above uranium by examining empirical data for uranium and synthetic elements.

Figure 1 shows alpha particle decay energies as a function of mass for the heaviest elements. The steady increase in the decay energy as atomic number increases is clearly visible. The decay energies for these high- Z elements are large and correspond to high decay probabilities (short half lives) as is apparent in Fig. 2 which shows the general trends for even-even nuclei in the transuranium elements for alpha decay energies above 5.5 MeV. Half lives for nuclei with an odd number of neutrons or protons are longer by factors of up to 1000 or more. See Ref. [2].

Alpha decay can occur with resultant formation of the daughter nucleus in the ground state or in an excited state. When transitions do occur to excited states a corresponding complexity arises in the alpha particle spectrum. The spectrum of alpha transitions is an important characteristic of each heavy nucleus which decays by alpha particle emission. The alpha-energy spectrum of an unknown nuclide

and of its daughter product can play an important role in new element identification, as discussed later.

From Fig. 1 it is possible to predict the approximate energy of alpha particles emitted by an isotope of element 104 or 105. It is clear from the systematic trends that there is little probability of confusion with alpha particles emitted from most elements of lower Z . One might also think that it was unnecessary to be concerned with the group of nuclides around mass 210-215, which do have alpha energies of 9 MeV or greater, on the grounds that they would not be formed in a nuclear reaction in which the goal was to prepare a sample of element 104 or 105. However, owing to the presence of traces of lead or bismuth in the target materials, target backing foils, or beam collimators it is possible to make several of these nuclides which emit alpha particles with energies of 8 MeV or higher. The much higher nuclear reaction cross sections of formation of these products (because of negligible losses by fission decay in the formation process), compared to the formation of the desired much heavier element, compensates for the small percentage of lead or bismuth contamination in the heavy element target.

c) Spontaneous fission

Nuclei of uranium and heavier elements can fission spontaneously from their ground state into two massive fragments as a result of quantum barrier penetration or they can be induced to fission as a result of shape distortions caused by nuclear excitation associated with the capture of a neutron or with charged particle reactions. The

minimum excitation energy required for induced fission is about 5 MeV for most transuranium nuclei.

Spontaneous fission was discovered in natural uranium in 1940 by Petrzhak and Flerov [3] who showed that it is a very rare process in this element; the half life for spontaneous fission of ^{238}U is greater than 10^{16} years, compared to the half-life for alpha decay which is 4.5×10^9 years. Thus decay of ^{238}U by spontaneous fission occurs only about twice per million alpha decays.

In the period 1950-1964 the half-lives for spontaneous fission of about 30 nuclei were measured and many authors commented in the trends in the data or presented semi-empirical representations of the data as a basis for the prediction of properties of unmeasured heavier nuclei. A review as of 1964 is presented by Hyde [2]. Hoffman reviewed data on the heaviest nuclei in 1979 [2a].

Spontaneous fission becomes a more and more prominent decay mode for heavier elements as is clear from the data displayed in Fig. 3. Half-lives are shortest and most regular for even-even nuclei. If we examine the data in Fig. 3 for $Z < 104$ we can note the following trends. The half-life decreases roughly exponentially with increasing Z^2/A , but if attention is fixed on the isotopes of a given element, this is not true. If Z is fixed, the half-life increases with A until the neutron number 152 is reached, after which there is a drastic decrease. Some authors have associated this effect with the existence of a subshell at 152 in the independent particle description of nuclear structure. Half-lives for spontaneous fission of nuclei with

an odd number of neutrons or protons, or both, are significantly longer than those interpolated from neighboring even-even nuclei.

The significance of the spontaneous fission empirical systematics as it was known in the 1960s when the search for element 104 began, is that it was expected that even mass isotopes would have half-lives of the order of seconds or less for spontaneous fission and that odd mass isotopes would have half-lives significantly longer, but probably no greater than minutes. There were differing views of the significance of the 152 neutron subshell effect. The Berkeley group anticipated a precipitous drop off in half-life for isotopes of element 104 with more than 152 neutrons, while the Dubna group believed from their interpretation of their own data that the 152 neutron effect did not influence the properties of element 104 in that dramatic fashion.

Any theory that attempts to describe the phenomena of nuclear fission must have at its core some mathematical method for evaluating the energy of the nucleus in its ground state and in the states it must pass through as it deforms, necks down, and then ruptures into two or more fragments. In the classical work of Bohr and Wheeler [4], the liquid drop model (LDM) was chosen for estimation of the binding energy (mass) of the nucleus in its spherical ground state and in deformed shapes. The LDM, in analogy to liquids, describes the nucleus as a charged liquid drop characterized by a volume energy, a surface energy, and the Coulombic effects of a uniformly distributed protonic charge. In this picture the mass of the nucleus consists of the sum of the masses of the constituent neutrons and protons decreased by the nuclear binding energy and modified by the

electrostatic energy. Many authors have contributed to the development of the LDM and greatly improved it as a semi-empirical method for the estimation of masses of nuclei in their ground state and in states of deformation [5]. Descriptions of fission based on the liquid drop model are qualitatively correct but quite inaccurate in many details, such as the height of the fission barrier and the division of mass between the fragments.

After the introduction of the independent particle (shell) model of nuclear structure in the late nineteen forties and its elaboration in the nineteen fifties [6], with remarkable success in describing many nuclear data, there arose the hope that the shell model could replace the liquid drop model for all purposes, including the calculation of nuclear masses in equilibrium shapes as well as in deformed shapes by a mathematical procedure that involved no averaging of the properties of the nucleons but only a summing of the energies of the constituent particle states. However, so far, the mathematical challenges to such an undertaking have proven to be too formidable for it to form the basis of a complete theory of fission. However, an effective interim theory has been developed by combining the two approaches. The liquid drop model is used to define the major average features of the nuclear potential energy in deformation coordinates and the single particle model is used to make corrections to these features. The most widely used and most successful theory of this general kind is one introduced by the Soviet theorist V. M. Strutinsky [7].

Strutinsky's choices for the single particle states were those defined by the Nilsson model [8]. In the Nilsson description strong variations occur in the spacing of the states as they are filled in a deformed nuclear potential well. The levels are closely spaced just before completion of a "shell" of neutrons or protons and widely separated immediately after. When energies of states are summed, those nuclei near closed shells are more tightly bound than they would be if all states had the average energy assumed by the liquid drop approach. Thus, strong corrections to the liquid drop mass result for such nuclei. The Nilsson states change in a complex way with nuclear shape changes, which results in a complex pattern of shell corrections to the liquid drop mass predictions for the shapes involved on the path to fission. The Strutinsky method generates a binding energy map as a function of shape changes which is highly specific for each heavy nucleus. This map shows the height and location in deformation space of the fission barrier or barriers and explains many features of the fission process and shows why some nuclei are strongly stabilized against fission and others not. The Strutinsky method accounts in a natural way for the occurrence of mass asymmetry in the splitting of the nucleus during fission. The calculations also reveal that the shape of the barrier can be complex. One of the most dramatic successes of the theory was the discovery that some nuclei have a double fission barrier with a sufficiently deep minimum between the barriers to permit the existence of nuclei within this minimum. This explained the existence of fission isomers. The lifetimes of these isomers are in the nanosecond to millisecond range and are enormously

shorter than for spontaneous fission decay from the ground state of the same nucleus. More than 40 of these isomers have been identified among the nuclides of uranium, neptunium, plutonium, americium, curium, and berkelium [9-11]. For a particular thorough review see Bjornholm and Lynn [10]. The existence of these isomers is significant in connection with the identification of new transuranium elements because it makes it a necessary part of the identification to show that an observed spontaneously fissioning species is not an isomer of some lighter element.

Some calculations by Randrup et al. [12a] for elements 104 through 110, based on the Strutinsky method, are shown in the lower right hand section of Fig. 3. These calculations support the possibility of an abrupt change in the systematic trends of spontaneous fission half-lives beyond element 102, nobelium. A quite recent paper by Moller, Nix, and Swiatecki [16] discusses the application of the Strutinsky method and reports barrier properties and half-lives for many nuclei including 6 even isotopes of element 104.

As was mentioned above, the experimental data on isotopes with an odd number of neutrons and protons show that fission is reduced in probability compared to nearby even-even cases, but this hindrance is not a fixed quantity because it depends crucially on the specific nucleons involved. Although the recent theoretical calculations based on the Strutinsky method do not in general treat the odd proton or neutron cases, the extra hindrance associated with the fission of nuclides having an odd number of neutrons or protons has been recognized for some time. Johansson [13] treated this effect as early

as 1959. More recently, Randrup et al. [12b] have plotted the hindrances associated with such cases, as shown in Fig. 4. The hindrance is typically of the order of 10^5 , but has been found to be as low as 10 and as high as 10^{10} . They also calculated the extra hindrance associated with $n = 157$ nuclei (due to the $9/2 + [615]$ neutron orbital in the notation of the Nilsson model) and reproduced the observations for ^{257}Fm , ^{259}No , and $^{261}_{104}$ rather well. See Fig. 4.

It was thought at the time of the initial studies of elements 104 and 105 that the spontaneous fission half-lives of the synthesized isotopes would be quite short, probably in the range of milliseconds or less. Predictions were based only on interpretations of the trends of data on the light elements up through element 102 and were only good enough to serve as rough guides to experiment and identification. The powerful theoretical methods of today, particularly those based on the Strutinsky method, were stimulated by the experimental data accumulated during the new element searches at Dubna and Berkeley and, in particular, by the discovery at Dubna of a mysterious 14 ms spontaneously fissioning nuclide, later identified as an isomer of ^{242}Am . The later calculations based on the new theoretical approaches have contributed greatly to an understanding of many of the data on properties of fission isomers, fission barriers, mass division in the fission process, and fission half-lives.

We conclude that the observation of a nuclear species decaying by spontaneous fission with a short half life is a clear indication that the nuclide has an atomic number in the transuranium region, but

precise assignment of a new spontaneous fission activity to the isotopes of element 104 or 105 on the basis of half life alone is impossible.

d) Fission from excited nuclear states during production

In addition to the fission instability of the ground state, it is important also to consider the greatly enhanced fission probabilities of the excited states of isotopes of elements 104 and 105. The nuclear reactions used to make isotopes of these elements necessarily produce compound nuclei with excitation energies of 20 MeV or more. Loss of element 104 or 105 nuclei by fission during this de-excitation process decreases the yield of final nuclei by approximately ten orders of magnitude. This constitutes a serious obstacle to the preparation of samples of these elements in sufficient quantity for clarification of their properties. This loss by fission during de-excitation of reaction products is discussed more fully in Section III.

e) Electron-capture and beta decay

The heavy ion reactions used to produce isotopes of elements 104 and 105 result in nuclei which are neutron deficient and stable toward beta decay. However, decay by electron capture is energetically possible for the isotopes under consideration [14]. For the even-even isotopes, decay via this mode will not compete with spontaneous fission, which is expected to have a much shorter half life. For the odd-mass isotopes, electron-capture decay to the next lower Z element can compete with fission or alpha-particle emission depending on the particular energies and single-particle states involved. For example,

the decay energy for electron capture, Q_{ec} , is estimated [14] to be around 2 MeV for $^{261}_{104}$ and around 5 MeV for $^{255}_{104}$. These decay energies would give half lives of the order of hours to seconds, respectively, for allowed electron-capture decay. Similarly, electron-capture decay half lives of minutes can be estimated for $^{260}_{105}$, $^{261}_{105}$, and $^{262}_{105}$. These estimated half lives are considerably longer than those estimated for alpha decay in every one of the selected cases in element 104 and 105 with the possible exception of $^{262}_{105}$ where the electron capture and α -decay rates might be similar. Owing to the clear experimental advantages of studying α -decay or fission compared to electron capture none of the studies to be cited later in this report were designed to investigate electron capture.

Observation of decay by electron capture of only a few atoms of a heavy element isotope would be particularly difficult because of the necessity for measuring the few photons of the associated characteristic x rays or for detecting the growth of a known alpha-decaying daughter activity.

III. Methods of Synthesis

It is impossible to synthesize elements 104 and 105 by nuclear reactions induced by protons, deuterons or helium ions because of the lack of the necessary target materials of elements 102 or 103. It is also impossible to build up samples of these elements by intense neutron irradiation of lighter elements because the neutron capture and beta decay sequences, which work so well for production of many elements above uranium, are interrupted by very short beta-decay half lives and the extremely high susceptibility to fission of many of the isotopes in the sequence. A particularly disastrous nucleus in the neutron-capture-plus-beta-decay sequence is ^{258}Fm which decays by spontaneous fission with a 380 microsecond half life and abruptly terminates the build up to higher Z elements. This leaves "heavy ion" reactions as the only practical path for synthesis. Accelerated beams of such particles as ^{11}B , ^{12}C , ^{15}N , ^{18}O , and ^{22}Ne make it possible to jump by 5, 6, 7, 8, or 10 units of charge via compound nucleus reactions in which there is a complete fusion of projectile and target nuclei. Target isotopes of curium (element 96) are available in multimilligram quantities, isotopes of berkelium (element 97) in milligram quantities and einsteinium (element 99) can with difficulty be prepared as target materials in amounts of several micrograms. These are small quantities but with the advanced bombardment and detection techniques available in the Dubna, Berkeley, Oak Ridge, Darmstadt, and other laboratories a significant number of atoms of the new elements can be produced and studied.

Owing to the high charges of the target and projectile nuclei, there is an extremely high Coulomb repulsion between the two nuclei to the point of contact and resulting coalescence into a compound nucleus. Some typical values of Coulomb barriers are given in Table 1. However, modern particle accelerators such as the Berkeley SuperHILAC and 88-inch Cyclotron, the Darmstadt UNILAC, the Oak Ridge National Laboratory Cyclotron and the Dubna U-300 and U-400 Cyclotrons are readily able to accelerate heavy ion projectiles to these and even higher energies and thus to overcome these barriers.

There is a difficulty, however, in the use of such high energy projectiles. For bombarding energies in the barrier region, the major fraction of the reaction cross section goes into compound nucleus formation. After addition of the negative reaction Q -value to the kinetic energy of the projectile one finds that the compound nucleus is still excited to many tens of MeV. See Table 1. This excitation is greater than the binding energy of the least tightly bound neutron or proton and is also well above the fission barrier of 5 to 6 MeV; hence this excitation is dissipated by emission of one or more neutrons, protons, α -particles and γ -rays, or by nuclear fission. For transuranium element targets the emission of protons or other charged particles is inhibited by the Coulomb barrier, and γ -emission is a slow process. Hence, the most probable de-excitation processes are neutron emission and fission. These are independent processes, and the competition between them is expressed in a statistical description by the ratio of the neutron to fission widths, Γ_n/Γ_f . As long as neutron emission leaves the nucleus with enough residual energy to emit

Table 1. Reaction data for reactions induced by
complex nuclear projectiles

Target nucleus	Bombarding particle	Q-value for compound nucleus formation	Coulomb barrier energy (MeV)	Excitation energy of compound nucleus at barrier in MeV Q + barrier
^{242}Pu	^{22}Ne	-59.51	+ 99.88	40.37
^{243}Am	^{22}Ne		+100.85	
^{246}Cm	^{16}O	-44.58	+ 83.91	39.32
	^{18}O	-44.39	+ 82.95	38.56
^{249}Bk	^{14}N	-32.25	+ 74.90	42.65
	^{16}O		+ 84.54	
	^{18}O		+ 83.58	
^{249}Cf	^{12}C	-31.52	+ 65.77	34.25
	^{13}C	-29.64	+ 65.30	35.66
	^{18}O		+ 84.43	

Values taken from The Nuclear Properties of the Heavy Elements Vol. I, p. 343, E. K. Hyde, I. Perlman, and G. T. Seaborg, Prentice Hall, Englewood Cliffs, 1964. Q-values calculated with mass tables of Foreman and Seaborg, J. Inorg. Nucl. Chem. 7, 305 (1958).

another neutron, the competition of fission and emission has to be taken into account. Because of this possibility for fission at each stage of de-excitation of a compound nucleus, the overall total probability for fission rises with increasing bombarding energy until the cross section for the formation of evaporation residues becomes negligibly small.

Because the Γ_n/Γ_f ratio is an important nuclear parameter in understanding cross sections and other features of fission induced by charged particles, many authors have attempted to derive its value from experimental data or theoretical models. This is not an easy task because it is not clear how the Γ_n/Γ_f ratio varies as a function of nuclear charge, mass, excitation energy, and angular momentum. The general trend of Γ_n/Γ_f values for heavy nuclei is shown in Fig. 5. Sikkeland, Ghiorso, and Nurmia [15] derived Γ_n/Γ_f values from experimental data on carbon ion induced reactions and proposed the following empirical formula for estimation of values in cases where no data are available:

$$\begin{aligned} \log(\Gamma_n/\Gamma_f) &= -0.276Z + 5.46 + 0.140N, \text{ for } N < 153 \\ &= -0.276Z + 19.23 + 0.050N, \text{ for } N > 153. \end{aligned}$$

Figure 5 indicates by broken lines the application of the empirical formula to elements 98, 100, 102 and 104. However, the extrapolation of this empirical relationship beyond element 102 may be inappropriate because of possible changes in the height and shape of the fission barrier. Such changes would probably increase the probability for fission (reduce Γ_n/Γ_f). The projected very low Γ_n/Γ_f values for element 104 have the consequence that only 1 in 30 or fewer nuclei

survive at each stage where neutron emission and fission are in competition. Consequently, in a reaction like $^{242}\text{Pu}(^{22}\text{Ne},4n)^{260}104$, where 4 neutrons must be emitted by the compound nucleus, the number of surviving nuclei of element 104 is very low. The value which the Dubna group reports for the reaction cross section for the above case is 10^{-34} cm^2 whereas the geometric cross section for formation of the excited compound nucleus is calculated to be 10^{-24} cm^2 , or about 10 orders of magnitude higher. Computer codes have been written to estimate cross sections of specific reactions as a function of bombarding energies. These estimates are usually good to only a factor of 2 to 4, but are useful guides for planning experiments and assessing the difficulties to be encountered.

A typical example is the estimation of the product yields for the reaction of ^{249}Bk with ^{15}N .

To illustrate the difficulties caused by the low survival of the heavy nuclei in the nuclear reaction used to produce them, let us consider the rate of production of atoms in a typical case. The number of atoms produced, P , can be expressed as

$$P = T \times I \times t \times \sigma$$

Where T is the number of target nuclei per cm^2 , I is the number of projectiles per second, t is the duration of the bombardment in seconds and σ is the reaction cross section in cm^2 . For a beam current of 0.5 particle microamperes, a target thickness of one milligram per cm^2 , a bombardment time of 10 hours and a cross section of 10^{-33} cm^2 one can compute that ~ 300 atoms of element 104 will be formed in the reaction of ^{15}N with ^{249}Bk .

This low production rate, reflecting mainly the low reaction cross section (σ) due to massive loss by fission of the excited compound nuclei, is one of the major reasons it has been so difficult to get definitive and reproducible results in experiments designed for element 104 and 105. The situation is also complicated by the short half lives of the isotopes themselves, but this in itself would not be a critical disadvantage if 100 to 1000 times larger samples could be prepared. A further complication is that the radiations of the few atoms of the desired product must be sought in a large background of other radioactive products of lower Z formed by different reaction channels such as direct transfer reactions or by reactions of the beam particles with trace amounts of target contaminants such as lead.

IV. Physical Methods of Detection

Isotopes of elements 104 and 105 decay predominantly by α -particle emission and spontaneous fission. Hence, physical techniques specifically designed for detection of these decay modes are needed for identification of these elements. For many years the Dubna group favored the detection of spontaneous fission whereas the Lawrence Berkeley Laboratory group placed most of its effort on the detection of alpha-decay.

a) Measurement of spontaneous fission

The kinetic energy of the two fragments resulting from binary nuclear fission is of the order of 200 MeV and, hence, fission fragments are easily detected by ionization chambers, silicon semiconductor detectors, or other types of detectors. The large pulses are easily discriminated from those caused by other types of radiation, and it is possible to study the kinetic energy distribution of the fragments. If the fissioning sample is mounted on a thin backing, time-coincident measurements of the kinetic energy or velocity distributions of both fragments can be made. In fact, in favorable cases one can measure not only the kinetic energy, from which the ratio of the masses of the two fragments can be deduced, but other phenomena such as neutron or charged particle emission measured in coincidence with the fission fragments. There is a rich literature on this subject and much ingenuity had been applied to an exploitation of the detailed characteristics of fission for the spontaneously fissioning isotopes of elements 94 to 100 by the time the first

attempts were made to synthesize and identify isotopes of elements 104 and 105. (See reviews in Refs. [2,17]).

Unfortunately, for the study of these elements it was not possible to prepare samples of sufficient intensity in these early experiments to make use of the elaborate techniques which had been applied to elements of lower atomic number. The very short half lives of the known isotopes was another serious limitation. Therefore, measurement of the spontaneous fission properties of elements 104 or 105 in nearly all the early experiments was limited to a registration of the simple fact of the fission event without simultaneous measurement of its properties.

It was shown many years ago that the passage of nuclear fission fragments through many insulating solids such as mica, glass, natural minerals or plastic materials, causes narrow paths of intense disruption on an atomic scale which makes the damaged material along these paths subject to rapid and preferential attack by a properly chosen chemical reagent; e.g., hydrofluoric acid in the case of mica. By controlled etching this track can be made visible in examination by an ordinary optical microscope. This detection technique has been reviewed by Fleischer, Price and Walker [18].

The solid track detector has great advantages for the study of new elements. It is simple, inexpensive and has a low background. It records each event with close to 100 percent efficiency. It is highly specific for fission fragments. Gamma-rays, α -particles, protons, neutrons or scattered beam particles do not interfere if moderate attention is paid to etching conditions. It is insensitive to a high

radiation background and, hence, the detectors can be placed close to the nuclear reaction zone which is hostile to all other detectors.

The Dubna group devoted much study to this method and to the development of special phosphate glasses for this purpose.

The major obstacle to identification of a new element by measurement of spontaneous fission is that the fission process itself effectively destroys any direct information concerning the proton number of the fissioning nuclide. Although it is possible in principle to measure the Z and A of coincident primary fission fragments and hence reconstruct the Z and A of the parent, the problems of making such sophisticated measurements on the decay of a few atoms of very short-lived new elements appear difficult even at present. The solid track detector also suffers from the limitation that it provides no timing information and thus no coincidence experiments are possible.

b) Measurement of α -decay energy

Detection and measurement of the energy of α -particles is usually achieved with an ionization chamber or silicon semiconductor detector coupled to suitable electronic circuitry [19]. In the work being reviewed silicon detectors were used exclusively. Such detectors are capable of an energy resolution of 10-15 KeV (full width at half maximum) but under the less-than-ideal conditions of new element experiments the typical energy resolution was poorer, about 30 keV. The total energy of the α -particle is nearly 10 Mev, so that an energy resolution of better than 0.3 percent is achieved. When several dozen or several hundred α -particles are recorded by the silicon detector

system, an energy spectrum is derived which is of great value for the identification of the disintegrating species. In Section IIb above, we noted that each heavy element species has a characteristic energy for decay to the ground state of the daughter nucleus and that the pattern of decay to the ground and excited states of the daughter (as revealed by the spectrum of α -particle energies) also contains information about the nuclear structure. When the α -spectrum of a nucleus is known, it is possible to limit the possible assignments of atomic number and mass because of the systematic trends in the α -decay of the heavy elements, and the previously known information for nearly all possible candidates of lower atomic number.

The daughter product of the α decay of an isotope of element 104 is an isotope of element 102 (nobelium) which also has a high probability for rapid decay by α emission. If the experiments are conducted so as to permit the recording of the α -particle energy and the half life of this element 102 daughter, and if these daughter properties are already known from previously established results, then one has a very powerful tool for identification of the element 104 parent because of this genetic coupling. A similar situation obviously applies to the measurement of an element 103 daughter activity produced by α -decay from an element 105 parent.

How does one prepare samples of the daughter for α -particle measurements. The simplest method is to examine the total spectrum of α -particles emitted by the sample of the parent activity and follow the development of the spectrum with time. If the parent and daughter half lives lie in a suitable range it may be possible to observe the

changes in time (growth and decay in the rate of α emission) of specific daughter α -energy groups. These changes reflect the half life of the parent (growth portion) and the half life of the daughter (decay portion). Another method, called the method of "time correlation," is applicable when the half life of the daughter activity is relatively short and the total decay rate of the sample is small. In this technique, whenever an alpha particle is emitted with an energy close to the known or suspected energy of the parent, the time elapsed until the emission of the next alpha particle is recorded together with its energy. A semilogarithmic plot can be made of the time distribution of alpha particles with energies expected for the known daughter. If this time distribution forms a straight line consistent with the known half life of the daughter, a strong presumption of a genetic linkage is established. Various techniques of electronic recording, tagging, and sorting of the alpha data can be devised to implement this method. As we shall see below, the method of "time correlation" of alpha pulses was used by several groups studying elements 104 and 105.

It is also possible to separate the daughter activity physically. This can be accomplished by taking advantage of the recoil momentum given to the daughter nucleus when the high energy α -particle is ejected from the parent nucleus. For a parent nucleus of element 104 emitting an α -particle of about 10 Mev, the recoil energy of the daughter is about 150 keV. If the parent is situated in a nearly weightless source on the surface of the sample backing, the recoiling daughter nuclei can be collected and transported to a different location for measurement of their subsequent radioactive decay.

The Berkeley group has used various techniques based on the recoil phenomenon to separate and examine the subsequent decay of daughter products.

c) Measurement of characteristic X-rays

Ever since Moseley established the regularities in the energies of X-rays as a function of the atomic number of the emitting atom, characteristic X-ray energies have served as a reliable identification of the atomic nature of any material which is excited in such a way as to stimulate the emission of these X-rays. From the experimentally known energies of the K and L X-rays of the known heavy elements from uranium (92) to fermium (100) it is easy to extrapolate or calculate the values expected for elements 104 and 105. Because of the extremely small number of atoms of these elements available for study the observation of the X-rays is difficult. In addition to the intensity problem there is the further worry about false identification of γ radiation from nuclear decay for the K or L X-ray of the element of interest, because of the overlap of X-ray and γ -ray ranges (e.g., the $K_{\alpha 1}$ X-ray energy for element 104 is 132 Kev). If samples of sufficient intensity are available this source of error can be eliminated by analyzing the X-ray spectrum for its expected intensity pattern.

In Sections VII and X we cite important confirmatory experiments on specific isotopes of elements 104 and 105. A group at Oak Ridge National Laboratory developed a method for the unambiguous identification of the atomic number of radioactive species by measuring the K or L X-rays emitted by the daughter element after its

formation by emission of α -particles from the parent. In this case the X-rays are emitted during the de-excitation of a state of nuclear excitation of the daughter nucleus. This de-excitation occurs within a very short time ($\sim 10^{-12}$ seconds) which makes it possible to measure the time coincidence of the α -particles and the X-rays, with energy measurement of both types of radiation.

d) Measurement of nuclear reaction properties

When the synthesis of a new element is attempted with a heavy ion reaction and a new radioactive isotope is found which is a candidate for identification of this element, this isotope must have production characteristics which agree with the expected properties of the nuclear reaction used to make it. One of the important properties is the "excitation function," better described as the reaction yield as a function of the bombarding energy. For each reaction of the (Heavy Ion, x neutron) type, where x can vary, there is, for each value of x, a rise of the yield with energy to a peak value and then a decrease. The peak value, the shape of the excitation function, and the bombarding energy at which the peak occurs can be estimated from well known nuclear theory, where important parameters are evaluated from experimental data in reactions of similar type in neighboring target elements. The competition of fission with neutron emission, which we have mentioned in Section III, is a critical matter in estimating the yield at the peak of the excitation function. In many of the experimental studies in elements 104 and 105, which we shall discuss later, the authors cite information on the yield and excitation function in support of their claim to discovery.

In case there is some question whether a candidate radioactive isotope has been made by a (HI, xn) reaction rather than by a competing reaction, either through incomplete fusion of the target and projectile or through emission of one or more charged particles in the deexcitation of the compound nucleus, it becomes important to study alternative reactions in which the compound nucleus has lower nuclear charge than the element being sought. If the candidate radioactivity appears in such a reaction then it must be discounted as a candidate for the new element. If it is observed only in reactions which involve compound nuclei with Z equivalent to the element where discovery is being sought, then the case for its identification as the new element is strengthened.

Compound nuclei absorb the substantial quantity of forward momentum of the incident heavy-ion projectile. Because the compound nucleus is massive (Atomic mass ~ 260) the emission of a few neutrons or protons does not much affect this forward momentum. Soviet investigators, as we shall note later in this report, took advantage of this strong forward collimation of products of the (Heavy Ion, xn) type to discriminate against products of reactions in which only a few nucleons were transferred between the projectile and the target nucleus. They did this by placing metal plates with a differing pattern of holes downstream of the target and noting the change in the relative yield of products made by reactions of the two general types just mentioned.

V. Chemical Identification on a "One-atom-at-a-time" Basis

The problems of chemical identification of transactinide elements having half lives of the order of seconds or less and available only on a "one-atom-at-a-time" basis have been thoroughly discussed in a review article by Keller and Seaborg [20]. They point out that chemical experiments designed to identify such a new element must be not only fast, but capable of giving the same results for one atom at a time as would be obtained in a macroscopic chemical experiment. (Statistically speaking, one atom goes through many identical chemical reactions rather than a large number of atoms undergoing only one reaction.) Thus, methods such as ion exchange, solvent extraction or gas chromatography in which one atom is involved in many identical reactions (provided, as is usually the case, the reaction kinetics are favorable) can give chemically reliable results, while methods such as co-precipitation in which the atom may react only once can often give unreliable, non-reproducible, non-specific results. However, if these methods are to be used in the identification of new elements, it is necessary that the technique selected be capable of "high resolution"; i.e., be able to differentiate between elements or single the element out from all others. In some cases, group separations may constitute useful supportive evidence, but by themselves could not constitute identification. An example of a high resolution technique was the characterization of element 61 by well known ion-exchange separation techniques [21] which showed that it eluted between known rare earth elements.

In Section VII we review applications to element 104 of chemical identification on a one-atom-at-a-time basis by the Soviet and American research teams.

VI. Criteria for Discovery of New Elements

Owing to the difficulty of preparing samples of element 104, element 105 and higher Z elements containing more than a few atoms, and because of the short half lives for disintegration of these nuclei, there are extraordinary difficulties in studying the chemical properties of such elements. Thus many of the traditional methods for establishing the uniqueness (atomic number) of the element are not applicable and one must face the question of the acceptability of the chemical or physical properties cited in proof of the uniqueness of a new element. This question has been carefully considered and statements on the proper criteria for discovery of new elements have appeared in the literature. We quote from two [22,23].

G. N. Flerov and I. Zvara [22] in a 1971 article entitled "The Second Hundred Chemical Elements" set down the following three statements on criteria. (This is our translation of the Russian.)

"1. An 'Element' is above all a concept from the fields of chemistry and atomic physics. If only radioactive properties of isotopes are studied and nuclear - physical evidence is used to verify the conclusions, the work may be considered as discovery of the element only in the case when the conclusions both as to atomic number and as to mass number are not subject to revision in subsequent investigations. It is necessary to understand that as a rule the determination of radioactive properties does not have value for nuclear physics when the mass number is erroneously determined.

"2. If the atomic number is established by chemical methods or by means of atomic physics (roentgen spectroscopy and other techniques) then even without a nuclear-physical identification it follows that the work may be acknowledged as a discovery. In this case, the isotope's mass number may in general remain unknown.

"These criteria, it seems to us, are suitable not only to the contemporary age of element synthesis with the aid of heavy ions but also by use of a wider circle of methods of synthesis and search for new elements.

"We further believe that printed accounts dedicated to the discovery of a new element must contain a statement in the form of an "experimental section" if possible close to the summary statement of the evidence. In it there must be a uniquely determined atomic number of the element and the measured decay properties of the isotopes with the accuracy corresponding to the good contemporary level of experimental technique. Possible inaccuracies in the determination of the mass number must be discussed."

The second statement we wish to cite is from a publication of a group of American, German and French nuclear chemists [23]. We quote the essential paragraphs of this article and indicate by dots the omitted material.

.....

"The basic criterion, of course, must be the proof, by some means, that the atomic number (Z) of the new element is different from the atomic numbers of all previously known elements. This means, in general, that the atomic number should be established. It should not

be necessary to establish the mass number, except insofar as this evidence is directly related to the method used for establishing the atomic number.

"Chemical identification constitutes an ideal proof that an element with a new atomic number has been produced. Two important requirements should be met in this kind of experiment. First, the chemical procedure should be of a type that is valid for application to individual atoms; the use, for example, of ion exchange adsorption-elution or partition between solvents has been shown to meet this criterion in many situations, and such methods also provide safeguards against complicating surface adsorption and entrainment effects. Second, it must be possible to determine the presence or absence of the new element in the appropriate chemical fractions in an unequivocal manner. If the new element is observed through its decay by high-energy alpha-particle emission or spontaneous fission, or both, the chemical identification can be confined to separation from all known elements with atomic number greater than lead ($Z=82$).

.....

"Also satisfactory is the identification of characteristic X-rays in connection with the decay of the isotope of the new element. In actual practice this is likely to involve measurement of the half-life and precise, unique energies of the alpha particles of the new element in coincidence with the characteristic X-rays of the daughter nuclide. However, it might be possible to measure characteristic X-rays of the new element itself (primary product) if these can be associated with the subsequent immediate decay of this nuclide. Thus, such short-lived

X-rays, which may be emitted in the course of, or as an aftermath of, the production of the primary product, might be followed very shortly by emission of alpha particles or fission fragments which could be detected by delayed coincidence techniques. The characteristic X-rays must, of course, be distinguished from gamma rays of similar energies - perhaps by identification of the complex structure of the X-rays.

"The proof of a genetic decay relationship through an alpha-particle decay chain in which the isotope of the new element is identified by the observation of previously known decay products should be acceptable. This method depends on measurement of the half-life and precise, unique energies of the alpha particles of the new isotope, and measurement and identification of the half-life and decay properties of the daughter, whose identity, including atomic number, has been previously established. Time correlation between parent and daughter should be established. Use of a genetic relationship as evidence for a new element implies that the mass number of the new element isotope is experimentally determined by its relationship to a daughter nuclide of known mass number.

"Detection of a spontaneous fission activity and measurement of its half-life cannot per se establish that an element with a new atomic number has been produced. Even when additional information, such as fragment mass and kinetic energy distributions, can be obtained, the atomic number assignment for new elements cannot be made on this basis alone since the systematics and theoretical predictions cannot be extrapolated with the necessary certainty into new regions.

Similarly, the use of the predicted half-lives for spontaneous fission decay and alpha decay and of predicted alpha-decay energies cannot yet be considered sufficiently reliable for establishment of the atomic number of a new element.

"The present understanding of production yields, excitation functions, angular distributions, and so forth is not sufficient to allow measurements to establish with certainty that a nuclide with a new atomic number has been produced, although such data may be useful as supportive evidence. It is particularly difficult to establish and interpret the difference between heavy-ion-induced compound nucleus reactions in which only neutrons are emitted (and consequently the atomic number of the product nucleus is the sum of the atomic numbers of the target and projectile) and those nuclear reactions (compound nucleus or otherwise) in which charged particles (such as protons and alpha particles) are also emitted so that the atomic number of the heavy product is less than the sum of projectile and target atomic numbers. An unambiguous differentiation between these reaction mechanisms would be necessary for the proof of atomic number by the use of such techniques. Information from cross bombardments can be useful, but again, interpretation of the results is subject to the same uncertainties concerning production yields and reaction mechanisms.

.....

"As a concluding thought, we suggest that composite nuclear systems that live less than about 10^{-14} second (the generally accepted upper limit for a compound nucleus lifetime) shall not be

considered as new elements. Nuclear molecular systems (those in which extranuclear electrons encompass two closely adjacent nuclei), which can be identified by their corresponding transitory X-rays, would also not qualify as new elements.

"The criteria described here should be necessary and sufficient for proof of the discovery of a new chemical element. We believe that any claim to such a discovery should be published in a refereed journal with sufficient data to enable the reader to judge whether the evidence is consistent with such criteria. We further believe that even when these criteria are met, the name for a new element should not be proposed by the discoverers until the initial discovery is confirmed."

This concludes the citation from Ref. [23] on the criteria for discovery of new elements.

VII. Chronological Sequence of Publication on Element 104

In this section we cite all the essential publications relative to the identification of isotopes of element 104 and summarize the results and claims made in each of these studies. Many of the reported results and claims are in disagreement but we reserve our principal comments on these discrepancies for Section VIII of our paper. We have neglected a few conference reports or other publications which present some of the results in summary form, but we believe we have included all the important original sources from the Joint Institute for Nuclear Research, Dubna, and from the Lawrence Berkeley Laboratory as well as some significant confirmatory studies from the Oak Ridge National Laboratory, the Lawrence Livermore National Laboratory, and the Gesellschaft für Schwerionenforschung (GSI) in Germany.

a) Soviet publications, 1960-1969

In the period 1960-1964 Polikanov and his co-workers [9] at Dubna initiated the study of the reaction of ^{22}Ne with ^{238}U and other heavy nuclei and discovered that in these reactions there was a large probability of formation of a 14-msec spontaneous fission activity of unknown nature. The half life was considered anomalously short compared to spontaneous fission half lives known at that time. This activity was later identified as an isomer of ^{242}Am . The isomerism turned out to be a remarkable new type related to a double minimum in the potential energy curve as the americium nucleus changes in shape

from spherical to more elongated spheroidal shapes on the path to nuclear fission. The Soviet theoretician Strutinsky [7] showed how corrections to the liquid drop model based on the wave functions of individual nucleons (shell effects) could produce such a double minimum and a metastable isomer of elongated shape in the second minimum.

This unexpected discovery of the 14-ms spontaneous fission activity triggered a world-wide search for more examples of such isomerism and an intense development of theory [9-11]. It is not an exaggeration to say that a sub-branch of nuclear physics was created as a result. The prestigious Bonner Award of the American Physical Society was given to S. Polikanov and V. Strutinsky for their part in this development in nuclear fission. Numerous other examples of spontaneous fission isomers have been found among the nuclei of U, Pu, Am, Cm and Bk.

The spontaneous fission isomers are important for the discussion of element 104 and 105 because the 14-ms ^{242m}Am is a prominent by-product in some of the reactions employed to synthesize these elements and because it is necessary before any spontaneous fission activity is assigned to a new element, to exclude an alternative assignment as an isomeric form of some element of lower atomic number.

In 1964 G. N. Flerov and his collaborators [24] published a description of their study of the reaction of ^{22}Ne with ^{242}Pu in which a new 300 ms spontaneous fission activity was observed. They assigned it to $^{260}104$, produced by the reaction $^{22}\text{Ne} + ^{242}\text{Pu} \rightarrow ^{260}104 + 4n$. A possible alternate assignment was $^{259}104$.

The experiments were carried out with the aid of a special probe, represented schematically in Fig. 6, which was placed inside the 300 cm heavy ion cyclotron. The products of the nuclear reactions were ejected from the ^{242}Pu target by the momentum imparted to them by the beam and were caught in a nickel conveyer belt which transported them past a series of fission detectors made of phosphate glass. When reaction products on the moving belt underwent spontaneous fission one of the fission fragments would enter one of the glass detectors, leaving a latent image (in the form of damage to the molecular structure) which could be developed and etched by later chemical treatment in the laboratory. The etched tracks could then be observed under a microscope and the distribution of tracks along the series of glass detectors could be determined. From this distribution and the known speed of the conveyer belt the half life could be deduced. In a series of experiments the speed of the belt could be varied over a wide range. The energy of the ^{22}Ne ions could also be varied.

When the incident particle energy was in the range 113–115 MeV, which is at the expected optimum value for the $^{242}\text{Pu}(^{22}\text{Ne},4n)^{260}_{104}$ reaction, the formation of a spontaneously fissioning isotope with a half life of about 0.3 ± 0.1 sec and a cross section of about $2 \times 10^{-34} \text{ cm}^2$ was observed. The data are shown in Fig. 7. This decay curve contains 58 events.

Figure 8 shows the dependence of the yield of the 0.3 sec activity on the beam energy. This curve has a shape and maximum consistent with assignment to the compound nucleus reaction written above.

The authors considered other possible assignments of the observed activity but dismissed them as improbable. Check experiments were done with other combinations of target and projectiles to eliminate several of these alternate possibilities. They concluded that the shape of the excitation function, the cross section value at the maximum, and the absence of the effect with other ions and targets provided sufficient grounds to propose the formation and observation of an isotope of element 104 with a mass number 260.

In a paper published in 1968, Donets and Schchegolev [25] described attempts to detect a branching decay by α -emission in the 0.3 sec activity assigned to $^{260}_{104}$. These experiments made use of recoil separation techniques followed by chemical separation of a fermium fraction from the appropriate recoil collection foil and a search for the α radiations of 22.7 hour $^{252}_{\text{Fm}}$ which is the granddaughter of $^{260}_{104}$ after a chain of two α -disintegrations. No atoms of $^{252}_{\text{Fm}}$ were found and an effective upper limit of $2 \times 10^{-34} \text{ cm}^2$ was set for the reaction $^{242}_{\text{Pu}}(^{22}_{\text{Ne}}, 4n)^{260}_{104}$, assuming that this nuclide decays predominantly by alpha emission. The authors expressed the view that this cross section was much lower than was to be expected from their previous studies of formation cross sections of compound nucleus reactions for atomic numbers above 100. They had predicted a value approximately ten times higher.

The team of physicists (Flerov et.al. [24]) that developed the evidence for the 0.3 second spontaneous fission activity realized that their claim to discovery would be strengthened by observation of the chemical properties that would characterize their 0.3 second isotope

as element 104. They expressed this thought in the following sentence in their initial report [24]. "However, taking into consideration difficulties in the syntheses of transfermium elements the authors believe it is quite desirable to conduct chemical experiments for additional identification as well as for the study of chemical properties of element 104." The challenging task of making these chemical measurements was accepted by Dr. Ivo Zvara and his coworkers at the same Joint Institute for Nuclear Studies, Dubna. They knew from the work of their physicist colleagues that they could expect to detect one atom of the 0.3 second isotope per 5 to 6 hours of beam time. Since no known chemical technique could be applied to such exceptional conditions, Dr. Zvara and his group took on the formidable task of developing what amounted to a whole new approach to the study of the chemistry of a radioactive element.

In 1966 this group issued its first report [26] on the chemical properties of the 0.3 second isotope, but owing to the lack of many details in that preliminary report, to understand these experiments we need to examine several more detailed papers published a few years later.

The development of the requisite techniques was made on the assumption that the actinide hypothesis was correct in predicting that element 104 would be eka-hafnium. Under this assumption, element 104 should have a volatile chloride whereas the chlorides of the heavy actinides should be non-volatile. Gas chromatography was chosen as the technique to be developed for these studies, since the needed speed and requisite degree of separation could be attainable by this

approach. Background experiments with hafnium, zirconium, and certain lanthanide elements gave encouraging results [27,28,29].

The apparatus and general experimental approach were described in detail in 1969 publications [30,31,32]. (These papers were submitted to Radiokhimiya in the fall of 1968. Experiments reported in summary form in 1966 presumably employed a similar approach) [26]. In addition to assuming that element 104 is eka - hafnium, the Dubna chemical experiments conducted between 1966 and 1969 were specifically designed to study an isotope of that element having a 0.3 second half life.

A diagram of the apparatus is given in Fig. 9. Sections I, II, and part of III were inside the cyclotron so that the target could be irradiated with the higher flux of the internal beam of ^{22}Ne . The target composition was: ^{242}Pu - 97%; ^{240}Pu - 1.5%; ^{238}Pu - 1.5% in the initial experiments and somewhat different in later ones, but the ^{242}Pu content was always above 92 . In some experiments a samarium target was irradiated along with the Pu target to produce simultaneously $^{170,171}\text{Hf}$. Recoil atoms from the target, caught in nitrogen carrier gas (section I), were swept into a chlorination chamber (section II) containing a mixture of niobium pentachloride and zirconium tetrachloride vapor. Section III was a 4 meter long chromatographic tube. The walls of the tube, which serve as the exchange surface, were stainless steel, Teflon, or glass in different experiments. Section IV is an inert filter used to trap large particles that are known to be produced, for example, as aerosols in the interaction of NbCl_5 vapor with oxygen present in the nitrogen

carrier gas. The filter was usually made up from crushed material of the chromatographic tube walls. Atoms that successfully passed the chromatographic tube and inert filter entered section V, which was 600 mm long. This section contained mica fission track detectors. Their purpose was to record the fissions of $^{260}_{104}$ nuclei that occurred during the fraction of a second they were in this chamber. It was known that traces of actinide nuclei could deposit out on the detectors as well. These nuclei could be recorded too, if they underwent spontaneous or neutron induced fission before the tracks in the mica were counted. The final section (VII) was a condensate trap for the chloride vapors that passed through all the previous sections. All sections were maintained at a uniform temperature of either 250 or 300 (± 15) $^{\circ}$ C for these experiments except in one case in which the apparatus was maintained at room temperature.

On the basis of ancillary experiments, the authors showed that only one event could arise in 300 days in the detectors from neutron induced fission of uranium impurities known to be present in the mica. They also showed that about one event might arise in the detectors from plutonium transferred from the target in a 2 to 3 day run.

The fissions counted in the mica detectors in the first series of experiments reported by Zvara and coworkers in 1969 are given in Table 2. One to five fission events were recorded for each bombarding energy that was expected to produce some $^{260}_{104}$ atoms. No $^{260}_{104}$ was seen in the case where the bombarding energy precluded production. The beam time for each experiment was 2 to 3 days. A

Table 2

Series	No. experiments	Material of walls gas channel	Carrier (vapor pressure in mm Hg)	Temperature of tube and filter (°C)	Energy of ²² Ne ions (MeV)	Integral flux of particles 10 ⁻¹⁷	Distance be- tween de- tectors (mm)	Measure- ment time (sec)	Number of fission tracts recorded
I	6	Stainless steel	NbCl ₅ (0.15)	250	114	20.9	10	1.2	2
II	3	The same	NbCl ₅ (0.10)		114	4.5	10	1.2	1
III	4	Teflon	+ZrCl ₄ (0.05)	300	114	17.7	6	0.72	1
IV	1	Glass	NbCl ₄ (0.16) +ZrCl ₄ (0.05)		114	2.9	The same	The same	3
	1				114	4.5	The same	The same	5
	1				128	3.2	The same	The same	0
	1			114	2.5	The same	The same	2	
									Total 14

total of 14 fission events was recorded in this series of experiments [31].

In addition to the fission events recorded there were other results in these experiments.

(a) There was complete transfer of the hafnium isotopes into the carrier condensate trap. If element 104 behaved like Hf, its atoms would also be transferred completely to the trap unless they decayed in flight while in the mica detector chamber. The passage time by the detectors was set at 0.7 seconds to optimize the detection efficiency for a 0.3 second half life) [30,31,32]. With NbCl_5 in the vapor phase, hafnium was determined to transfer [31] from the target to the detectors in 1 sec at 250°C . With ZrCl_4 in the vapor phase, the transfer occurred at 250°C in 0.5 sec and at 300°C in 0.2 sec. Since these transfer times were near the half life of 0.3 sec (which was thought at the time to be correct for $^{260}_{104}$), the apparatus and experimental parameters were deemed suitable for demonstrating the chemical similarity of element 104 to Hf.

The physicochemical conditions in the chromatographic channels were set by the Dubna researchers with considerable care on the basis of calibrations using HfCl_4 . The conditions to be used for the 104 experiments were chosen so that the retention time (t_R) of $(104)\text{Cl}_4$ in the channel would be equal to the time for the carrier gas to go through [32]. The design of the experiment was such that the half life of the 104 isotope entered into the calculation [33] of t_R . From the value of t_R so derived, the heat of adsorption, Q_A , of $(104)\text{Cl}_4$ could be calculated using chromatographic

theory [31,33]. Obtaining the value of Q_A was important [32] because, as the authors state: "This makes it possible to identify Z of unknown nuclides by comparing the heats of adsorption or directly comparing the values of t_R of compounds of the same type from different elements." Thus the quantitative evidence for identifying element 104 was to make its chloride and to compare its heat of adsorption to that of HfCl_4 .

(b) The half life of $^{260}_{104}$ could be determined in these chemical experiments if it had a value in the range of the transit time (0.7 seconds) across the mica detectors since the nuclei would decay while they were in the chamber before reaching the condensate trap. The Dubna group found that the distribution of the fission events along the mica detectors was compatible with a half life of 0.3 seconds (Fig. 10). This result supported the earlier finding of Flerov et al. [24] The authors [31] point out that the fission track distribution "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 sec and 3.7 sec."

(c) About 2% of the total amount of actinide isotopes produced were found on the mica detectors or in the condensate trap. The presence of these isotopes was recognized by the Dubna researchers as a source of error in their experiments. They made many ancillary studies designed to show that the fission tracks they were attributing to $^{260}_{104}$ did not arise from these other elements.

A second series of chemical experiments [33] was submitted to J. Inorg. Nucl. Chem. in the late summer of 1969 and published in 1970. The apparatus and the physical and chemical parameters and principles involved were functionally the same as in Ref. 31, except that: (1) no Hf calibration was done, (2) Pu and Am targets were irradiated with the external rather than the internal beam of the cyclotron, (3) thionylchloride was added to the carrier gas in addition to $ZrCl_4$ and $NbCl_5$, and (4) the transfer time of the isotopes through the chamber containing the mica detectors was set at around 1.5 seconds. Twenty 40 - 50 hour experiments were carried out. Experimental conditions for these experiments and the number of fission tracks recorded in the mica detectors are given in Table 3. A total of 67 events were recorded. Most of these were with the Pu target and at a ^{22}Ne beam energy near 115 Mev, the energy most favorable for the production of $^{260}_{104}$. At Ne beam energies that would be unfavorable for the production of the 0.3 sec $^{260}_{104}$ reported by Flerov and coworkers,²⁴ only a few events were seen. The same was true in experiments with a different combination of projectile and target nuclei (O + Am) in which the activity should not have been produced.

In this second series of 1969 chemical experiments, [33] in essential correspondence with the previous series, [31] the distribution of fission tracks in the mica detector column (Fig. 11) was stated to "imply a half-life of somewhat less than one second." A value of 0.5 seconds was accepted by them as the correct value of the half life from their complete set of data. The authors parenthetically inserted a brief statement that the true half-life might be

Table 3 [From Ref. 33]

Series	Target	Bombarding particle energy (MeV)	Capillary column material	Temp. (°C)	No. of expts	Particle current $\times 10^{-17}$	N_0	N	$N/N_0(\%)$
1*	^{242}Pu	^{22}Ne	glass	300	3	8.6	172	13(2)	8
		(113-117)		350	1	4.8	96	6(2)	6
2	^{242}Pu	^{22}Ne	glass	300	8	12.1	242	28(10)	12
		(113-113)		350	2	6.5	130	16(4)	12
3	^{242}Pu	^{22}Ne	KCl	300	1	3.4	68	0	0
		(116-118)							
	^{242}Pu	^{22}Ne	glass	300	2	6.0	12	2(1)	16
		(129)							
4	^{242}Pu	^{18}O	glass	300	1	2.6	0	1(1)	
		(83)							
	^{243}Am	^{18}O	glass	300	2	6.0	0	1	Total 67(20)
		(92-94)							

*Chloride carriers (mm Hg): $\text{NbCl}_5(0.15) + \text{ZrCl}_3(0.05)$. N_0 = calculated number of atoms of 0.3 second isotope produced. N = number of atoms observed (the number in parentheses refers to events in which both fission fragments were detected).

longer (than 0.5 seconds) since the 104 chloride molecules could be passing through the column "more slowly than the carrier gas." No experimental evidence was given to support this assertion. The experimental evidence obtained previously on the HfCl_4 transfer time calibrations [30,31,32] was not discussed or replaced. This isolated statement on the possible longer half life was not used in any way in the paper. Indeed, after making it, the authors calculated the heat of adsorption of $(104)\text{Cl}_4$ from their data assuming a half life of 0.5 seconds [33]. The value of 24 kcal/mol that they obtained in this manner was shown to correlate with HfCl_4 and with analogous elements (Fig. 12). It is important to note that the heat of adsorption correlation was the only quantitative evidence presented by the Dubna chemistry group to identify element 104 as eka-hafnium in their 1966 - 1969 experiments.

At the Welch Foundation Conference in the fall of 1969, Zvara [34] announced that Dr. Druin and coworkers had determined a new value of 0.1 sec for the half life of the isotope assigned by Dubna as $^{260}_{104}$. (Later the accepted half life was to become 0.02 seconds.) It became evident at this conference that the newly determined short half life of $^{260}_{104}$ presented serious difficulties for the interpretation of the Dubna 104 chemistry experiments because the technique employed was too slow to observe such an isotope. This problem caused Zvara [34] to revise somewhat the Dubna chemical group's half life determination of $^{260}_{104}$ as given previously [31,33] by stating: "The detector assembly is unfortunately not so good for measuring the half life as it might appear at first sight. There are some peculiar effects due

to the aerodynamics involved and due to the fact that the assembly itself necessarily acts as a chromatographic column. So it probably gives only a rough estimate of the half life, perhaps its lower limit and in no case can the decay curve shown in Fig. 11 be separated into the possible components. The conclusion that can be drawn is that we deal with the kurchatovium activity (or activities) with the half-life of about 1 second or more and also with tails of the decay curve of the 0.1 second kurchatovium activity."

A paper published by Zvara and coworkers [35] in 1969 on the chemistry of elements 102 and 103 is also of interest in this discussion because of additional important information it gives on the Dubna 104 chemistry experiments. In these experiments the apparatus for the internal beam of the cyclotron was used. This is the same apparatus used in Refs. [26] and [31] and is functionally similar to that used in Ref. [33]. The results for element 102 are given in Fig. 13. As can be seen, Pu behaves very much like Hf in the chromatographic column. Concerning this similarity between Pu and Hf chemistry the authors only state: "The peculiar behaviour of plutonium indicates higher volatility of its chloride." Plutonium clearly represents a serious potential source of error in the Dubna experiments. Ancillary experiments designed to show that Pu was not the source of the fission tracks attributed to elements 104 were carried out by the Dubna group. The appearance of the heavy actinides in the mica detector region for the 102 experiment [35] as shown in Fig. 13, was also noted in the 104 publications [31,33].

b) Berkeley publications, 1969-1970

In 1969, Ghiorso, Nurmia and Harris [36] attempted in Berkeley to confirm the existence of a 0.3 second isotope, of $^{260}_{104}$ or $^{259}_{104}$. They reported that they were unable to find evidence for this activity in experiments in which it should have been made by the reactions $^{246}_{\text{Cm}}(^{18}_{\text{O}},4n)^{260}_{104}$, and $^{248}_{\text{Cm}}(^{16}_{\text{O}},4n)^{260}_{104}$.

In 1969 this same group with 2 additional investigators (K. Eskola and P. Eskola) claimed [37] the first positive identification of two alpha-particle emitting isotopes of element 104.

It was claimed that in the bombardment of a 60 microgram target of monoisotopic $^{249}_{\text{Cf}}$ with $^{12}_{\text{C}}$ ions from the Berkeley heavy ion linear accelerator (HILAC) a new alpha-particle activity was produced by the $(^{12}_{\text{C}},4n)$ reaction.

The transmuted atoms recoiling from the target were swept by helium gas from the target region operating at a pressure of about 500 Torr through a 0.4 mm-dia. orifice into vacuum to be deposited onto the periphery of a 45-cm-diam. wheel. The digitally controlled wheel was rotated every three seconds to place these atoms sequentially into positions adjacent to four Si-Au surface-barrier crystal detectors so that alpha-particle spectra could be obtained during four sequential three second periods after collection of the sample of reaction products. The energy and half-life data were stored in a computer. The beam current was monitored by a water-cooled Faraday cup. Because of the very high beam densities it was also necessary to edge-cool the windows, the degrader foils, and the target by water-cooled frames.

The ^{12}C bombardments of the ^{249}Cf target produced a new alpha-particle activity with a complex energy spectrum (Fig. 14) and a half-life of 4.5 ± 1.0 sec. The spectra were submitted to a computer analysis with the result that four prominent alpha-particle groups with energies 8.70, 8.78, 8.95, and 9.00 MeV and relative intensities 0.15, 0.2, 0.3, and 0.35, respectively, were assigned to the same activity. For alpha-energy calibration, the 6.043-MeV peak of $^{209,210}\text{Rn}$ and 7.43-MeV peak of ^{250}Fm were used and extrapolation was made by use of a pulse generator. The absolute accuracy of the energy values was estimated to be 0.02 MeV. The amount of activity was substantial (~5 counts per microampere hour of beam) and it was possible to measure its excitation function with good accuracy (Fig. 15). The function, with a maximum cross section of approximately 10^{-32} cm^2 , corresponds to that expected for the (^{12}C , 4n) reaction to produce $^{257}_{104}$.

To make certain of the mass and atomic number of this activity, a series of experiments was carried out to identify the ^{253}No daughter, an alpha-particle emitter with an energy of 8.01 MeV and a half-life of 105 sec. Samples of $^{257}_{104}$ were collected as before on the periphery of a wheel which was then moved every three seconds to position these samples sequentially in front of 4 semiconductor detectors. After repeating this process for 200 seconds, the four semiconductor detectors were moved by an automatic mechanism to positions opposite four similar detectors. This was done in order to measure the activity from the nobelium (element 102) daughter atoms which had been transferred by alpha-particle recoil to the detectors

when they were opposite the element 104 deposit on the wheel. During the next 200-sec period it was then possible to measure the half-life and energy of the daughter atoms free from the interference produced by those atoms produced directly during the bombardment. This cycle was repeated automatically with the beam being turned off during the daughter-measuring intervals. A composite spectrum of the mother and daughter activities is shown in Fig. 16. The activity from the 55-sec, 8.10 MeV ^{254}No which was produced directly in good yield during the bombardment served to monitor the amount of nobelium that reached the surfaces of the crystals by a transfer mechanism of unknown origin; such transferred nobelium atoms were observed only in the first detector. The amount of ^{253}No daughter activity, as well as its energy and half-life, correspond quite well with the genetic sequence proposed.

It was further reported by Ghiorso et al. [37] that in bombardments of the same ^{249}Cf target with ^{13}C ions, a new alpha emitter was produced which had a half life of about 3 seconds and two prominent alpha particle groups at 8.77 and 8.86 MeV. Daughter atoms from this alpha emitter were collected by the recoil separation technique and the alpha decay properties of the daughter were determined with silicon semiconductor detectors. A known isotope of element 102, nobelium, was identified; namely, 185 second ^{255}No , which emits 8.11 MeV alpha particles. The genetic relationship of the 3 second activity to the ^{255}No daughter fixes its identity as $^{259}_{104}$. The same 3 second $^{259}_{104}$ activity was produced by the $(^{16}\text{O},5n)$ reaction in bombardments of ^{248}Cm .

In a third set of experiments, mica detectors were used as recorders of fragments from the spontaneous fission of the products of the reaction of ^{12}C and ^{13}C ions with ^{249}Cf targets. In these experiments evidence was found for an isotope decaying by spontaneous fission with a half life of 13 ± 2 msec. The excitation functions and yields were consistent with its assignment to $^{258}_{104}$ but the authors stated that they could not be certain of this attribution because spontaneous fission fragments by themselves did not provide a Z or A signature.

Additional details on these experiments leading to the synthesis of $^{257}_{104}$ and $^{259}_{104}$, as well as confirmatory experiments done with more elaborate experimental apparatus, are published in a paper delivered by A. Ghiorso at the 1969 Conference of the Robert A. Welch Foundation in Houston, Texas [38]. In the spectra published in these references, several dozen events are recorded for each isotope of element 104.

An additional alpha-emitting isotope of element 104 with a half life of 65 ± 10 seconds and a mass number of 261 was synthesized [39] by the $^{248}\text{Cm}(^{18}\text{O}, 5n)^{261}_{104}$ reaction by the Berkeley group in 1969 and reported in a 1970 publication. A preliminary account of this work was presented by Ghiorso at the 1969 Houston Conference [38]. This isotope emits 8.28 MeV alpha particles. The daughter isotope, separated from its parent by the alpha-recoil process, was shown to have the properties of 26-second ^{257}No which emits 8.22, 8.27, and 8.32-MeV alpha particles.

The comparatively long half life of the isotope, $^{261}_{104}$, made it an attractive candidate for use in radiochemical experiments to determine some chemical properties of the element. In a paper [40] published in 1970 (and in a preliminary report [38] in 1969), the Berkeley group described experiments in which samples of $^{261}_{104}$, synthesized by the $^{248}_{98}\text{Cm}(^{18}_0\text{O},5\text{n})$ reaction, were separated from the target by a physical technique, then dissolved in an aqueous solution from which they were adsorbed on a short column of the cation exchange resin, Dowex 50, and then eluted from the resin with a solution of 0.1 molar α -hydroxyisobutyric acid adjusted to a pH of 4.0. Summing up the results of several hundred individual experiments, it was observed that 17 atoms of the 65-second isotope were rapidly eluted from the column as was to be expected if element 104 were chemically similar to its homolog, hafnium, in its elution properties from a cation exchange resin in the presence of a complexing agent. Under the conditions of the experiments it was known that the actinide elements would not have been eluted from the column. These experiments indicated that the ion exchange column behavior of the α -activity, which had been identified as $^{261}_{104}$ by Ghiorso and co-workers [39] on the basis of physical evidence, is consistent with that of hafnium and zirconium, but differs entirely from the chemical behavior of the trivalent and divalent actinide elements.

c) Cross criticisms, exchange of views, 1969

At the Robert A. Welch Foundation Conference on the Transuranium Elements, held in Houston, Texas, November 17-19, 1969, Dr. Albert Ghiorso, leader of the Lawrence Berkeley Laboratory group, summarized all the work done to that time by his group and suggested the name rutherfordium (symbol Rf) for element 104. He gave his reasons for disbelief in the correctness of assignment to $^{260}_{104}$ of the 300 msec, spontaneously fissioning isotope, reported by the Dubna group, but stated that if later work established the correctness of the earlier work by the Dubna group that he would withdraw his suggestion for the name and accept the name, Kurchatovium, put forth by the Russian scientists. This paper which was published in the Proceedings of that Conference [38] contains many experimental details concerning the Lawrence Berkeley Laboratory work on the isotopes of mass 257, 259 and 261.

In a series of papers [41-43] in the period 1969-1971, the Dubna group raised questions about the validity of the Berkeley work on the alpha-emitting activities assigned to $^{257}_{104}$ and $^{259}_{104}$. These questions were based on the appearance in the published spectra of alpha particle groups resulting from the presence of lead impurities in the californium target and on some peculiarities of the measured half lives of some of these alpha groups. These criticisms were answered in a 1971 paper published in Nature [44] by members of the Berkeley group. These papers are not discussed further in this document because there is no evidence from later publications or

contacts between the groups that the essential correctness of the original Berkeley work on the isotopes, $^{257}_{104}$ and $^{259}_{104}$, is now challenged by the Dubna group.

d) Later Soviet publications, 1969-1975

In 1969 Druin, Lobanov, and Kharitonov [45] developed a new experimental technique of general importance for determining whether some observed nuclear reaction product is produced by a mechanism which involves the formation of a compound nucleus followed by the evaporation of only a few nucleons or whether it is produced in some other reaction in which only a limited number of nucleons are transferred between the projectile and the target nucleus. In the first case the reaction product would recoil from the target within a cone with a half angle of only a few degrees from the direction of the beam. In the second case the products of non-compound nucleus interactions would recoil with considerably larger angles.

The technique chosen by Druin et al. [45] to exploit this fundamental difference was to place a foil behind the target in which a grid of evenly spaced holes had been drilled. The yield of specific products was measured in a series of experiments in which the hole diameter was kept constant but the collimator thickness was increased to increase the degree of collimation. They tested the influence of these changes in recoil collimation on a series of products from known nuclear reactions and were able to verify the predicted effects and to derive calibration curves for distinguishing compound nucleus reaction

products from products of reactions in which many nucleons or nuclear clusters were evaporated or otherwise ejected.

This collimation technique was applied by Oganessian and co-workers [46] in 1969 to the spontaneously fissioning activities produced in the reaction ^{242}Pu and ^{22}Ne . They found that the activity which the Dubna group had assigned to $^{260}_{104}$ did behave in a manner consistent with the strong forward peaking expected of a compound nucleus reaction. The 14 msec ^{242}Am by-product activity, on the contrary, had a much broader angular distribution. This was taken by them as confirming evidence for the correctness of the assignment of the first activity to element 104.

In the course of this study, performed with the apparatus shown in figure 17, Oganessian and co-workers determined that the half-life of $^{260}_{104}$ was much shorter than had been originally reported in 1964. In the new study the half life was revised downward from 300 msec to 100 ± 50 msec. See figure 18.

This paper is a major restatement of the evidence in favor of the assignment of the 100 msec spontaneous fission isotope to $^{260}_{104}$.

Another major restatement of the evidence for the 100 ± 50 msec spontaneous fission activity and its assignment to $^{260}_{104}$ was made by Dr. G. N. Flerov in an address [47] delivered at the International Conference on Heavy Ion Physics held at Dubna, February 11-17, 1971. The new work mentioned in Ref. 46 was reviewed again. A measurement of the yield of 100-msec activity for three beam energies in the 108-120 MeV range showed the yield curve had a maximum at 114 MeV with a half width of ~ 10 MeV. These values are in good agreement with

theoretical yield curves for the ($^{22}\text{Ne},4n$) reaction. The reaction cross section at the yield maximum was $7 \times 10^{-34} \text{ cm}^2$, compared to the $2 \times 10^{-34} \text{ cm}^2$ value reported in the earlier paper [24] when the half life was measured as 0.3 second.

In the ^{22}Ne beam energy interval, 114–140 MeV, evidence was found for two more spontaneously fissioning isotopes with half lives of 4.5 ± 1.5 seconds and much greater than 5 seconds. The dependence of the formation cross section of the 4.5 second isotope on beam energy had the characteristics expected for the ($^{22}\text{Ne},5n$) reaction so that the authors suggested that this 4.5-second activity was associated with the spontaneous fission decay of $^{259}_{104}$. Hence the authors believed that they had observed the spontaneous fission branching of the isotope whose alpha decay had been observed previously by the Berkeley group. Further evidence for this was found by measuring the effect of grid collimators placed in back of the plutonium target on the yield of the 4.5 second isotope. The effect was of the nature expected for a compound nucleus reaction.

This paper also emphasized a systematics of the spontaneous fission half lives for isotopes of element 104 which was at variance with that used by the Berkeley group. The main difference was that the Berkeley group believed there should be a pronounced maximum in the half lives of even Z-even N isotopes at $N = 152$, whereas the Dubna group proposed that there is a departure from the behavior observed for lighter actinide elements and that there is only a gentle dependence of spontaneous fission half lives on neutron number for element 104 and above. See Fig. 3.

In 1971 Zvara and his group published a paper [48] that described a new experiment on element 104 chemistry. This experiment resembled the earlier one in being based on gas chromatography of volatile chloride compounds but differed from it in some crucial ways. The motivation for the restudy as explained in the introductory section of the paper came from recent measurements of Flerov et al. [47] which resulted in the conclusion that two isotopes of element 104 had been observed with half lives of 0.1 and 4.5 seconds for $^{260}_{104}$ and $^{259}_{104}$, respectively. Zvara et al. also stated that the chemical experiments carried out by them earlier could not have permitted them to observe an activity with a half life as short as 0.1 second. They stated that they now believed that the half life of roughly 0.5 second which they had observed in the earlier experiment was caused by some absorption effect and that they really had been observing the chemical properties of the 4.5 second $^{259}_{104}$. A central purpose of the new experiment was to prove that their previous experiments had in fact involved the study of the 4.5 second $^{259}_{104}$ and that the conclusions regarding its chemical properties were still valid.

The techniques employed in the new experiments were greatly superior to those used earlier for ascertaining chemical properties.

The same type of target and chlorination assembly was used as in the earlier experiments (see Fig. 9), although TiCl_4 was substituted for NbCl_5 and ZrCl_4 as the exchange agent. The most important change was that the new design used a linear temperature gradient instead of a constant temperature in Section III of the chromatographic tube. The mica fission track detectors were placed in

this section. Section V of Fig. 9, where the 0.3 sec $^{260}_{104}$ nuclei had supposedly been detected in the earlier experiments (as they supposedly decay during their rapid passage), was eliminated. Indeed, for this new set of experiments the Dubna group perfected and employed a new thermochromatographic chemical separation technique. In the thermochromatographic technique for each element with a volatile chloride there was a temperature at which it would condense and form a peak. A peak identified with hafnium is shown in Fig. 19. Peaks were also formed for Sc and heavy transuranium elements (^{256}Fm) at the hot end of the column (Section I and the beginning of Section II). In Section III, where the steep temperature gradient is maintained, fission tracks were recorded at the same position of deposition as HfCl_4 . The large increase in chemical resolution of this experiment relative to the previous ones gives confidence that these fission tracks arise from a volatile chloride species of element 104 that behaves similarly to HfCl_4 . The authors state that the resolution was sufficient to separate HfCl_4 from plutonium chlorides, for example. Furthermore, most of the length of Section II is free of activity so that a large buffer zone is maintained. This gives confidence that all of the relatively nonvolatile chloride molecules were stopped by the beginning of Section II. This was not true in the previous experiments [31,33] where a significant percentage of the actinides was found in the detector region and in the condensate trap.

The thermochromatographic technique developed by Zvara and co-workers for this paper [48] for 104 chemistry has been applied by them in many more studies of great importance in the actinide region [49].

The Berkeley group raised several objections [50] to the Dubna chemistry results in the references just cited. One of these was that the spontaneous fission branching of $^{259}_{104}$ was set at less than 20 percent by Berkeley measurements and might be less than one percent.

Druin et al. [51] (1973) measured spontaneous fission activity in the reaction of $^{246}_{\text{Cm}}$ with 100 MeV $^{18}_0$ ions which was expected to produce $^{259}_{104}$ by the ($^{18}_0,5n$) reaction. They saw 31 spontaneous fission events with a half life of 3.8 ± 0.8 seconds, in agreement with the previously reported half life for $^{259}_{104}$. The measured cross section was $4 \times 10^{-34} \text{ cm}^2$. The expected total cross section for α -decay (as well as spontaneous fission) was estimated from various data to be about $6 \times 10^{-33} \text{ cm}^2$. From these numbers a branching ratio (spontaneous fission/ α) of about 7 percent was estimated. This result was supported by a later measurement carried out at Oak Ridge National Laboratory by Bemis and coworkers, [52] who found the SF branching to be 0.06 ± 0.037 . Their value of the half life was 3.0 ± 1.3 sec.

In 1974 Oganessian et al. [53] introduced a new reaction concept, later named cold fusion, in which a target such as $^{208}_{\text{Pb}}$ is bombarded with a very heavy projectile like $^{50}_{\text{Ti}}$ in order to form a compound nucleus of element 104. $^{208}_{\text{Pb}}$ is a doubly-magic nucleus. During compound nucleus formation the breaking of this shell structure of the target nucleus absorbs energy, which results in a colder compound nucleus. This cooling causes the emission of less nucleons in the de-excitation of the compound nucleus, hence in fewer chances for losses by fission. By use of this approach Oganessian et al [53] reported the discovery of two spontaneous fission activities with half lives of 4 sec and 6 ms

in the reaction of ^{50}Ti with ^{207}Pb and ^{208}Pb . They attributed these activities to $^{255}_{104}$ and $^{256}_{104}$, respectively, produced in the reactions $^{207}\text{Pb}(^{50}\text{Ti}, 2n)^{255}_{104}$ and $^{208}\text{Pb}(^{50}\text{Ti}, 2n)^{256}_{104}$. These spontaneous fission half lives are in agreement with the Dubna view of the systematics of spontaneous fission of isotopes of 104. The data on element 104 show an abrupt change from the trends observed in lighter even-Z elements. This revised systematics is, however, not in conflict with a 20 or 100 ms $^{260}_{104}$. These results on the activities assigned to $^{255}_{104}$ and $^{256}_{104}$ were reproduced later at the UNILAC accelerator in Darmstadt by Münzenberg and coworkers [54,55] who found an 8.726 MeV alpha particle associated with the decay of $^{255}_{104}$ and a half life of 1.4 ± 0.6 seconds. They observed an 8 msec half life for the spontaneous fission decay of $^{256}_{104}$.

e) ORNL confirmation of $^{257}_{104}$

The Berkeley group's claim [37] of the identification of a 4.5 second $^{257}_{104}$ in the reaction of $^{249}_{\text{Cf}}$ with $^{12}_{\text{C}}$ was verified [56] by an independent group of American scientists at the Oak Ridge National Laboratory in 1973. The experiment consisted of the identification of the characteristic $K\alpha_1$, $K\alpha_2$, $K\beta_1$, and $K\beta_2$ X-rays of element 102 in coincidence with the α particles of the 4.5 sec activity attributed to $^{257}_{104}$. (See Figs. 20 and 21). The element 102 X-rays come from the element 102 daughter product from the α decay of $^{257}_{104}$. This experiment is regarded as extremely strong verification because of the uniqueness of the energies of the X-rays and the high resolution of the instrumentation for discriminating against X-rays of neighboring elements.

f) Attempts to get agreement on the half life of $^{260}_{104}$

In 1975 the Dubna group [57] restudied the synthesis of element 104 isotopes by the reaction of a $^{246}_{\text{Cm}}$ target (0.8 mg cm⁻² surface density) with an $^{18}_{\text{O}}$ beam (3 microamperes). These workers observed the formation of a spontaneously fissioning isotope with a half life of 80 ± 20 msec which they assigned to the isotope $^{260}_{104}$. This half life was considered more accurate than previous values reported by the Dubna group. The excitation function and the angular distribution of the 80 msec activity as it recoiled from the target were judged to be consistent with the assignment to $^{260}_{104}$. The

maximum formation cross section was determined to be 1.5×10^{-33} cm^2 . The authors emphasized that these results contradicted the negative results reported by Ghiorso [37] for the same reaction. An important feature of the experimental apparatus used in this new set of experiments was a recoil collector tape 10–20 microns thick and 200–800 meters in length. Use of this tape markedly reduced the background of the by-product ^{256}Fm which undergoes spontaneous fission with a 2.6 hour half life.

Stimulated by the results described in Ref. 57, Nitschke and his coworkers [58] restudied the reaction $^{249}\text{Bk}(^{15}\text{N},4n)^{260}\text{104}$ in a major attempt to prepare and observe the 80 ± 20 ms spontaneously fissioning isotope, assigned as $^{260}\text{104}$ by the Dubna group. A team of scientists was assembled from four American laboratories and an observer from Dubna was invited to be present during the experiments. The best semi-empirical estimate of the cross section for the isotope $^{260}\text{104}$ was 25 nanobarns ($25 \times 10^{-33} \text{ cm}^2$) at a ^{15}N energy of 82 MeV (the energy for a maximum in the production cross-section for the $(^{15}\text{N},4n)$ reaction). This cross section is the highest of those estimated for any of the competing reactions except $^{11}\text{B} + ^{253}\text{Es}$ and hence the $^{249}\text{Bk} + ^{15}\text{N}$ combination was regarded as the most favorable for the production of $^{260}\text{104}$.

The experiment was designed so that atoms of element 104 recoiling out of the $85 \mu\text{g cm}^{-2} \text{ } ^{249}\text{Bk}$ target would impinge on the edge of a large metallic drum which rotated at high speed. To reduce the level of ^{256}Fm contamination the drum was also moved laterally. The collected deposit of element 104 and other reaction products rotated

past a series of mica sheets which served as detectors of fission fragments. Any atom disintegrating by spontaneous fission would send one of the fission fragments into the mica detector. After many hours of running, the beam was turned off and the mica sheets were removed for chemical treatment to develop the tracks caused by the fission fragments. The tracks could then be observed on a low power (100x) microscope. The position of the track in the mica relative to the location of the interaction point of the ^{15}N beam and target, together with the speed of the collector drum rotation, gave a measure of the time of decay of the collected product. Measurements were made at beam energies of 78, 82, 86, 88 and 100 MeV with integrated beam intensities of 9-47 microampere hours at each energy. Even at the most favorable energy of 82 MeV no trace of the 80 ms activity was found; a conservative upper limit of 0.7 nanobarns was set. There was, however, evidence for an unknown spontaneously fissioning species with a half life of 20 msec (and an apparent cross section with a peak value of 19 nanobarns at a beam energy of 82 MeV) above a small background from 2.6-hour ^{256}Fm .

A. Ghiorso presented a paper [59] on this experiment at the 3rd International Conference on Nuclei Far From Stability, Cargese Corsica, France, May 26, 1976. The paper which appears in the proceedings of that conference contains a lengthy introduction reviewing the history of the discovery of element 104 and of the controversy surrounding it as seen from his perspective.

V. A. Druin analyzed [60] the data presented by A. Ghiorso at the Corsica Conference [59] and criticized the interpretation given by Ghiorso. These criticisms were repeated in a longer paper [61] discussed below.

A team of Dubna scientists [61] in April and in September 1976 studied the products of interaction of ^{249}Bk with ^{15}N ions and detected an activity with a half life of $76 \pm 8\text{ms}$ which they attributed to ^{260}Ku produced with a cross section of 8 nanobarns. An observer from Berkeley was present during one set of these experiments.

In these experiments the ^{249}Bk target material was chemically purified from its ^{249}Cf daughter just before use. Its surface density was 180 micrograms per cm^2 . Products of the nuclear reaction with the ^{15}N beam recoiled out of the target and were caught on a moving nickel tape, whose motion was controlled by a "tape recorder" system. A long (350-600 meter) tape was used so that background activity of 2.6 hour ^{256}Fm , which is produced in a by-product nuclear reaction, would not accumulate on the tape and interfere with the measurement of the 80 ms activity. As in previous experiments a series of fission fragment track detectors was placed along the path of the moving nickel tape.

Experiments were performed at beam energies of 75, 78, 82, 89, 90 and 93 MeV. The expected maximum for the $^{249}\text{Bk}(^{15}\text{N},4n)^{260}\text{104}$ reaction was at a beam energy of 82 MeV.

At 82 MeV the observed spontaneous fission tracks were distributed in the track detectors in a pattern corresponding to a 2-component decay curve. See Fig. 22. Above a long lived background (presumably

2.7 hour ^{256}Fm) there appeared a 76 ± 8 ms component with a cross section of 8 ± 2 nanobarns. At lower and higher ^{15}N beam energies the cross section decreased. The 3 point excitation function had the appearance of the predicted yield curve for the $4n$ reaction. This report on the observation of a 76 ms second component produced with an 8 nanobarn cross section is in clear contradiction to the results reported in Berkeley [58], where an upper limit of 0.7 nanobarns was set for the production of an activity with this half life. However, both experiments agreed on the cross section for production of ^{256}Md .

The Russian authors sharply criticized the experimental techniques used by their American competitors. Their major criticism was that the drum collector technique used in Berkeley provided a moving surface for collection of reaction products with an effective length of 0.8 meters in some experiments and 30 meters, at most. In their view, even 30 meters was inadequate to spread out the atoms of the long-lived 2.6 hr ^{256}Fm and hence that the spontaneous fission background built up to a level which interfered with the observation of the 80 ms $^{260}_{104}$. They further claimed that the 20 ms activity observed in Berkeley could be a mixture of 80 ms and 13 ms components of $^{260}_{104}$ and $^{242\text{m}}\text{Am}$, respectively. They forced a computer fit to the Berkeley data assuming the existence of these two activities and obtained a cross section of 1.0 ± 0.5 nanobarns for the 80 ms component. This is to be compared with the upper limit of 0.7 nanobarns set by the American authors [58] for their data and the 8 nanobarns set by the Dubna authors for theirs [61].

The Dubna experimentalists also ran an experiment on the $^{249}\text{Bk} + ^{15}\text{N}$ reaction at the most favored beam energy using a recoil collector device with an 8 meter catcher belt to simulate the conditions of the reported experiments in Berkeley. They obtained data which looked very similar to those reported by Ghiorso in Ref. 58. The short lived component had a half live of 14.6 ± 3.6 ms and a cross section of 10 ± 6 nanobarns. This is in reasonable agreement with the Berkeley results. Druin et al. [61] however interpreted the 14.6 ms component as an artifact of a mixture of 13ms $^{242\text{m}}\text{Am}$ and 80 ms $^{260}\text{104}$; they reanalyzed the data under this assumption and obtained cross sections $\sigma(13\text{ms}) = 10 \pm 2$ nanobarns and $\sigma(80 \text{ ms}) = 1.3 \pm 0.6$ nanobarns respectively. They conclude that this shows experimentally how background conditions can lead to the loss of the 80 ms activity and to the misinterpretation of experimental data. They did not explain why they obtained a cross section of 8 ± 2 nanobarns in one set of data and 1.3 ± 0.6 in another.

J. M. Nitschke working at the Lawrence Berkeley Laboratory, constructed a 2000-meter long metal tape system with mica detectors located near the surface of the tape to detect the spontaneous fission of the recoiling atoms collected on the tape. This arrangement made it possible to study half lives ranging from milliseconds to hours with cross sections in the nanobarn region in the presence of background activities having microbarn cross sections. Using this system, Somerville et al. [62] could find no evidence for the 0.3-s, 0.1-s, or 79 ms activities variously reported by the Dubna Group in the reactions $^{249}\text{Bk} + ^{15}\text{N}$ and $^{248}\text{Cm} + ^{16}\text{O}$ and were able to

set cross section limits lower by factors of 24 and 2, respectively, than those reported by the Dubna workers. However, they were able to observe very clearly the 20 ms activity, as shown in figure 23. In experiments at other bombarding energies they did find three other spontaneous fission activities with half lives of 19 ms, ~ 55 ms, and ~ 1.3 sec in the bombardment of ^{248}Cm with ^{18}O ions. The excitation function for production of the ~ 1.3-sec activity was too broad to correspond to production of a compound nucleus followed by neutron evaporation. Subsequent measurements of the fission properties of this activity by Hoffman et al. [63] using an 80-position horizontal wheel system showed that it has a very high total kinetic energy, a narrowly symmetric mass distribution, and a half life of 1.6 ± 0.3 s. These very unusual properties are consistent with those reported [64] for 1.5-s ^{259}Fm produced via the ^{257}Fm (t,p) reaction. ^{259}Fm is the only known isotope with this half life which exhibits predominantly symmetric fission with very high total kinetic energy. The production of ^{259}Fm in the $^{248}\text{Cm} + ^{18}\text{O}$ bombardments may be viewed as an effective transfer of ^{11}Be to the ^{248}Cm . More experiments will be needed to determine the reaction mechanism. The production of ^{259}Fm in the $^{248}\text{Cm} + ^{18}\text{O}$ reaction with a peak cross section of ~15 nb indicates that neutron-rich products can be produced in heavy-ion bombardments and that extreme caution must be exercised in making isotope assignments based on the assumption of compound nucleus formation. Somerville et al. [62] suggest that the 20-ms spontaneous fission activity, which is also produced in $^{15}\text{N} + ^{249}\text{Bk}$ and $^{16}\text{O} + ^{248}\text{Cm}$ bombardments, is

$^{260}_{104}$ and that the 55-ms activity may be $^{262}_{104}$. Definitive assignments of these activities probably must await measurement of the mass via a mass separator. If the nuclide $^{264}_{106}$ were formed in some future study and found to be an alpha emitter some properties of its daughter $^{260}_{104}$ could be measured indirectly.

The publication of Somerville et al. [62] presents a very thorough study of the spontaneous fission properties of all isotopes of element 104 as well as a review of previous work.

A 1985 publication from the Dubna Laboratory is quite significant in resolving the dispute on the $^{260}_{104}$ half life. Ter-Akopyan and coworkers [65] made one more restudy of the $^{249}_{\text{Bk}} (^{15}_{\text{N}}, 4n) ^{260}_{104}$ reaction. They measured a half life of 28 ± 6 msec, which is much closer to the 21 ± 1 msec half life reported by the LBL group [62] than to the 76 ± 8 msec value obtained earlier in Dubna [61]. Based on an idea of Ghiorso's [59] they discussed a possible explanation for the distortion of the pattern of spontaneous fission tracks observed in the earlier Dubna experiments. The essential difference between the method used in the latest set of experiments compared to the earlier ones (Druin et al, [61] for example) consisted in the presence of helium gas in the space between the target and the collector of the nuclear recoils in the earlier experiments. It is possible that a small fraction of the longer lived mendelevium and fermium nuclei were stopped in the gas and transported along the direction of motion of the nickel collector tape and were deposited on the surface of the fission track detectors. The subsequent decay of these nuclei caused a distortion of the distribution of fission fragment tracks.

g) LLNL measurement of total kinetic energy of
fission fragments of $^{260}_{104}$

In April 1983 E. K. Hulet of the Lawrence Livermore National Laboratory reported [69a] on some on-line measurements of the coincident fragment energies in the fission of a 20 millisecond activity produced in the bombardment of $^{249}_{\text{Bk}}$ with $^{15}_{\text{N}}$ ions. Hulet concluded that the measured properties, together with the previous publications from Berkeley and Dubna, gave high probability to the conclusion that the fission characteristics of $^{260}_{104}$ had been observed. The mass distribution was symmetric, with a width of 35 mass units (full width at half maximum). The corresponding total kinetic energy distribution had a most probable value of 199 MeV. The only other nuclei among the transuranium elements which are known to have a symmetric mass distribution in the fission process are $^{258}_{\text{Fm}}$, $^{259}_{\text{Fm}}$, $^{258}_{\text{Md}}$, $^{259}_{\text{Md}}$, and $^{258}_{\text{No}}$.

VIII. Conclusions Concerning Claims and Counterclaims on Discovery of Element 104

In this paper we have presented the results of all published papers relating to the first synthesis and identification of the isotopes of element 104 and have summarized the claims and counterclaims on priority of discovery. In this section we now present our conclusions with respect to the correctness of these claims.

The claim to priority of the Soviet scientists from the Joint Institute for Nuclear Studies depends critically on the physical evidence presented in 1964 (primarily Ref. [24]) and a few other reports between then and 1969 when the American group published its claims. The Soviet claim also depends on the chemical evidence presented in the 1966 - 1969 year period [26,31,33].

The physical evidence involved the observation of a 0.3 second spontaneously fissioning nuclide produced by the bombardment of ^{242}Pu with ^{22}Ne at a beam energy where the $(^{22}\text{Ne},4n)$ reaction was expected to be favored in a compound nucleus reaction. In experiments in which the ^{22}Ne energy was varied from 108 to 124 MeV the yield of the product showed a rise and fall consistent with the proposed reaction. The formation cross section was measured to be about $2 \times 10^{-34} \text{ cm}^2$ which is only a factor of 10 lower than

expected from a semi-empirical theoretical calculation of the expected value. The new activity was not observed in several other reactions such as $^{238}\text{U} + ^{22}\text{Ne}$, $^{242}\text{Pu} + ^{22}\text{Ne}$, and $^{242}\text{Pu} + ^{18}\text{O}$. In these reactions $^{260}_{104}$ could not be formed but most other candidates for assignment of the 0.3 second activity could have been produced. Possible confusion with a spontaneously fissioning nuclide of much lower atomic number was ruled out by later experiments with a collimating grid placed behind the target [45].

This evidence makes it plausible to assign the 0.3-second activity to element 104 but it is indirect and inconclusive. The authors themselves commented on the desirability of the conduct of chemical experiments for additional identification. The basic problem, which is quite fundamental, is that the mere observation of spontaneous fission in itself is insufficient to identify the atomic or mass number of the fissioning nuclide. This fundamental difficulty is not present when the alpha decay properties are measured.

We turn now to a discussion of the chemical evidence. Owing to the insufficient physical evidence for discovery of element 104, the gas chromatographic technique was developed and applied in the 1966 - 1969 period with the goal of identifying element 104 conclusively. It was necessary that the resolution of the experiment be sufficient to establish with a high degree of certainty that element 104 could be clearly distinguished from all other elements or

at least from all elements which could decay by spontaneous fission. As the following analysis will show, the Dubna chemistry experiments conducted between 1966 and 1969, which we described in Section VII, fell far short of reaching this goal. The basic fallacy was that the experimental apparatus was specifically designed to study an isotope with a 0.3 sec half-life. The Dubna researchers then misinterpreted the very small number of events they recorded by thinking they were consistent with the assumed half life.

Since the 1966 paper [26] is simply a summary with no details about the experiments, we will first focus on the second paper [31] entitled: "Experiments on the Chemistry of Element 104 - Kurchatovium II. Chemical Investigation of the Isotope Which Undergoes Spontaneous Fission With a Half - Life of 0.3 Sec." This paper [31] - along with the three companion papers [30,32,33] - shows in detail how all of the work in the 1966 - 1969 period was carried out.

The actinide hypothesis suggests that element 104 will behave chemically like Hf. Thus if the 0.3 second isotope assigned as $^{260}_{104}$ could be shown to resemble Hf in its chemical properties, the earlier physical identification would be strengthened. Calibrations were made to show that HfCl_4 was transferred to the condensate trap of the apparatus (Fig. 9) under the chosen conditions of the experiment [31]. Therefore, if $(104)\text{Cl}_4$ behaved like HfCl_4 it

would be transferred to the trap also. But the design of the experiment was such as to prevent this from happening because the flow of the carrier gas was set such that the $^{260}_{104}$ (of the $T_{1/2} = 0.3$ sec) would decay along the detector chamber path before it reached the trap. Its decay pattern along the detector pathway was, indeed, used by the Dubna researchers to measure the half life as around 0.3 seconds, which was compatible with the value determined earlier by the physical methods [24]. It was stated [31] "positively" that the half life could not be 3.7 seconds. The later claims by the Dubna group [48] that they had supposed since 1966 to be studying $^{259}_{104}$ (actual $T_{1/2} = 3.0$ sec) rather than $^{260}_{104}$ (actual $T_{1/2} = 0.021$ sec) therefore cannot be accepted. The results of the second series of experiments [33] in 1969 were the same, and the experimentally measured half life of 0.5 seconds was used to calculate a heat of adsorption of $(104)Cl_4$ that correlated well with analogous elements (Fig. 12). All of these results were plausible and fitted together very well, but the credibility of the experiment was destroyed when it was shown that the half lives of the two possible isotopes ($^{260}_{104}$ and $^{259}_{104}$) were 0.02 seconds and 3.0 seconds. From the rate of gas flow through the system it was clear that the 0.02 second half life was too short for any significant transfer and thus of any opportunity to observe its chemical behavior. The 3.0 second half life had been ruled out "positively" by the Dubna group when they stated (on the

basis of the measured 0.3 second half life) that a 3.7 second half life was impossible [31].

The design and execution of the experiments conducted between 1966 and 1969 inextricably intertwined the half life determination and the determination of the chemical behavior of element 104. The plan of the experiment was such that the quantitative evidence for the identification of element 104 would come from the comparison of the heat of adsorption of its chloride with that of hafnium chloride and analogous elements [32]. The value of the half life entered into the calculation of the heat of adsorption [33]. The half life determination of 0.5 sec was wrong, so their calculated heat of adsorption [31,33] could not have been correct.

The identification of 104 on general qualitative grounds cannot be made either since the resolution was very low and various transuranium isotopes could reach the mica detector chamber. These isotopes were capable of spontaneous or neutron induced fission. There is no evidence internal to the experiment itself to distinguish an actinide element from a transactinide element. The ancillary experiments carried out to show that the fission tracks could not arise from actinide elements could be as wrong as the half life determinations of $^{260}_{104}$. The explanation of these chromatographic results appears to be that the Dubna researchers were misled by the small number of events they recorded into a false determination of the half life as

being 0.3 sec and thence into a false interpretation of the chemical properties of whatever they were seeing. In any case the conclusive evidence required in new element identification is lacking in the Dubna element 104 chemical experiments conducted during the period 1966 - 1969.

In 1971 the Dubna group took a new approach [48], based on a different principle, that did successfully achieve the chemical identification of element 104. This new approach was based on the concepts of thermochromatography.

The Dubna group stated that the results of these new experiments proved that their old experiments were correct. But the differences between the two methods are too great to establish such a linkage. In the (unsuccessful) approach taken between 1966 and 1969 the chemical channel was set at a constant temperature (Fig. 9), and the detection depended on the $^{260}_{104}$ nuclei decaying during their brief passage next to the mica detectors. This technique is therefore very sensitive to the half life of the isotope under study. The chemical resolution in the early 1966-1969 experiments was very poor (no chromatographic peaks were formed). In the second (successful) approach there was a steep temperature gradient in the chemical channel which caused the $(104)Cl_4$ to deposit out as a peak under the $HfCl_4$ peak (Fig. 19). Although the resolution of this experiment cannot be considered as good, the Dubna researchers showed that it was good enough to separate difficult elements such as Pu from Hf (and therefore 104). The thermochromatographic technique is very

insensitive to half life and cannot in general (even in principle) measure it. Since these 1971 results do not relate to those obtained earlier either in principle or in practice, they have no bearing on the priority of discovery of element 104.

Thus, the claim of priority by the Dubna scientists rests entirely on the quality of the physical evidence which they presented in the period 1964-1969 to back up their claim that they had observed $^{260}_{104}$ produced by the reaction $^{22}_{\text{Ne}}(^{242}_{\text{Pu}}, 4n)^{260}_{104}$. If we first consider the reported half life of the spontaneous fission activity originally assigned to $^{260}_{104}$ by Flerov et al. [24], the difficulty in establishing its correct value is disturbing. Aside from causing the "discovery" chemistry experiments to be designed erroneously around a 0.3 second activity, as just discussed, it is not so serious that the original value of about 300 milliseconds [24] was later reduced to 100 ± 50 milliseconds [46], to 80 ± 20 milliseconds [57], to 76 ± 8 milliseconds [61], and to 28 ± 6 milliseconds [65], if one makes due allowance for the difficulty of the experiments and the small number of events observed in any one experiment. However, it is disturbing that the Berkeley group was unable to observe the 76-millisecond component in any of the reactions where the isotope $^{260}_{104}$ should have been prepared. Figure 23 makes this point quite strongly. The Berkeley group's final conclusion [62] was that the true half life of $^{260}_{104}$ is 20 milliseconds. In a 1985 publication [65] by Dubna physicists the correctness of the shorter half life was apparently accepted: their measurement produced a value of 28 ± 6 milliseconds and they presented a possible reason for

distortion of the pattern of fission tracks observed in the earlier Dubna experiments.

It is also disturbing that the two groups were unable to agree on the formation cross section, or the upper limit to it, when both groups studied the identical reaction such as $^{249}\text{Bk}(^{15}\text{N},4\text{n})^{260}\text{104}$. These disagreements were reviewed in Section VII. The most detailed presentation and discussion of the conflicting results is given by Somerville et al [62].

If we turn now to the claims of the Lawrence Berkeley Laboratory group led by A. Ghiorso, we believe that this group presented credible evidence early in 1969 for three isotopes: 4.5-second $^{257}\text{104}$, 3-second $^{259}\text{104}$, and 65-second $^{261}\text{104}$. In the experiments done on the first two isotopes a pattern of alpha particle intensities and energies was observed that corresponded to no known nuclides. Daughter activity was collected by the alpha particle recoil technique discussed in Section II and this daughter activity was found to emit alpha particles with the unique properties known for $^{253}_{102}\text{No}$ and $^{255}_{102}\text{No}$, respectively. This genetic linkage is very powerful evidence for the atomic number and mass number assignments of the parent nuclei. Evidence for $^{261}\text{104}$ was found in samples prepared by the $^{248}\text{Cm}(^{18}\text{O},5\text{n})^{262}\text{104}$ reaction in experiments published [39] in 1970. An isotope decaying by the emission of 8.3 MeV alpha particles was found to produce a daughter activity identified as $^{257}_{102}\text{No}$. As was mentioned above in Section VII, the half life of the isotope $^{261}\text{104}$ is sufficiently long to make it an attractive candidate for chemical experiments. The well-calibrated ion exchange

elution method, in which a mixture of trace elements is absorbed from aqueous solution onto a Dowex 50 cation exchange resin and then selectively eluted by a solution of 0.1 molar alpha-hydroxyisobutyric acid adjusted to a pH of 4.0 was used by Silva, Harris, Nurmia, Eskola, and Ghiorso [40] to study the behavior of the 65-second activity attributed to element 104. They observed the rapid elution of the 65-second alpha emitter, together with tetravalent hafnium and zirconium tracers, well ahead of the known transuranium elements curium and californium and of added trivalent rare earth tracers. This elution behavior clearly distinguished the chemical identity of the 65-second activity as different from the actinide elements. The behavior is consistent with that predicted for element 104. The chemical experiments thus support the physically derived assignment of element 104. Additional extraction chromatographic experiments of a similar nature were carried out in 1975 by Hulet and coworkers [64].

A strong confirmation of the original claim by Ghiorso et al. [37] to the identification of 4.5-second $^{257}_{104}$ produced by the $^{249}_{\text{Cf}}(^{12}_{\text{C}}, 4n)^{257}_{104}$ reaction was made by a group working at the Oak Ridge National Laboratory. Bemis et al. [56] were able to reproduce the Berkeley results. In addition they measured the spectrum of X-rays emitted in coincidence with the alpha-particles attributed to $^{257}_{104}$ and found a pattern of K X-rays with the characteristic energies and relative intensities for element 102. The $K_{\alpha 1}$, $K_{\alpha 2}$, $K_{\beta 1}$, $K_{\beta 2}$ pattern was obtained (see Fig. 21). This is very strong evidence for the assignment of the alpha activity to element 104 owing to the uniqueness of the X-ray properties of the

daughter. (In 1985 a group at the UNILAC in Darmstadt [68] produced $^{257}_{104}$ by the $^{208}_{\text{Pb}}(^{50}_{\text{Ti},\text{In}})^{257}_{104}$ reaction and measured a half life and an α particle spectrum close to those reported by Ghiorso et al. [37] and by Bemis et al [56]. These results are included in Table 4.)

The ORNL group also repeated [52] the experiments of Ghiorso et al. [37] on the synthesis of $^{259}_{104}$ and obtained nearly identical results.

We conclude that the information published in 1969 by Ghiorso, Nurmia, Harris, Eskola, and Eskola [37] was correct and that no other publications on the three isotopes of mass number 257, 259, and 261 preceded theirs. Their information fully meets the criteria for discovery of new elements stated by Harvey et al. [23] and those stated by Flerov and Zvara [22] which were discussed in Section VI. The earlier claims by the Dubna group for definitive identification of an isotope of mass number 260 based on their results published in 1964 or in subsequent years up through 1969 cannot be accepted, nor do any of the results published later by this group reestablish the basis for their claim for priority of discovery. Therefore, we believe the Lawrence Berkeley Laboratory group should have the traditional right to suggest a name for the element. We suggest that their choice of the name Rutherfordium, with symbol Rf, be adopted.

Table 4. Reported Information on Isotopes of Element 104

Isotope	Half-life	Ref.	Decay Mode (E_α in MeV)	Ref.	Method of Preparation	Ref.
253	<u>1.8 sec</u>	[67]	SF	[67]	$^{206}\text{Pb}(^{50}\text{Ti}, 3n)^{253}104$	[67]
254	<u>0.5±0.2 msec</u>	[66]	SF	[66]	$^{206}\text{Pb}(^{50}\text{Ti}, 2n)^{254}104$	[66]
255	2 sec	[66]	SF (50%)	[66]	$^{207}\text{Pb}(^{50}\text{Ti}, 2n)^{255}104$	[66, 53]
	<u>1.7±0.2 sec</u>	[53]	SF	[53]		[55, 68]
	<u>1.4±0.6 sec</u>	[54, 55]	α 8.726 (55%) SF (45%)	[54, 55]		
	<u>1.4±0.2 sec</u>	[68])	α 8.766, 8.715 <u>8.625 (48±7%)</u> SF (52±7%)	[68]		
256	<u>6.7±0.2 msec</u>	[53]	SF (98%)	[53]	$^{208}\text{Pb}(^{50}\text{Ti}, 2n)^{256}104$	[53, 55, 68]
	<u>7.4±0.9 msec</u>	[55, 68]	SF	[54, 55]		
257	<u>4.5±1.0 sec</u>	[37]	α 8.70 (15%)	[37]	$^{249}\text{Cf}(^{12}\text{C}, 4n)^{257}104$	[37, 56]
	<u>4.8±0.5 sec</u>	[38, 44]	8.78 (20%)			
	<u>4.76±0.53 sec</u>	[69]	8.95 (30%) 9.00 (35%)			
	<u>4.3±1.3 sec</u>	[68]	α 7 groups from 8.597 to 9.012 SF (2.4%)	[68]	$^{208}\text{Pb}(^{50}\text{Ti}, 1n)^{257}104$	[68]
257	<u>3.8±0.8 sec</u>	[62]	SF 14±9% SF 8±5% EC (18%)	[62]	$^{249}\text{Cf}(^{12}\text{C}, 4n)^{257}104$	[62]
				[54, 55]	$^{208}\text{Pb}(^{50}\text{Ti}, 1n)^{257}104$	[53, 55, 68]
258	<u>13±2 msec</u>	[38]	SF	[38, 37]	$^{249}\text{Cf}(^{12, 13}\text{C}, 3, 4n)^{258}104$	[38, 37]
	<u>13±3 msec</u>	[52, 62]	SF	[62]	$^{246}\text{Cm}(^{16}\text{O}, 4n)^{258}104$	[62]
259	3 sec	[37]	α 8.77 (60%)	[37]	$^{249}\text{Cf}(^{13}\text{C}, 3n)^{259}104$	[37, 52]
	<u>3.0±1.3 sec</u>	[52]	<u>8.86 (40%)</u>		$^{248}\text{Cm}(^{16}\text{O}, 5n)^{259}104$	[37]
	<u>3.4±1.7 sec</u>	[62]	SF 6.3±3.7%	[52]	$^{249}\text{Cf}(^{13}\text{C}, 3n)^{259}104$	[52]
	<u>3.2±0.8 sec</u>	[51]	SF 7-12%	[47, 51]	$^{246}\text{Cm}(^{18}\text{O}, 5n)^{259}104$	[51]
260	300, 80 msec	[24, 57, 61]	SF	[24, 57, 61]	$^{242}\text{Pu}(^{22}\text{Ne}, 4n)^{260}104$	[24, 61]
	<u>21±1 msec</u>	[62]	SF		$^{249}\text{Bk}(^{15}\text{N}, 4n)^{260}104$	[58, 62, 65]
					$^{248}\text{Cm}(^{16}\text{O}, 4n)^{260}104$	[62]
					$^{249}\text{Cf}(^{18}\text{O}, \alpha 3n)^{260}104$	[62]
					$^{242}\text{Pu}(^{22}\text{Ne}, 5n)^{259}104$	[47]
261	<u>65±10 sec</u>	[39]	α 8.28	[39]	$^{248}\text{Cm}(^{18}\text{O}, 5n)^{261}104$	[39]
			SF < 10%	[62]		
262	<u>47±5 msec</u>	[62]	SF	[62]	$^{248}\text{Cm}(^{18}\text{O}, 4n)^{262}104$	[62]
					$^{244}\text{Pu}(^{22}\text{Ne}, 4n)^{262}104$	[62]

Best current values of half life and α energies are underlined.

IX. Summary of Properties of Isotopes of Element 104

It may assist the reader in evaluating the historical record on element 104 presented in this document to have a reference table in which the currently known information on all isotopes of the element is collected. We present such a table here for this purpose, based on the cited references. The data base maintained at the Lawrence Berkeley Laboratory by the Nuclear Data Group was utilized to determine whether any important reference had been overlooked.

X. Chronological Sequence of Publications on Element 105

In this section we cite essential publications relative to the identification of isotopes of element 105 and summarize the results and claims from each of these studies. In the following Section XI we present our evaluation of these results and claims.

a) Soviet publications, 1968-1970

During 1968 the Dubna group led by G.N. Flerov studied [70] the reaction of 123 MeV ^{22}Ne ions with targets of ^{243}Am , where the target thickness was about one milligram per cm^2 . At this energy it was expected that complete fusion of the projectile and target would occur and, after removal of excess energy by the evaporation of 4 or 5 neutrons, it was expected that isotopes of element 105 with mass numbers 260 and 261 would be produced. The alpha-particle energy for $^{260}_{105}$ was expected to be about 9.6 MeV and for $^{261}_{105}$ about 9.4 MeV. Because of the strong loss of product nuclei by fission competition at each stage of neutron emission the formation cross section was expected to be small but measurable, i.e. about 10^{-33}cm^2 .

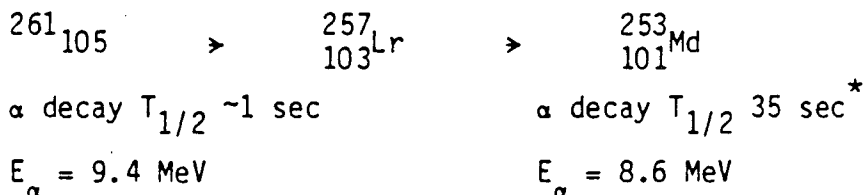
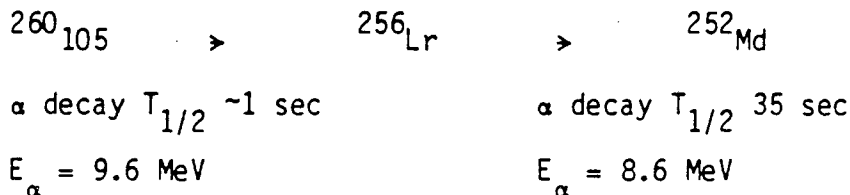
The experiments were carried out at the 300 cm Dubna cyclotron using the apparatus shown in Fig. 24. The target was continuously bombarded with a $^{22}\text{Ne}^{+10}$ ion beam at a current of about 5 (electrical) microamperes. The reaction product nuclei recoiled out of the target into the gas filled reaction chamber and were

transported in the gas stream (a nitrogen and helium mixture) along the capillary (5), emerged from the capillary tip, passed through the central hole of an annular silicon semiconductor detector and finally impinged on and stuck to the metal surface of the collector wheel (7).

Some of the alpha particles emitted by reaction products collected on the wheel were intercepted by the alpha detector (6), whose active surface faced downward. Pulses from the alpha detector were amplified in suitable circuitry and subjected to pulse height analysis. The alpha particle spectrum so obtained is shown in Fig. 25. Many of the alpha particle peaks in this spectrum are the result of reactions of ^{22}Ne with minute amounts of lead in the target. This is particularly true of the energy region 8.8–9.1 MeV. Some of the pulses fall in the energy region 9.1–9.8 MeV expected for isotopes 260 and 261 of element 105.

In an attempt to strengthen the assumption that some of these 9.1–9.8 MeV alpha particles were correctly assigned to element 105 the method of "time correlation" of parent and daughter alpha pulses was used.

First the assumption was made on the best available information in 1968 that the decay sequences would be the following.



[*Authors' note. This was the half life assumed for ^{257}Lr in 1978. In later work the correct value of 0.7 seconds was measured at Berkeley. This modifies the conclusions from the study.]

The essential feature of these two decay sequences is that decay of a nucleus of element 105 should be followed within 160 seconds, with nearly 100 probability, by the emission of an 8.6 MeV alpha particle from its element 103 daughter. The experimental procedure to take advantage of this expected time correlation was to stop the ^{22}Ne bombardment of the target whenever an alpha pulse in the range 8.8 to 10.3 MeV was recorded and register any alpha particles that were emitted in the next 160 seconds (3.6 half lives of the daughter). For each pair of "start" and "stop" pulses the alpha energies of both pulses were recorded. After suitable sorting and analysis of these data it was reported by the authors that start pulses of 9.1-9.8 MeV (the region of expected energy for $^{260}_{105}$ and $^{261}_{105}$) were followed in 40 percent of the cases with a stop pulse in the alpha particle range of 8.35 to 8.6 MeV. If the start pulses fell below 9.1 MeV or above 9.8 MeV then stop pulses of 8.35-8.6 MeV occurred only 10 percent of the time. These results were taken as tentative evidence that alpha particles with energies from 9.1 to 9.8 (with weak peaks at 9.15, 9.4 and 9.7 MeV) were emitted by isotopes of element 105 with mass numbers 260 and 261.

Additional conclusions drawn from the experiments reported in Ref. [70] were that the half life for the 9.4-MeV alpha emitter, tentatively assigned to $^{261}_{105}$, was in the range of 0.1 to 3 seconds. The emitter of 9.7 MeV alpha particles tentatively assigned

to $^{260}_{105}$ had a formation cross section of 10^{-34} cm² and a half life >0.01 second.

In a later publication [71] Flerov and his co-authors commented on this initial report about the possible alpha decay of element 105. They observed that the formation cross section of 10^{-34} cm² was lower than expected by perhaps a factor of 10. Part of this missing activity might be attributed to alpha particles of energy less than 9.1 MeV which, though possibly present in the spectrum, were obscured by background activity in the 8.8 to 9.1 MeV region produced by microadmixture of lead in the target. They also commented that part of the missing cross section might be attributed to a spontaneous fission competition in the decay of element 105. They carried out additional experiments to search for evidence of this spontaneous fission branch. These experiments were described in a series of Dubna reports [71,72] and in a published article [73]. The last reference provides the best summary for our purpose.

These experiments were performed with the technique used earlier in the search for element 104. The apparatus shown in Fig. 17 was used for the bombardment of the ^{243}Am target with 119-MeV ^{22}Ne ions and the capture of recoil products on a moving nickel tape which transported them past a series of detectors made of phosphate glass. As the reaction products disintegrated by spontaneous fission, one of the fission products would leave the nickel carrier tape and impinge on a glass detector where its track would later be seen after chemical treatment. This distribution of tracks along the detectors, taken in connection with the known speed of the nickel tape, allowed the

determination of the half life of the fissioning species as shown in Fig. 26. The 2 second component was attributed to element 105 for reasons we will discuss. The best value of the half life, quoted in the summary section of the paper, was 1.8 ± 0.6 sec.

Owing to the fact that registration of fission fragments does not supply definitive information on the charge and mass number of the emitter, the authors carried out additional experiments and cited additional arguments to strengthen their assignment of this 2-second activity to element 105.

First, they did an experiment with a collimator grid behind the target and measured the reduction in yield caused by use of grids with different degrees of collimation. Products with mass number very low compared to the compound nucleus, such as ^{242m}Am , were expected to be reduced in yield by about a factor of 5. Products close to the compound nucleus, such as elements 102 to 105, were expected to be reduced in yield only by a factor of 2 under the conditions studied. The results indicated a factor of 5 reduction for the ^{242m}Am , as expected, but only a factor of 2.2 for the 2 second activity. The authors state clearly that their angular correlation (collimation) method could not distinguish between compound nucleus reactions and direct reactions of the $(^{22}\text{Ne},\text{pxn}), (^{22}\text{Ne},\alpha\text{xn})$ type. However, they were able to cite reasons why no known or expected isotope of elements 102, 103, or 104 could account for the observed 2 second activity. Thus, by elimination, they assigned it to element 105. They commented that additional and unambiguous identification would come by high speed radiochemical methods of study of the chemical properties.

As additional supporting evidence for their conclusions, the authors studied the yield of the 2-second nuclide as a function of the ^{22}Ne ion energy in the energy range 110 to 130 MeV. According to the theory of compound nucleus reactions (with parameters in the theory evaluated semiempirically) this is the energy region in which the $^{243}\text{Am}(^{22}\text{Ne},4n)^{261}_{105}$ reaction, and the corresponding $(^{22}\text{Ne},5n)$ reaction leading to $^{260}_{105}$, should have a characteristic rise and fall in reaction cross section. The results are shown in Fig. 27. The experimental points behave in the manner predicted by the theory, which the authors take as supporting evidence for the formation of an isotope of element 105 with mass number either 260 or 261, but with preference for 261. The formation cross section at the peak (^{22}Ne ions of 117 MeV) was $(5 \pm 1.5) \times 10^{-34} \text{ cm}^2$. The half life of the isotope was measured to be 2.2 ± 0.5 seconds. The experimental method was calibrated with a study of the $^{235}\text{U}(^{22}\text{Ne},5n)^{252}_{102}$ reaction in which the properties of the product were known from published research.

When these studies of the spontaneously fissioning products of the interaction of ^{243}Am and ^{22}Ne were completed and the conclusion had been reached that a 1.8 second isotope of element 105 with probable mass number 261 had been discovered, a new study of the alpha decay of element 105 was started by Druin and his coworkers [74]. This study used the same principles and techniques (but different apparatus) as had previously been used by Flerov et al. [70] and described above. The new set of experiments was designed to look for an α branch of the same isotope which decayed by spontaneous fission

with a half life of 2 seconds. A strong effort was made to remove troublesome lead contamination in the ^{243}Am target. A collimator grid behind the target further reduced the contribution of α emitters produced by transfer reactions, having wide angular distributions, from lead impurities in the target.

The alpha spectrum obtained by Druin et al. [74] is shown in Fig. 28. The 11.65 MeV peak in this spectrum is caused by $^{212\text{m}}\text{Po}$ (45 seconds). The 9.1 MeV activity was shown to have a half life of 1.4 seconds within a few tenths of a second error. The decay of the 8.90 MeV peak was complex including a short lived part with a half life value of roughly 1.5 seconds. Peaks in the 8.30–8.60 MeV region had a half life of 35 seconds which agrees with the half life reported earlier for ^{256}Lr and ^{257}Lr . [We should note that it was discovered later by LBL that the correct half life for ^{257}Lr is 0.7 seconds rather than 35 seconds.]

The authors of this paper made use of the method of amplitude-time "correlation" of parent and daughter activity to attempt to show a genetic relationship between the peaks in the 8.8 to 9.2 MeV range (assigned by them to element 105) and the peaks in the 8.3–8.6 MeV range (assigned to ^{256}Lr and ^{257}Lr). As in the previous study [70] the recording of an alpha particle in the higher (parent) energy range caused a stoppage in the collection of new recoil activity and the start of a waiting period of 140 seconds during which the daughter alpha spectrum was recorded. Some enhancement of the pulses in the 8.3–8.6 MeV region over the chance rate was observed.

The conclusions of the authors were expressed as follows.

"The results of analysis of amplitude-time correlations provide a basis for suggesting that we are observing an α emitter with atomic number 105. The total number of primary α particles with energy 9.1 and 8.9 MeV which could be assigned to element 105 was ~60, while 4 pulses were recorded which corresponded to an energy ~9.4 MeV, which is consistent with the 1968 data" [70].

They state further that: "It should be noted that the half life of the observed α emitter agrees within experimental error with the spontaneous fission data [72]. Therefore it appears likely to us that we have detected the α -decay of the same isotope for which spontaneous fission has been observed."

b) Soviet publications on the chemical identification
of element 105, 1970-1975

A JINR internal report by Zvara and coworkers [75], which appeared in 1970, described experiments to investigate the chemical properties of the spontaneously fissioning activity with a half life of 2 seconds which we have described above. The purpose was to provide chemical evidence for the assignment of this activity to an isotope of element 105. These experiments exploited the expected difference in the volatility of gaseous chlorides of different elements and selective absorption of different elements on the surface of a glass tube in which a strong thermal gradient had been established. This method had been developed for use in the heavy element region by Zvara and his

group and used previously in their studies of element 104 in 1971, as we have discussed earlier in this report.

It was proposed by the authors that the chemical properties of element 105 must be analogous to tantalum and specifically that its higher valent chloride (and also possibly its oxychloride) must be a comparatively volatile compound. The experiment was designed to take advantage of the volatility of the chloride compounds of element 105 in order to purify the element and to separate it from other elements of higher or lower volatility.

Owing to the 2-second half life it was necessary to work with atoms as they recoiled out of the ^{243}Am target during bombardment with ^{22}Ne ions. These recoil atoms were slowed down in a stream of hot nitrogen at 300°C and transported away from the nuclear reaction chamber. About 4 cm from the chamber the nitrogen gas stream merged with another nitrogen stream containing the vapor of substances used as chlorinating agents. In some experiments the chlorinating agent was thionyl chloride (SOCl_2); in most cases it was a mixture of SOCl_2 and TiCl_4 . The gas stream containing the chlorinating agents and the recoil atoms continued to move through a straight glass tube of 170 cm length and 4 mm internal diameter, divided into two sections. The first 30 cm section was held at a temperature of 300°C and was designed to remove from the gas stream those elements which form non-volatile chlorides. Indentations on the inner surface of the tube caused strong turbulence which aided the deposition of the nonvolatile compounds. In agreement with this, by-product ^{44}Sc deposited nearly completely in this part of the apparatus. Only a few

percent of any non-volatile chloride could be swept through this first tube. In the second part of the glass tube a temperature gradient starting with an initial 300°C and dropping to 50°C was created. It was expected that different elements would be deposited along the tube in an order determined by the volatility of their chloride compounds.

Inside this part of the tube there was placed a series of mica plates upon which the molecules of the chloride compounds of any spontaneously fissioning isotope might deposit. The fission fragments from these isotopes would cause track formation in these mica plate fission detectors, which upon later chemical development and examination would reveal the location of the deposit of the fissioning species.

It was necessary to have some calibration of this method so that the distribution pattern of known elements could help in the interpretation of the results. In addition to experiments done off line with radioactive tracers, it was also possible to use the α , β , and γ properties of fission products and some heavy element by-products (e.g. ^{246}Cf) of the $^{243}\text{Am} + ^{22}\text{Ne}$ reaction to locate the positions of deposit of several elements. In addition, the interaction of the ^{22}Ne beam with the aluminum support of the target provided reaction products such as ^{24}Na and ^{44}Sc . Finally, a light coating of samarium oxide was placed over part of the americium target so that ^{170}Hf would be produced along with the 2-second spontaneous fission activity. ^{170}Hf was an important isotope for monitoring and calibrating the operation of the system.

Four experiments were done with a continuous beam on target for periods ranging from 37 to 59 hours. The parameters of the four runs are summarized in Table 5 and the results are shown in Fig. 29.

In Run I where SOCl_2 alone was used as the carrier and chlorinating gas no fission events were registered. In Runs II-IV a total of 18 events were recorded with the distribution of individual events as shown in the figure. It is important to note also in figure 29 the zone of deposition of the actinide elements (Cm and Cf) and of hafnium, and of several other elements with volatile chlorides.

Alternate explanations of the nuclear reaction origin of the nuclide giving rise to the fission tracks, such as neutron induced fission of traces of uranium in the glass or mica or as short-lived spontaneously fissioning isomers of lighter elements, were considered and dismissed; the amount of uranium in the glass or mica and the cross sections for formation of the isomers would be too low to explain the observed tracks. Nor could any long lived isotope of an actinide element be produced in sufficient yield to account for the effect. Moreover, actinide and rare earth chloride compounds would deposit in the first 30 cm section of the tube which was held at 300°C , whereas the observed fission events did not deposit there but further along in the second section of the tube where the temperature gradient was maintained.

In a discussion of the results shown in Fig. 29, the authors of Ref. 75 commented that the 18 fission tracks observed in experiments II-IV are located in a section of the tube roughly equivalent to the section where ^{170}Hf deposited and in a section at a higher

Table 5. Conditions of the Experiments Designed for the Chemical Identification of Isotopes of Element 105 (Zvara et al [75])

Number of the experiment	Carrier Partial Press (mm. Hg.)	Consumption of gas in the gradient tube (liters)	Duration of experiment (hours)	Integrated beam particles	Energy of ^{22}Ne Ions (MeV)	Number of registered acts of spontaneous fission
I	$\text{SOCl}_2(25)$	3.0	37	6.2×10^{17}	114	0
II	$\text{SOCl}_2(12)$ $\text{TiCl}_4(1.1)$	4.5	57	4.9×10^{17}	118	6
III	$\text{SOCl}_2(4.8)$ $\text{TiCl}_4(1.3)$	4.0	59	4.5×10^{17}	119	8
IV	$\text{SOCl}_2(6.5)$ $\text{TiCl}_4(1.3)$	4.1	45	3.6×10^{17}	119	4

temperature than the section in which ^{90}Nb deposited. They concluded that the results showed that the spontaneously fissioning activity comes from an element forming a comparatively volatile chloride as might be expected for ekatantalum (element 105). They were unable to measure the half life with any precision, but set a lower limit of 0.2 seconds and argued that it was most likely the 2-second isotope reported by Flerov et al [71,72]. They also pointed out that the observed pattern of the fission tracks could be distorted by the short half life (to indicate somewhat less than the correct chloride volatility) because of the decay in flight during the time of gas transport through the tube.

The experiments described in Ref. 75 were presented later at a conference in Germany [76]. There is also a short review of them in a 1976 article, available in English [77]. This last article goes on to describe later experiments done with a bromide rather than a chloride system. Use of a brominating carrier consisting of a mixture of BBr_3 and Br_2 gave rapid equilibration and offered several experimental advantages such as lower temperature of absorption of the compounds under study and less corrosion of the walls of the reaction chamber.

Bromide components of lower volatility deposited in the first section of the nickel tube which was held at about 250°C . Among these elements were rare earths, scandium, and the actinide elements. The behavior of many elements had been learned in a previous set of experiments to develop the method [78]. In the long section of nickel tubing where a decreasing gradient of temperature was maintained, the

absorption zone of volatile NbBr_5 was defined by the presence of ^{90}Nb . Spontaneous fission tracks were found in detectors located at a somewhat higher temperature in a region where the bromide of hafnium was shown to deposit. See Fig. 30. This was taken as evidence of strong discrimination from the actinide elements, whose tribromide compounds would not be this volatile. Arguments are repeated in this article for excluding alternate explanations of the origin of the observed spontaneous fission tracks.

c) Lawrence Berkeley Laboratory publications, 1970-1971

Early in 1970 Ghiorso, Nurmia, Eskola, Harris and Eskola [79] announced the synthesis of an isotope of element 105 with mass number 260 by the bombardment of ^{249}Cf with ^{15}N ions at the heavy ion linear accelerator (HILAC) at the Lawrence Berkeley Laboratory. This isotope was observed to decay by alpha particle emission with a half life of 1.6 ± 0.3 seconds. Alpha groups were reported at 9.06 (55%), 9.10 (25%) and 9.14 MeV (20%). Branching decay by spontaneous fission was set at less than 20%. The Z and A of $^{260}_{105}$ were unambiguously identified by alpha recoil separation and measurement of the known ^{256}Lr daughter.

In these experiments a 300 microgram/cm² target of californium was bombarded with ^{15}N ions at four energies from 79 to 103 MeV. The element 105 reaction products recoiled out of the thin target into a helium gas stream which was pumped through a small orifice into an adjacent vacuum chamber. These heavy nuclear products were deposited

on the periphery of a vertically mounted wheel. The wheel was periodically rotated to place the collected transmutation products next to a series of silicon solid-state detectors which measured the energies of the emitted alpha particles. From the distribution of the α -particle decays among these detectors and the decay times, the half lives of the α -emitting isotopes were determined. The series of alpha-particle energy spectra are shown in Fig. 31.

An important feature of the experiment was that daughter activity from the decay of approximately half of the deposited element 105 alpha activity could recoil into the surface of the silicon detector. To measure this daughter activity each of these primary detectors was periodically shuttled to a position opposite another similar silicon detector where at high geometry the two detectors together could analyze the daughter alpha activity which had recoiled off the wheel.

Twenty detectors in all were involved in a complex shuttling operation. A computer was used to tag and record the spectra from individual detectors. Figure 32 shows the main result of these measurements of the daughter activity. The most important feature is the presence of alpha particles in the 8.2 to 8.6 MeV region (with a major peak at 8.4 MeV) which the authors assigned to ^{256}Lr on the basis of the following observations.

- (1) The spectrum in the 8.2- to 8.6-MeV region closely resembles that of a known sample of ^{256}Lr (see insert).
- (2) The 8.4 MeV daughter activity decays with a half life of 30 ± 19 seconds, which agrees with the previously known half life for ^{256}Lr .

- (3) The number of alpha particles in the ^{256}Lr peaks agrees closely with the number to be expected if the 9.1 MeV peak in the previous Fig. 31 was correctly assigned to the parent $^{260}_{105}$.
- (4) Finally the excitation function (yield versus ^{15}N energy) plotted from the experimental data (see Fig. 33) agrees with a calculated yield curve for the $^{249}\text{Cf}(^{15}\text{N},4n)^{260}_{105}$ reaction.

At the time these experiments were performed (early 1970) the Berkeley group was unaware of any results from Dubna except the first publication [70] of that group on alpha decay properties, a short summary of which we have presented above. Ghiorso and coworkers [79] presented their views as to why these Dubna results were insufficient to establish correctly the properties of either alpha emitting $^{261}_{105}$ or $^{260}_{105}$. In particular, there appeared to be no correspondence between the alpha spectra observed by the two groups. The Berkeley group then claimed discovery of the element and suggested the name Hahnium (Symbol Ha) in honor of the German chemist and codiscoverer of neutron-induced fission, Otto Hahn.

This first study [79] by the LBL group was followed a year later by a second publication [80] presenting evidence for two additional alpha-emitting isotopes of element 105. By bombardment of a ^{250}Cf target with ^{15}N ions and ^{249}Bk targets with both ^{16}O and ^{18}O ions they produced an 8.93-MeV 1.8-second activity which they assigned to $^{261}_{105}$ and an 8.45-MeV 40-second activity which they assigned to $^{262}_{105}$. Genetic relationships between the new activities and the known ^{257}Lr and ^{258}Lr daughters were established by α -recoil of

these daughter nuclei into separate silicon detectors and by correlating in time their α decays to the parent α decays. A few details of these studies are given in the following paragraphs.

The experimental techniques were very similar to those used in the earlier study [79] of $^{260}_{105}$. Nuclear reaction products which recoiled out of the target were transported by a helium gas stream and deposited on the periphery of a wheel. This wheel was periodically rotated to place the collected recoil atoms next to a series of peripherally mounted silicon detectors. There were seven detector stations arranged at 45° intervals around the wheel. Two movable and two stationary detectors were used at each of seven locations. While one set of movable detectors was recording alpha events from atoms on the wheel, the other set and its stationary complement were analyzing the daughter activities which had recoiled off the wheel into the movable detectors. Alpha particle pulses were recorded together with tag information on the detector location and time. From the spectra from all detectors including the tag information and the time constants for movement of the collector wheel and for shuttling of the moveable detectors from the parent to the daughter data-taking mode, it was possible to deduce spectral and time decay information both for the parent and the daughter.

The alpha-particle spectra displayed in Fig. 34 resulted from bombardments of the $^{250}_{\text{Cf}}$ target with 83 MeV $^{15}_{\text{N}}$ ions. The complex group of peaks at about 8.9 MeV was assigned to $^{261}_{105}$ and its alpha-decay daughter, 8.87 MeV 0.6 second $^{257}_{\text{Lr}}$, whose properties were known from a previous study [81]. The whole complex

decays with a half life of 1.8 ± 0.6 seconds, or with the same half life as the 8.93 MeV group. This is consistent with the ^{257}Lr daughter being supported (i.e. replenished) by the decay of its parent. Only a few counts in the region of 8.87 MeV were recorded in the off-wheel detectors when daughter activity was under investigation, but these were distributed in a time pattern consistent with the short 0.7 second half life of ^{257}Lr . More convincing evidence for the relationship of parent alpha pulses of 8.93 MeV to daughter alpha pulses of 8.87 MeV came from an examination of "time-correlated" events. In this examination the authors sorted the data to see what alpha pulses in the 8.7 to 9.1 MeV interval were followed within 6 seconds (i.e. within several half lives of ^{257}Lr) with a second pulse in this energy region. A very clear correlation of an 8.93 MeV parent with a daughter emitting a complex spectrum with peaks at 8.79 and 8.86 MeV was noted. The correlation exceeded the expected random effect by a large factor.

In the case of the isotope of mass 262, the alpha-particle spectra displayed in Fig. 35 resulted from a 118 microampere hour bombardment of a ^{249}Bk target with ^{18}O ions. There is a complex alpha spectrum with prominent peaks at 8.45 and 8.66 MeV which decay with a 40 ± 10 second half life. These peaks were assigned by the authors to the mass 262 isotope of element 105. In their notation the isotope is ^{262}Ha . The 8.66 MeV peak is masked by the 8.6 MeV cluster of peaks belonging to the daughter ^{258}Lr .

The complex spectra in Fig. 35 include peaks from lighter elements like francium which result from trace lead impurity in the target. The ^{256}No and ^{257}No activity was produced from the ^{249}Cf that gradually grows into the purified ^{249}Bk target as it decays.

As in the study of the 1.8-second isotope $^{261}\text{105}$, the genetic linkage of the 40-second isotope $^{262}\text{105}$ to its daughter ^{258}Lr was studied by examination of the alpha activity which recoiled into the detector from the primary deposit on the wheel. The detector was moved away from the wheel and subsequent alpha emission was recorded. Alpha activity in the 8.5–8.7 MeV range was found with a half life of 4.5 ± 2 seconds in agreement with the known properties of ^{258}Lr ($T_{1/2} = 4.2$ seconds, α groups at 8.60 MeV). Examination of the "time correlation" of the α peaks from parent and daughter nuclei also supported the genetic linkage of the two.

The LBL authors of this study of two isotopes of element 105 commented on several of the Soviet papers which we have reviewed earlier in this report.

In the case of the study by Druin et al. [74] of a 1.4 ± 0.6 -second alpha-emitter, produced by bombarding ^{243}Am with ^{22}Ne ions, the Soviet authors gave their reasons based on "time-correlated" mother-daughter events for assigning the 1.4-second activity either to $^{260}\text{105}$ or $^{261}\text{105}$. Ghiorso et al. [80] point out that only the former is a possibility because the half life of 30 seconds assumed by Druin for ^{257}Lr was incorrect. In the case of the report of Flerov and coworkers [71,73] on a possible 1.8-second spontaneously fissioning isotope of element 105, Ghiorso et al. [80] comment that

their rough limits on the spontaneous fission branching of the 1.6-second alpha-emitting $^{260}_{105}$ and 1.8-second $^{261}_{105}$ are compatible with the possibility that the 1.8 second activity reported by Flerov and his coworkers could be due to one of these isotopes.

d) Oak Ridge National Laboratory publications, 1977

In 1977 a group of scientists [82] at the Oak Ridge National Laboratory repeated the experiments of the Lawrence Berkeley Laboratory on $^{260}_{105}$, duplicated the results, and did further measurements to confirm the mass and atomic number assignment.

The nuclide, $^{260}_{105}$, was produced in the $^{249}_{98}\text{Cf}(^{15}_{7}\text{N},4n)$ reaction at the Oak Ridge Isochronous Cyclotron and its decay properties were investigated. Measured alpha-particle energies and intensities were: 9.041 ± 0.014 MeV (48 ± 5), 9.074 ± 0.014 (25 ± 3) and 9.12 ± 0.017 MeV (17 ± 3). The half life was measured as 1.52 ± 0.13 seconds (see Fig. 36). Alpha branch decay was measured as 90.4 ± 0.6 and spontaneous fission branching as 9.6 ± 0.6 . All those values are close to those reported by Ghiorso et al. in 1970.

Bemis et al. [82] provided a very definitive proof of the assignment of atomic number 105 to the 1.52-second alpha activity by measurement of the spectrum of L X-rays in coincidence (within 200 nanoseconds) with the alpha particles in the energy range associated with the 1.52-second activity (i.e. 9.00-9.20 MeV). Figure 37 shows the photon spectrum so obtained. The energies and relative intensities of the peaks match with those expected from the L X-ray

spectrum for element 103 and are separated from the peaks of neighboring lower-Z elements by several times the energy resolution of the detector systems. This L X-ray "fingerprint" is unique for element 103. The genetic relationship of the 1.52-second alpha activity (assigned as $^{260}_{105}$) to a 26 second $^{256}_{\text{Lr}}$ daughter was established by the "time correlation" method. When alpha particles associated with the 1.52-second activity (i.e. α groups in the 9.00–9.20 MeV range) were recorded in a specific semiconductor detector all subsequent alpha particles in the next 35 second period were recorded and tagged with time as well as pulse-amplitude information. Those daughter alpha particles agreed both in time distribution and energy distribution with the known properties of 25.9 ± 1.7 -sec $^{256}_{\text{Lr}}$. See Fig. 38.

Bemis et al. [82] concluded their paper with these comments. (We have changed the reference numbers to correspond to ours.)

"1.) We have produced in the reaction of $^{15}_{\text{N}}$ with $^{249}_{\text{Cf}}$ a 1.52-sec alpha activity with alpha particle groups at 9.04, 9.07, 9.12 MeV, similar to the results of the earlier experiments of Ghiorso et al [79]; 2.) in addition, we have definitively established the atomic number for this activity as 105 and established the mass number as 260; 3.) we see no evidence of the 8.9-, 9.4- or 9.7-MeV alpha particle groups in the decay of $^{260}_{105}$ as reported in Ref. 70 and 74; 4.) we do detect a spontaneous fission activity with a half life similar to that for $^{260}_{105}$ derived from alpha decay data, but beyond this similarity, we are unable to establish a definitive link between these activities.

Our results for $^{260}_{105}$ completely corroborate and extend the earlier experiments of Ghiorso et al [79]. The unique identification provided for element 105 in our present experiments unequivocally supports the discovery claims for element 105 proffered by Ghiorso et al [79]."

This same group of ORNL scientists in collaboration with scientists from the Lawrence Livermore National Laboratory [83] restudied the properties of the nuclide $^{262}_{105}$. They produced it by the same reaction, $^{249}_{\text{Bk}}(^{18}_0,5n)^{262}_{105}$, reported in the original paper by Ghiorso, Nurmia, Eskola and Eskola [80]. They measured a more accurate value for the half life of 34.1 ± 4.6 seconds and for the spontaneous fission branching ratio of $78 \pm 6\%$. They also carried out a study [84] of the mass asymmetry and total kinetic energy release in the spontaneous fission of $^{262}_{105}$ in the hope that they would observe some unusual features (symmetric mass division and exceptionally high kinetic energy release) predicted by some theorists, but their results showed no evidence for them.

e) Soviet and German publications on light isotopes
of element 105, 1976-1985

In connection with our discussion of element 104 we cited discovery of some low-mass isotopes by the bombardment of $^{207}_{\text{Pb}}$ and $^{208}_{\text{Pb}}$ targets with $^{50}_{\text{Ti}}$ by Oganessian and coworkers [53]. The "cold fusion" of the projectile and the shell-stabilized lead target resulted in the production of a compound nucleus of such low

excitation that only 2 neutrons had to evaporate before the residual nucleus was deexcited enough to survive.

In 1976, Oganessian and his Dubna coworkers [85] applied this approach to the synthesis of light isotopes of element 105 by bombardment of ^{209}Bi with ^{50}Ti , of ^{208}Pb with ^{51}V , and of ^{205}Tl with ^{54}Cr . In all three cases the compound nucleus was $^{259}\text{105}$. By use of bombarding energies which would emphasize the evaporation of two neutrons from the compound nucleus to form $^{257}\text{105}$, these workers observed a 5 ± 1.7 second spontaneous fission activity which they assigned to $^{257}\text{105}$. They estimated the spontaneous fission branching as about 20%. The best results were obtained with the $^{209}\text{Bi} + ^{50}\text{Ti}$ combination.

In the same set of experiments Oganessian et al. [85] looked for the production of element 107 by bombardment of ^{209}Bi with ^{54}Cr and ^{208}Pb with ^{55}Mn ; in both cases the compound nucleus was $^{263}\text{107}$. They again saw the 5 second spontaneously fissioning isotope and attributed its presence to $^{257}\text{105}$ formed by the alpha decay of a short lived $^{261}\text{107}$ formed by the reaction, $^{209}\text{Bi} (^{54}\text{Cr}, 2n) ^{261}\text{107}$.

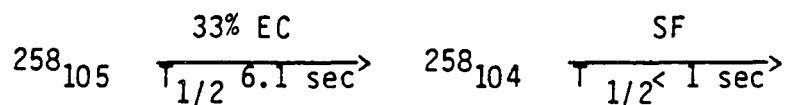
A group at the UNILAC accelerator of the Gesellschaft für Schwerionenforschung (GSI), Germany, also studied the production of light isotopes of element 105 by the "cold fusion" of ^{50}Ti with ^{209}Bi . Hessberger and coworkers [86] made use of an on-line velocity filter called SHIP to separate the recoiling products from this reaction, to collect these products by implantation into an array of position-sensitive surface-barrier detectors, and to measure the

α -decay and spontaneous fission of the implanted products. The ^{50}Ti beam energy was chosen close to the Coulomb barrier so that the ($^{50}\text{Ti},1n$) and ($^{50}\text{Ti},2n$) reactions would be emphasized.

In these experiments a 4.4 ± 0.9 second alpha emitter was identified and assigned to the nuclide, $^{258}_{105}$. The alpha spectrum consisted of four groups: 9.299 (8%), 9.172 (59%), 9.078 (28%) and 9.008 MeV (5%). In addition a 1.4 ± 0.6 -second alpha emitter was identified and assigned to $^{257}_{105}$. The alpha spectrum consisted of three groups: 9.160 (30%), 9.071 (30%), and 8.970 MeV (40%).

The Z,A assignments of these nuclides were greatly strengthened by use of the "time correlation" of alpha events from parent activities with the daughter and granddaughter activities whose alpha energies were known from previous studies. Thus $^{258}_{105}$ was linked to its daughter products $^{254}_{\text{Lr}}$, $^{254}_{\text{No}}$ and $^{250}_{\text{Md}}$ and $^{257}_{105}$ was linked to its decay products $^{253}_{\text{Lr}}$, $^{249}_{\text{Md}}$, and $^{245}_{\text{Es}}$.

Hessberger et al. [86] observed spontaneous fission activities with half lives close to those assigned to $^{258}_{105}$ and $^{257}_{105}$ but found the interpretation of the fission events to be much less clearcut. For SF events observed at ^{50}Ti bombarding energies where production of 4.4-second $^{258}_{105}$ was maximal they observed a 6.1 ± 1.0 -second fission decay which they interpreted, based on a number of considerations, as the fission of $^{258}_{104}$ formed by electron capture decay of $^{258}_{105}$.



For SF events observed at ^{50}Ti bombarding energies corresponding to maximum production of 1.4-second $^{257}_{105}$ Hessberger et al. [86] observed a 1.7 ± 0.5 -second fission activity, which may correspond to a 17% fission branch for $^{257}_{105}$. They commented on the previous report by Oganessian et al. [85] of a 5-second spontaneous fission half life, which they had assigned to $^{257}_{105}$, in the following words.

"The existence of a 5-second spontaneous fission activity observed in the reaction $^{50}\text{Ti} + ^{209}\text{Bi}$ and assigned to the isotope $^{257}_{105}$ in Ref. [85] could be confirmed. Our analysis showed that it consists of at least three components, SF of $^{257}_{105}$ (2n-channel), SF of $^{258}_{104}$ produced by EC-decay of $^{258}_{105}$ (1n-channel), and SF of $^{258}_{104}$ (1p-channel)."

XI. Conclusions Concerning Claims and Counterclaims on Discovery of Element 105

In the preceding section of this article we have summarized all of the publications which are relevant to the claims of first synthesis and positive identification of element 105. In this section we present our conclusions with respect to the correctness of these claims.

Our first conclusion is that the earliest study of a possible alpha-emitting isotope carried out in 1968 and described in an unpublished Dubna report [70] was too preliminary in character to be considered as a claim to discovery. The authors themselves described it as a preliminary study. In later articles [73] by the Soviet group in which they review their most important work on the identification of element 105, they state with respect to the 1968 work that "suggestions but not definite conclusions on $^{260}_{105}$ and $^{261}_{105}$ could be drawn from these experiments." Later studies of the alpha particles emitted by the isotopes with masses 260 and 261 do not confirm the alpha particle properties listed in this early reference.

The first papers which put forth claims for a convincing identification of element 105 were published in 1970 almost simultaneously by the Berkeley and Dubna groups. In both cases the experiments actually started in 1969 and were completed and written up for publication without knowledge of the results of the other laboratory.

Let us consider first the physical evidence presented by the Soviet group [71,73] as proof that the 1.8-second spontaneous fission activity they observed was due to $^{261}_{105}$ or $^{260}_{105}$. The physical evidence must be considered in its entirety because spontaneous fission is a general property of the heaviest nuclei and it is essential not only that the observed fission properties be reasonable for element 105, but also that all other possible candidates of lower atomic number be unequivocally ruled out. This is particularly important because the measured cross section of $5 \times 10^{-34} \text{ cm}^2$ for formation of the 1.8-second candidate activity is very small. The arguments given by Flerov and coworkers [73] are the following:

1) The compound nucleus from the $^{243}\text{Am} + ^{22}\text{Ne}$ reaction is $^{265}_{105}$. For the range of ^{22}Ne energies used, the evaporation of 4 or 5 neutrons from the excited compound nucleus would be expected, which would produce an isotope of mass number 261 or 260.

2) The observed half life is reasonable for decay by spontaneous fission or alpha decay.

3) The excitation function has the shape expected for the formation of a compound nucleus followed by evaporation of 4 or 5 neutrons. Comparison to excitation functions computed by semiempirical theory indicates that mass number 261 is the favored choice and that mass number 260 is also possible.

4) The observed cross section of $5 \times 10^{-34} \text{ cm}^2$ is somewhat low compared to the estimated value of $2 \times 10^{-33} \text{ cm}^2$ but this deficit could be explained by attributing the major decay to alpha decay, which was not looked for in the initial experiments.

5) Other possible assignments of the 1.8-second spontaneously fissioning activity can be ruled out after considering the two general classes of candidates. The first class is spontaneously fissioning isomers of nuclides of much lower Z formed by a non compound nucleus reaction in which only a few nucleons are transferred between the projectile and the target. The formation of 14-millisecond ^{242m}Am is an example of such a reaction. The Soviet group cite their experiments on the variation of the yield as a function of the collimation of the recoil products as strong evidence that their 1.8-second nuclide was made by a compound nucleus reaction and could not be a spontaneously fissioning isomer of a lower element.

The second class of spontaneous fission emitters which must be ruled out is the group of isotopes of elements 104, 103 and 102 which could be made by reactions of the $(^{22}\text{Ne}, \text{pxn})$, $(^{22}\text{Ne}, \alpha \text{xn})$, and $^{22}\text{Ne}(\alpha \text{pxn})$ type. The collimation grid experiments would not distinguish such reactions from the $(^{22}\text{Ne}, 4\text{n}-5\text{n})$ reaction which is postulated as the source of the 1.8-second activity.

Flerov and coworkers made a plausible case for eliminating any possible assignment to elements 104, 103, and 102 in the following ways.

In the case of element 104, isotopes which would have to be produced by reactions of the $^{243}\text{Am} (^{22}\text{Ne}, \text{pxn})$ type, there is considerable evidence from studies of targets of ^{238}U , ^{241}Am and ^{239}Pu that the ratio of cross sections for the competitive (Heavy Ion, pxn) reactions to the (Heavy Ion, xn) reactions is smaller than 0.02 for all $x \geq 1$, provided the bombarding energy corresponds to the

maximum of the excitation function for the (Heavy Ion, 4 or 5n) reaction. On the basis of this argument, such isotopes as $^{259}_{104}$ or $^{260}_{104}$ would have to be made in much smaller yield than was observed for the 1.8-second activity assigned to element 105. Similar arguments could be made to exclude isotopes of element 102 made by (Heavy Ion, α pn) reactions.

The same argument could not be used to exclude isotopes of element 103 because yields of products from (Heavy Ion α xn) reactions are higher in many cases than the (Heavy Ion, xn) reaction products. The Russian group working at the Joint Institute for Nuclear Studies investigated the properties of the isotopes $^{256,257}_{103}$ and established that the partial half life for spontaneous fission of both is $>10^5$ seconds. The decay properties of all other isotopes of 103 which could conceivably have been made were known to be inconsistent with assignment of the 1.8-second activity to them.

Thus, by elimination, the Soviet team argued that all possible alternate assignments for their 1.8-second spontaneously fissioning activity had been ruled out. Only assignment to element 105 remained. In their opinion, the mass assignment of the activity was most likely 261 with 260 a possibility.

In our opinion also these arguments are plausible and reasonable, but they still lack the force of convection that we believe is essential for such an important matter as the discovery of a new element. In Section VI entitled Criteria for Discovery of New Elements we cited a statement published by a group of American, German, and French nuclear chemists [23] on this subject. One of

their recommendations was that "the present understanding of production yields, excitation functions, angular distributions, and so forth, is not sufficient to allow measurements to establish with certainty that a nuclide with a new atomic number has been produced, although such data may be useful as supportive evidence."

We can also comment on the work carried out by a second group of 8 investigators at Dubna led by Dr. Druin [74] which was initiated as a search for the alpha decay branching of the 1.8-second activity found by Flerov et al [73]. We discussed these results in Section X and showed some of the data in Fig. 28. They concluded that had seen alpha particles at 9.1 and 8.90 MeV which decayed with a 1.4-second half-life. Unfortunately the quality of the alpha-particle data is poor. Druin et al. did not discuss their alpha-particle energy resolution but from the appearance of the data in Fig. 28 the half width must have been greater than 100 keV. Their spectrum in the 8.8 to 9.20 MeV range shows no close resemblance to the much better data published at the same time or earlier by Ghiorso, Nurmia, Eskola, Harris, and Eskola [79] and confirmed later by Bemis et al [82]. Druin et al. reported that they had applied the alpha correlation technique to their weak samples and found evidence that their 1.4-second alpha particles were weakly correlated in time to alpha particles in the 8.3 to 8.6 MeV range. For 62 start pulses in the 8.8 to 9.2 MeV range they observed 7 pulses in the 8.3 to 8.6 MeV range during a subsequent 140-second period. In their uncorrelated spectrum the alpha particles in the 8.4 to 8.6 MeV range had a 35-second half-life which they attributed to $^{256}_{103}$ or $^{257}_{103}$, because at

that time the half lives of these two isotopes were believed to be the same. Thus their alpha correlation data led them to state that they had "a basis for suggesting we are observing an alpha emitter with atomic number 105." This still left the mass assignment of the supposed 105 parent uncertain between 260 and 261. Later work at Berkeley [81] shows that $^{257}_{103}$ had the much shorter half-life of 0.7-seconds. If this had been known to Dr. Druin et al. it would have forced the conclusion that a genetic linkage could only be made to $^{260}_{105}$. Indeed, in the Druin article there is a note added in proof which cites the contemporaneously published work of Ghiorso et al. [79] on the isotope $^{260}_{105}$ and comments that these results (i.e. Ghiorso's) "are in good agreement with the data of Flerov et al. [71,72] and with the present work." However, this would have made it necessary for Flerov and his coworkers to change their favored mass assignment from 261 to 260 and, as far as we are aware, they did not do this in subsequent publications. Our judgment is that the work reported in Ref. 74 does not strengthen the case for the discovery of element 105 at Dubna.

We turn now to a statement on the chemical properties measured by Zvara et al. [75] and used as evidence for the element 105 assignment.

In Section X under the heading, "Soviet Publications on the Chemical identification of Element 105" there is a description of experiments done by the thermochromatographic technique. The main results were presented in Figs. 29 and 30. These experiments were designed under the assumption that element 105 would be the chemical homolog of niobium and tantalum. The earliest work, performed in a

chlorination system, was published only as an internal JINR report [75] with a short synopsis given later in a conference proceedings [76]. This work appeared in 1970 in time to be considered as supporting evidence for the discovery of element 105 at Dubna, but it was never published in a standard scientific journal.

The results of this chlorination study are not definite in establishing the contention that the distribution of fission tracks (attributed to element 105) is that expected for a chemical homologue of niobium and tantalum. In Fig. 29 there is no peak in the fission track distribution. In fact, the tracks occur in the ranges for a number of the calibrating elements, and none of them appears in the range of niobium. From these results it is not possible to conclude that the element represented by these fission tracks is acting chromatographically like niobium and tantalum as distinct from other elements. This is in marked contrast to the element 104 results [48] of 1971 where the fission tracks attributed to element 104 were concentrated in the expected position directly under the hafnium peak.

Some years later (in 1975) Zvara and his group published experiments [77] done in a bromination system, which offered several experimental advantages over the chlorination system. This publication occurred years after the physics papers on the discovery of element 105 had appeared from Berkeley and Dubna; so, strictly speaking, this work cannot be regarded as helping to establish priority of the work of the Soviet team. It is interesting, nonetheless, briefly to analyze the data presented in this paper (see Fig. 30). It can be seen in the figure that the fission tracks

attributed to element 105 fall in a broad range occupied by ^{169}Hf and ^{170}Hf , but not in the region defined for ^{90}Nb . It thus appears that, if these fission tracks are from element 105, elements 104 and 105 deposit in the same range in the thermochromatographic column and therefore, cannot be distinguished on the basis of these low-resolution gas chromatographic experiments.

A more complete discussion of the Dubna 105 chemical experiments is given in Ref. 20.

At this point we examine the claims of Ghiorso and his coworkers concerning the discovery of element 105, based on the published articles [79,80] which we summarized in the part of Section X labeled Lawrence Berkeley Laboratory Publications 1970-1971.

In their first paper, published in 1970, they presented definitive evidence for a 1.6-second alpha emitter $^{260}_{105}$, produced by the reaction of ^{15}N with ^{249}Cf . The nuclear reaction is suitable and the excitation function (see Fig. 33) shows a maximum at the expected ^{15}N beam energy. The alpha particles assigned to the isotope have a unique pattern and the energies are in the appropriate range for element 105. The isolation of daughter alpha activity by the alpha recoil technique and the observation of an alpha spectrum for the recoil activity with the proper energies and half life for $^{256}_{103}\text{Lr}$ is very definite genetic information for establishment of the atomic and mass number of the parent. For identification of an unknown nuclide, the use of alpha particle information instead of the less specific spontaneous fission tracks has a great intrinsic advantage.

In their second paper Ghiorso, Nurmia, Eskola and Eskola [80] presented information of comparable quality to identify a 1.8-second $^{261}_{105}$ decaying by the emission of 8.93-MeV alpha particles to produce a 0.6-second $^{257}_{\text{Lr}}$ daughter. The genetic relationship of the $^{261}_{105}$ isotope to its $^{257}_{\text{Lr}}$ daughter was established by alpha recoil and alpha particle time-correlation experiments. Similarly, the isotope $^{262}_{105}$ produced by the $^{249}_{\text{Bk}}(^{18}_0,5n)^{262}_{105}$ reaction was identified by its parent-daughter relationship to 4.2 second $^{258}_{\text{Lr}}$.

For all three isotopes the original claims and assignments of properties have stood the test of time, except for minor refinements on the alpha particle energies, intensities and half life. It is significant that a restudy [82] by an independent laboratory completely confirmed the results. The ORNL study of Bemis et al. [82] added additional X-ray evidence for the confirmation of the identity of the $^{260}_{105}$ isotope. The properties of $^{262}_{105}$ published by Ghiorso et al. were completely confirmed in a restudy [83] by scientists from ORNL and the Lawrence Livermore National Laboratory.

We conclude that the evidence presented by Ghiorso, Nurmia, Eskola, Harris, and Eskola [79,80] was definite and convincing and that it fully meets the requirements of both statements [22,23] on Criteria for Discovery of New Elements which were cited in Section VI. In particular, the proof of the atomic number by genetic linkage to known isotopes of the daughter element 103 is very strong. Their work was performed and published at the same time as the Soviet work

on spontaneous fission properties [71-75]. We recommend that Ghiorso, Nurmia, Eskola, Harris, and Eskola be credited with the discovery of element 105 and be given the traditional right to name the element. We suggest that their choice of the name Hahnium, with symbol Ha, be adopted.

XII. Summary of Properties of Isotopes of Element 105

A summary of published information on all isotopes of element 105 is presented for reference in Table 6.

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Table 6. Published Information on Isotopes of Element 105

Isotope	Half Life	Ref.	Decay Mode	Ref.	α -Particle Energies (MeV)	Ref.	Method of Synthesis	Ref.
255	<u>1.5 sec</u>	[85],67]	SF(20%)				$^{207}\text{Pb}(^{51}\text{V}, 3n)$ $^{206}\text{Pb}(^{51}\text{V}, 2n)$	[85,67] [85]
257	<u>1.4\pm0.6 sec</u>	[86]	α (83 \pm 10%) SF(17% upper limit)	[86]	9.160(30%) <u>9.071(30%)</u> 8.970(40%)	[86]	$^{209}\text{Bi}(^{50}\text{Ti}, 2n)$	[86]
257	5.0 \pm 1.7 sec	[85]	SF(20%)	[85]			$^{208}\text{Pb}(^{51}\text{V}, 2n)$ $^{205}\text{Tl}(^{54}\text{Cr}, 2n)$ $^{209}\text{Bi}(^{50}\text{Ti}, 2n)$	[85] [85] [85]
258	<u>4.4\pm0.9 sec</u>	[86]	α (67%) EC(33%)	[86]	9.299(8%) <u>9.172(59%)</u> <u>9.078(28%)</u> <u>9.008(5%)</u>	[86]	$^{209}\text{Bi}(^{50}\text{Ti}, 1n)$	[86]
260	1.4 sec	[74]	α	[74]	8.90, 9.1	[74]	$^{243}\text{Am}(^{22}\text{Ne}, 5n)$	[74]
260	1.6 \pm 0.3 sec	[79]	α (>80%) SF(<20%)	[79] [79]	9.06(55%) 9.10(25%) 9.14(20%)	[79]	$^{249}\text{Cf}(^{15}\text{N}, 4n)$	[79]
260	<u>1.52\pm0.13 sec</u>	[82]	α (90.4%) SF(9.6%)	[82] [82]	9.041(48%) <u>9.074(25%)</u> <u>9.12(17%)</u>	[82]	$^{249}\text{Cf}(^{15}\text{N}, 4n)$	[82]
261	1.8 \pm 0.6 sec	[73]	SF	[73]			$^{243}\text{Am}(^{22}\text{Ne}, 4n)$	[73]
261	<u>1.8\pm0.6 sec</u>	[80]	α SF(<50%)	[80]	<u>8.93</u>	[80]	$^{250}\text{Cf}(^{15}\text{N}, 4n)$ $^{249}\text{Bk}(^{16}\text{O}, 4n)$	[80] [80]
262	40.0 \pm 10 sec	[80]	α	[80]	8.45, 8.66	[80]	$^{249}\text{Bk}(^{18}\text{O}, 5n)$	[80]
262	<u>34.1\pm4.6 sec</u>	[83]	α (22 \pm 6%) SF(78 \pm 6%)	[83]	8.450(75%) <u>8.530(16%)</u> 8.670(9%)	[83]	$^{249}\text{Bk}(^{18}\text{O}, 5n)$	[83, 84]

Best current values of half life and α energies are underlined.

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FIGURE CAPTIONS

- Figure 1. Alpha decay energies, Q_α , as function of mass number for heaviest elements. Q_α is the total ground state to ground state decay energy including alpha particle and recoil nucleus energies. The predicted region of Q_α values for elements 104 and 105 is shown in the upper right hand corner of the diagram. This figure is based on evaluated data published by Wapstra, et al., (Nucl. Phys. A432, 1-362 (1985)).
- Figure 2. Experimental values of half-life versus alpha energy (Q_α) for even-even nuclei. The figure is restricted to Q_α values above 5.5 MeV. For a nucleus with significant branching decay by spontaneous fission only the partial alpha half-lives shown. The true half-life is shorter in such a case. The curve for element 104 is extrapolated from the positions of the curves for lower Z elements.
- Figure 3. Comparison between experimental half-lives and partial half-lives calculated or estimated by Randrup, et al [12a] for spontaneous fission of even-even nuclei. The insert at the lower left shows a comparison between calculated and experimental half-lives for fission isomers. This figure is taken from Ref. [62].

Figure 4. Spontaneous fission half-life hindrance factors for odd-Z and odd-N nuclei, as obtained by comparing their empirical half-lives with values obtained by interpolation among half-lives of adjacent doubly even nuclei. The calculated hindrance factors for $N = 157$ are displayed as dots for comparison. The notation in brackets refers to the asymptotic quantum numbers of the Nilsson model description for odd-N nuclei. From Randrup, et al [12b].

Figure 5: Experimental Γ_n/Γ_f values for four even-Z heavy elements plotted versus the neutron number of the compound nucleus during in the neutron evaporation cascade. The circles, triangles and squares represent data for Cf, Fm, and No, respectively. The broken lines represent the empirical relationship quoted in the text of section III. See Sikkeland, Ghiorso, and Nurmia [15] for more complete explanation.

Figure 6: Probe inserted in the 300 cm heavy-ion cyclotron of the Joint Institute for Nuclear Research, Dubna, for use in preparation of element 104 and detecting spontaneous fission of product nuclei. The ^{22}Ne beam indicated by the arrow passed through a vacuum window foil and energy degrading foils before traversing the foil supporting the ^{242}Pu target. Product nuclei were ejected from the target and impinged on the moving nickel conveyor belt which transported them past glass detectors. If a product nucleus underwent spontaneous fission as it passed one of these glass detectors one of the fission fragments would strike the detector and create damage that would reveal its position later when the glass was suitably etched with chemical reagents. From Ref. [24].

Figure 7: The decay of the spontaneously fissioning product formed in the interaction of ^{22}Ne with ^{242}Pu as determined by the apparatus shown in Fig. 6. From Ref. [24].

Figure 8: The energy dependence of the cross sections for the formation of spontaneously fissioning products in the reaction $^{22}\text{Ne} + ^{242}\text{Pu}$. The filled rectangles are the experimental values for synthesis of an isotope with a spontaneous fission half-life of 0.3 sec. The open triangle are the experimental values for the synthesis of $^{256}_{102}$. The open circles are the experimental points for the synthesis of an isomer of ^{242}Am , for which the right hand cross section scale applies. From Ref. [24].

- Figure 9. Schematic of the type of gas chromatographic apparatus used at Dubna [28-31] for chemical studies during the period 1966 - 1969.
- Figure 10. Distribution of fission tracks along the mica detectors compatible with a half life of 0.3 seconds for "kurchatovium". Crosses mark positions where both fission fragments were recorded. From Zvara et al [31].
- Figure 11. Distribution of fission tracks along the mica detectors implying a half life of "somewhat less than one second" for "kurchatovium". The crosses identify events in which both fission fragments were recorded. From Zvara et al [33].
- Figure 12. Heat of adsorption and heat of sublimation correlation for "kurchatovium". The circles represent data for Nb, Ta, Hf, In, Th, Na, Sc, and Am. Points with error bars mark the positions for Pa, U, and Pu. The point for "kurchatovium" is placed on the curve using a heat of adsorption calculated on the basis of a half life of 0.5 sec. From Zvara et al [33].
- Figure 13. Schematic of the chromatographic apparatus for studying volatility of chlorides of element 102 vs. Cf, Cm, Hf and Pu. Element 102 was detected by its Fm daughter rather than directly. The distributions of isotopes along the column and in the detector are shown. (The column was evidently analyzed in two sections.) Much more 102 is deposited in the first part of the column than in the second part, as expected for a heavy actinide. Surprisingly plutonium chloride is sufficiently volatile to deposit near hafnium chlo-

ride raising a potential source of error in interpretation for the 104 experiments. From Chuburkov et al [35].

Figure 14: A series of alpha-particle spectra produced by bombardment of a ^{249}Cf target with 71 MeV ^{12}C ions. The top spectrum is the sum of the individual spectra from the four solid-state detectors taken at 3 second intervals. The 8.60-MeV peak is due to ^{258}Lr ; the peaks above that energy belong to $^{257}\text{104}$ with exception of the one at 8.87 MeV which comes from $^{211\text{m}}\text{Po}$ produced from a small lead impurity in the target. From Ghiorso, et al, [37].

Figure 15: Excitation curves (i.e., yield of a nuclear product versus the energy of the bombarding ^{12}C ions) for several activities produced by bombardment of ^{249}Cf with ^{12}C . Note the high yield of the nobelium isotopes produced in (^{12}C , αxn) reactions. From Ghiorso et al [37].

Figure 16: Spectra from the mother-daughter experiment conducted by Ghiorso et al [37] which demonstrates the genetic relationship between $^{257}\text{104}$ and ^{253}No . The left hand "mother" spectra 1 through 4 are successive 3 second measurements after deposition of the element 104 samples. The bottom spectrum is the sum of 2,3, and 4. Peaks at 8.70, 8.78, 8.95 and 9.00 MeV are assigned to $^{257}\text{104}$. The right hand "daughter" spectra are 200 second measurements of element 102 daughter activity which recoiled out of the mother samples. The energy and half life of the peak near 8.0 MeV agrees with the known properties of ^{253}No .

Figure 17: Experimental apparatus used by Oganessian et al [46] and Flerov et al [73] in the external beam of the JINR/Dubna cyclotron for the collection and measurement of short-lived, spontaneously-fissioning products recoiling out of heavy element targets placed in the path of a beam of heavy ions. The speed of the endless nickel belt could be adjusted to suit the half life expected for the product nuclei. The expanded view of the collimator shows how the grid of holes serves to collimate the products recoiling out of the target.

Figure 18: Time distribution of the fission fragments from reaction products of ^{242}Pu bombarded with 113 MeV ^{22}Ne ions. A 15-ms component is assigned to $^{242\text{m}}\text{Am}$ and a 100 ± 50 -ms component is assigned to $^{260}104$ by Oganessian et al [46].

Figure 19. (a) Schematic of the type of gas chromatographic apparatus used at Dubna for chemical studies of element 104 in 1971. See Ref. [48].

(b) Temperature gradient in the column.

(c) Deposition position of $^{259}104$ (black circles) compared to deposition position of Sc and heavy actinides (white circles) and Hf.

Figure 20: Alpha-spectrum of products from the reaction of ^{249}Cf with 73 MeV ^{12}C ions as measured by Bemis and co-workers [56]. The resemblance to the spectrum obtained by Ghiorso and co-workers [37] which appears in Fig. 14 is close. The most important feature is the reproduction of the peaks in the 8.70 to 9.00 region, which are assigned to $^{257}104$.

- Figure 21: Characteristic K-series x-ray spectra expected for elements with $Z = 100$ through 103. The experimental photon spectrum coincident with α particles in the energy range 8.5–9.1 MeV is shown in histogram fashion under the curve labeled 102 and forms the basis of a conclusive identification of element 104. From Bemis [56].
- Figure 22: Time distribution of the SF tracks from products of the reaction of $^{249}\text{Bk} + 82 \text{ MeV } ^{15}\text{N}$ ions as measured by Druin and coworkers [61]. The 76 ± 8 -ms component is attributed to $^{260}_{104}$. The cross section for its formation was determined as 8 ± 2 nanobarns.
- Figure 23: Decay curves showing ~ 20 -ms SF activity, assigned to $^{260}_{104}$, and produced by the following reactions:
 (a) $^{248}\text{Cm}(^{16}\text{O}, 4n)^{260}_{104}$, (b) $^{249}\text{Bk}(^{15}\text{N}, 4n)^{260}_{104}$, and
 (c) $^{249}\text{Cf}(^{18}\text{O}, \alpha 3n)^{260}_{104}$. The dashed lines show the amounts of ^{256}Fm backgrounds and short lived components determined in the fits to the experimental points; the solid curves show the sums of these components. From Somerville et al [62]. Section (b) is in strong disagreement with the results presented by Druin et al. [61] in Fig. 22. In particular, there is no evidence for a 76-ms component.
- Figure 24: Diagram of the target chamber and the gas stream collector system used by the Russian team at the Joint Institute of Nuclear Research, Dubna, [70] in a first search for isotopes of element 105. Numbers refer to (1) target, (2) reaction chamber, (3) aluminum foil, (4) Faraday cup, (5) capillary, (6) annular detector and (7) wheel catcher.

- Figure 25: Alpha particle spectrum obtained in the experiments of Flerov et al [70] in the reaction of ^{243}Am with 123-MeV ^{22}Ne ions.
- Figure 26: Time distribution of tracks from spontaneously fissioning nuclides produced in the reaction of 114 MeV ^{22}Ne ions with ^{243}Am . From Flerov et al [73].
- Figure 27: Dependence of the formation cross section of the spontaneously fissioning isotope with a half life of 2 seconds on the energy of the ^{22}Ne ions. The dashed curve shows the calculated excitation functions for the reaction $^{243}\text{Am}(^{22}\text{Ne},4n)^{261}_{105}$ and the corresponding ($^{22}\text{Ne},5n$) reaction leading to $^{260}_{105}$. The dot-dashed curve shows the calculated dependence of the fission cross section (righthand scale) on the ^{22}Ne energy. The experimental data are shown by the open circles. From Flerov et al [73].
- Figure 28: Alpha spectrum obtained by Druin et al [74] in a study of products of $^{243}\text{Am} + ^{22}\text{Ne}$ (120 MeV).
- Figure 29: Distribution of registered of fission tracks (black squares) in the temperature gradient portion of the glass tube in three separate experiments (a,b,c in the figure correspond to II, III, IV in Table 5). The position of deposition of known elements is also shown. From Zvara et al [75].

Figure 30: Locations of fission tracks (circles) attributed to element 105, in the gas chromatographic column compared to zones found for ^{90}Nb ($t_{1/2} = 14.7$ hr), ^{170}Hf ($t_{1/2} = 12.2$ hr), and ^{169}Hf ($t_{1/2} = 3.3$ min) using bromine chemistry. The "calc" curve (marked 105) compensates for the relatively shorter half life of element 105. Adapted from Fig. 1 in Ref. [77] (Zvara, Belov, Domanov and Shalayevisky).

Figure 31: A series of alpha-particle spectra of products of nuclear reactions of ^{15}N with ^{249}Cf . Spectra 1 through 5 represent data taken at 1 second intervals after deposition of reaction products on the moving wheel collector. The top spectrum is the sum of spectra 1-5. The peaks in the 9.06 to 9.14 MeV region are those assigned to $^{260}\text{105}$. The alpha peak at 9.00 MeV is that of ^{216}Fr , the daughter of 23-millisecond ^{220}Ac produced from a trace impurity of lead in the target. Most of the other peaks in the 6.04 ~ 8.1 MeV range also come from reactions with lead. Most of the ^{211}Po derives from electron-capture of 7.2-hour ^{211}At . From Ref. [79] (Ghiorso et al.)

Figure 32: A series of alpha-particle spectra from the same $^{249}\text{Cf} + ^{15}\text{N}$ bombardment on those in Fig. 31 but recorded by the silicon detectors in the off-wheel position, when the alpha-recoil daughter activity was under measurement. The spectrum of a known sample of ^{256}Lr produced by the reaction $^{249}\text{Cf}(^{11}\text{B}, 4n)^{256}\text{Lr}$ is shown in the inset. A similar group

of alpha particles in spectra 1-5 and the sum spectrum are assigned to ^{256}Lr . The spectra 1-5 represent 12.5-second time subgroups. From them a 30 ± 10 second half life is derived in agreement with the known 25.9-second ^{256}Lr half life. From Ref. [79].

Figure 33: Excitation curves for element 103 and 105 activities in bombardments of ^{249}Cf with ^{15}N ions [79].

Figure 34: Alpha spectra of recoil products from bombardment of ^{250}Cf with ^{15}N ions [80]. Spectra 1 through 7 represent 1.5-second periods after deposition of the sample. The step movement of the wheel placed the deposited sample in front of 7 detector stations in turn. The sum spectrum is shown on top.

Figure 35: Alpha spectra of recoil products from bombardment of a ^{249}Bk target with ^{18}O ions [80]. Spectra 1 through 7 represent data taken in seven 40-second time intervals after collection of the sample. The top curve is the sum of these spectra.

Figure 36: Time distribution of alpha-particle events in the energy range 9.00-9.20 MeV from reaction products of $^{249}\text{Cf} + 86.0 \text{ MeV } ^{15}\text{N}$ ions. This activity is assigned to $^{260}_{105}$. See Bemis et al [82].

Figure 37: Prompt coincidence photon spectrum in the energy range 10-35 keV associated with α events in the energy range 9.00-9.20 MeV. The solid lines represent the pattern expected for the L x-rays of element 103. From Bemis et al [82].

Figure 38: Alpha-particle energy spectrum (left section) for activities in time correlation with a preceding alpha decay of $^{260}_{105}$ in the energy range 9.00–9.20 MeV. The time distribution (since emission of the preceding alpha particle) of these events in the 8.27–8.56 MeV energy range is shown in the right hand section. The derived half life agrees with the published value of 25.9 ± 1.7 seconds for $^{256}_{\text{Lr}}$. From Bemis et al [82].

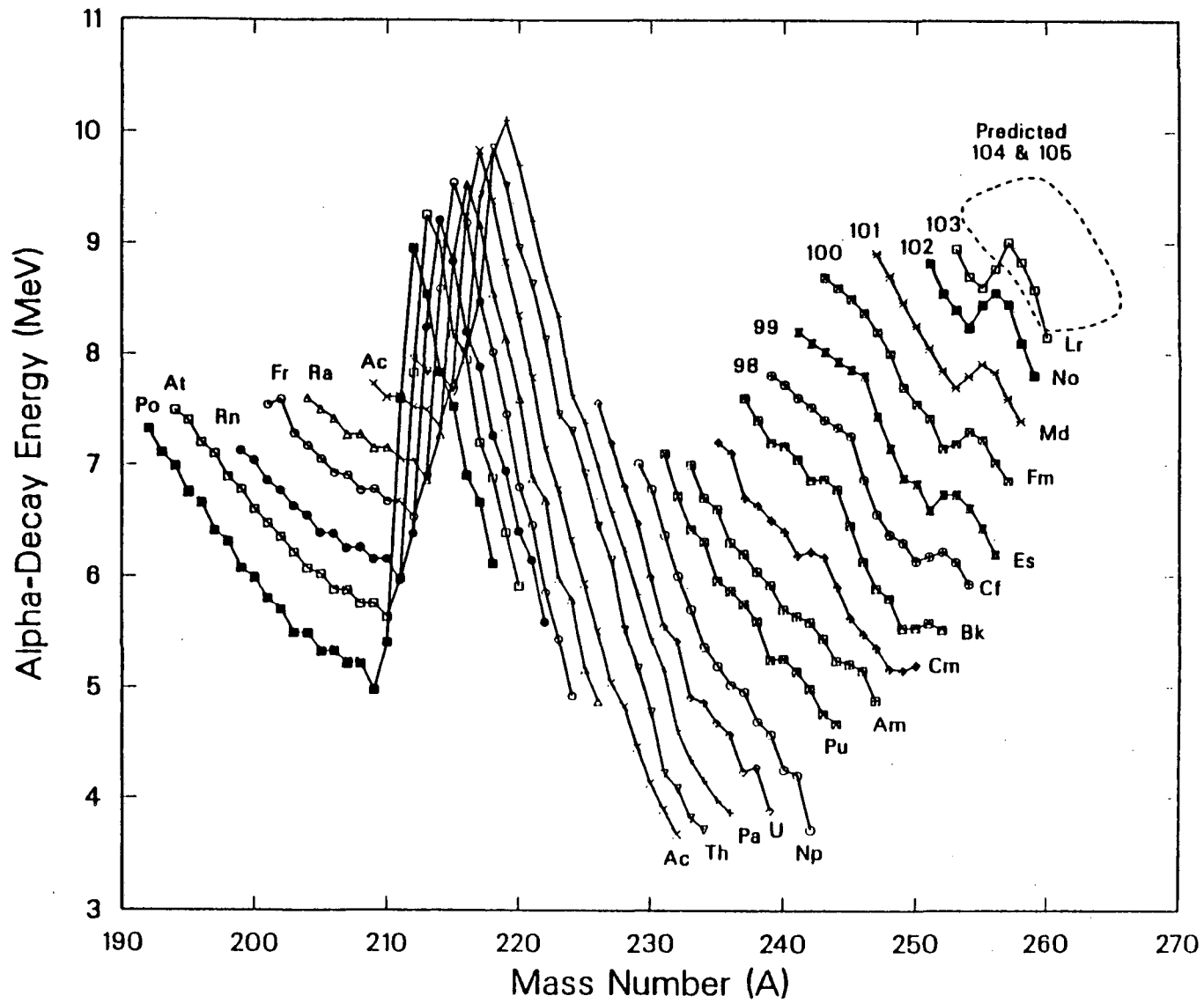
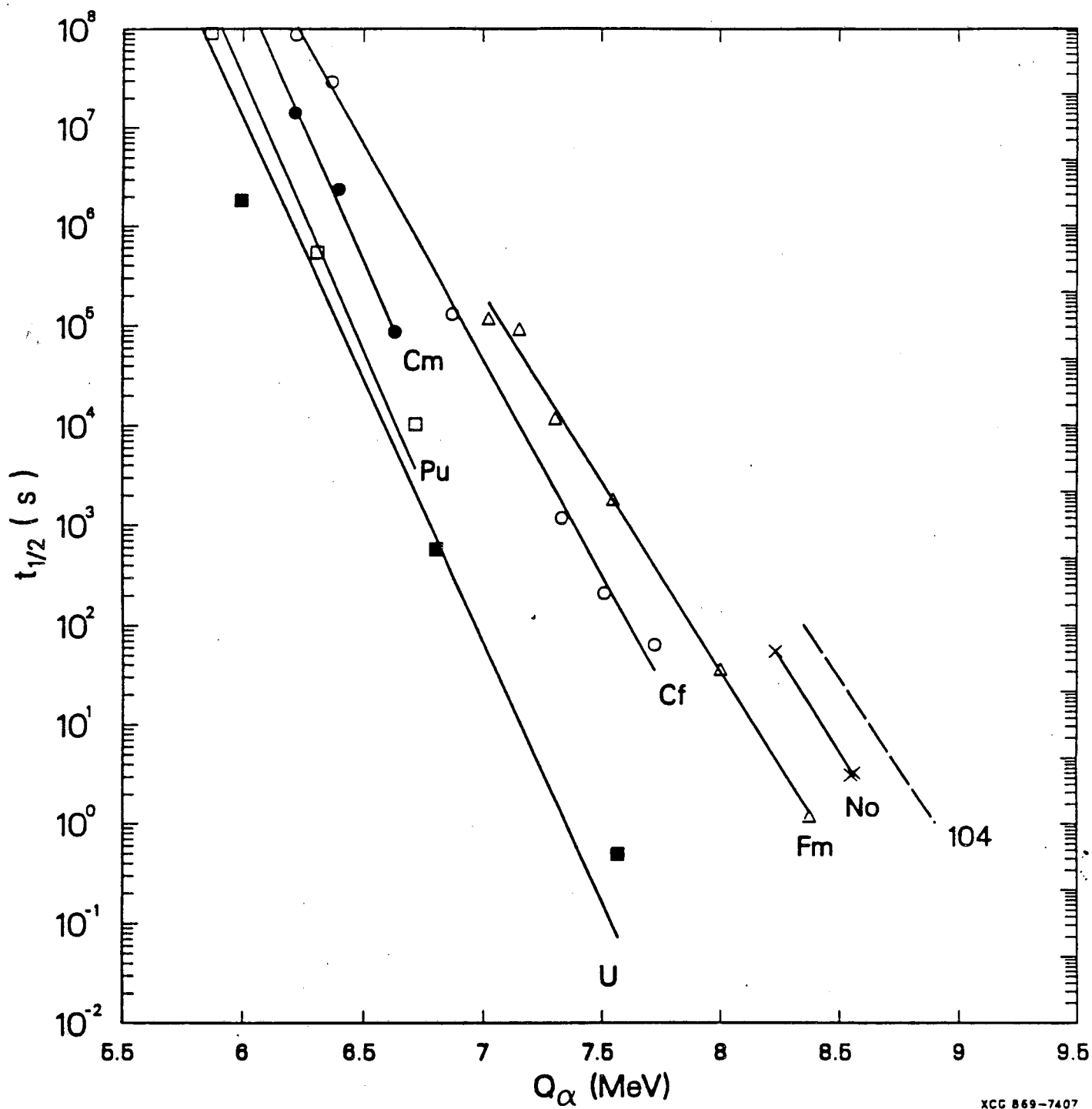


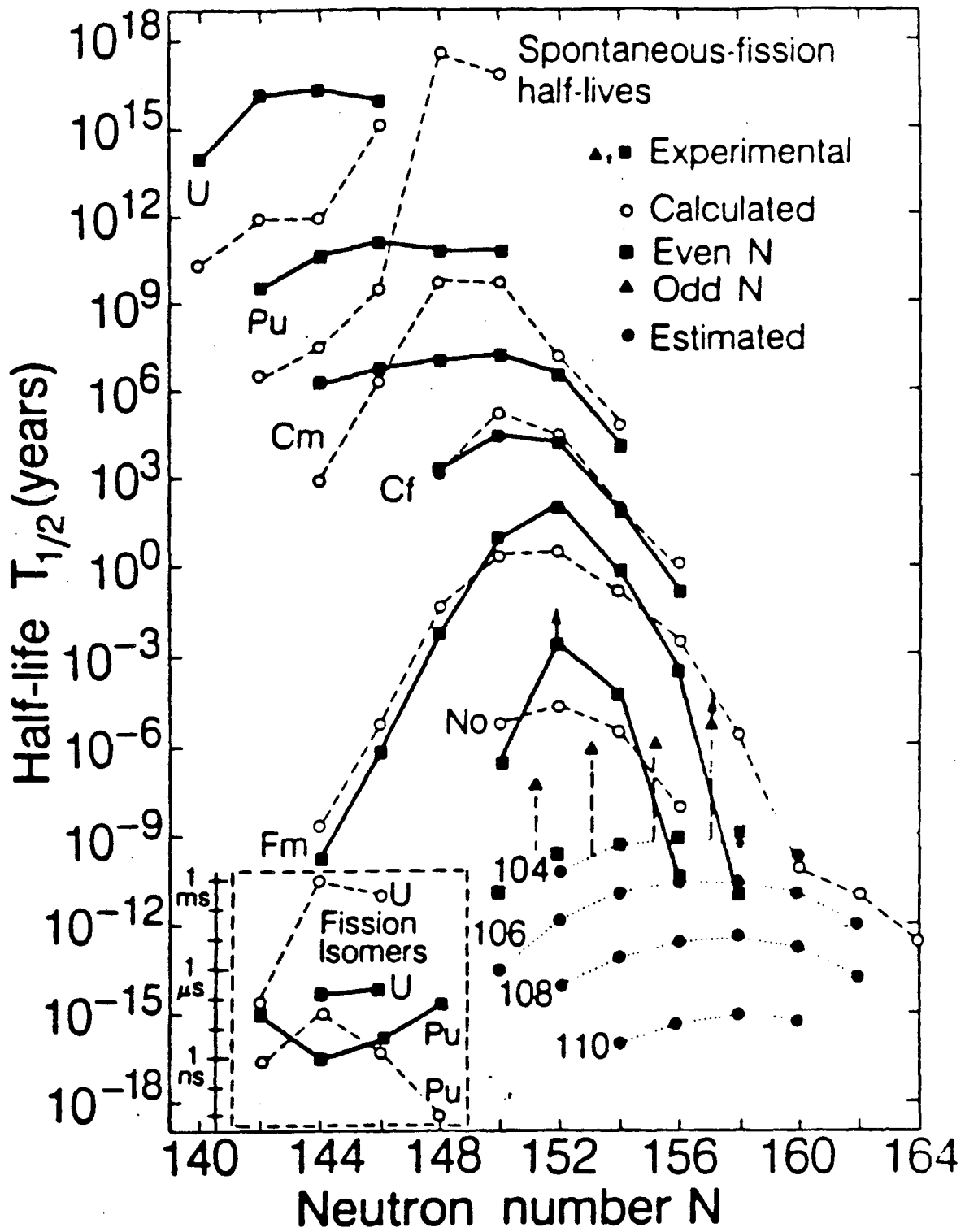
FIGURE 1

XCG 862 -7104



XCG 869-7407

FIGURE 2



XBL 862-580

FIGURE 3

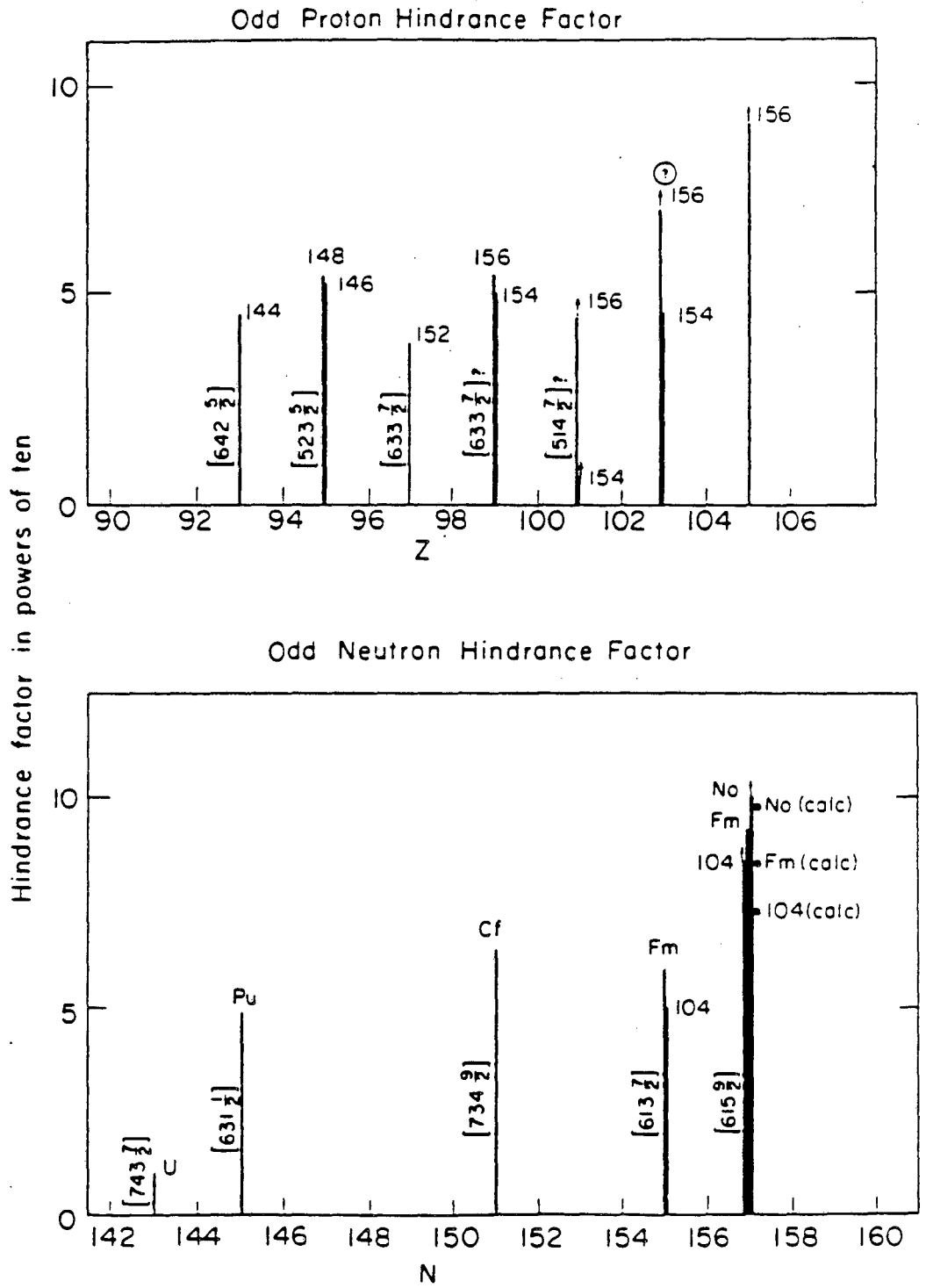
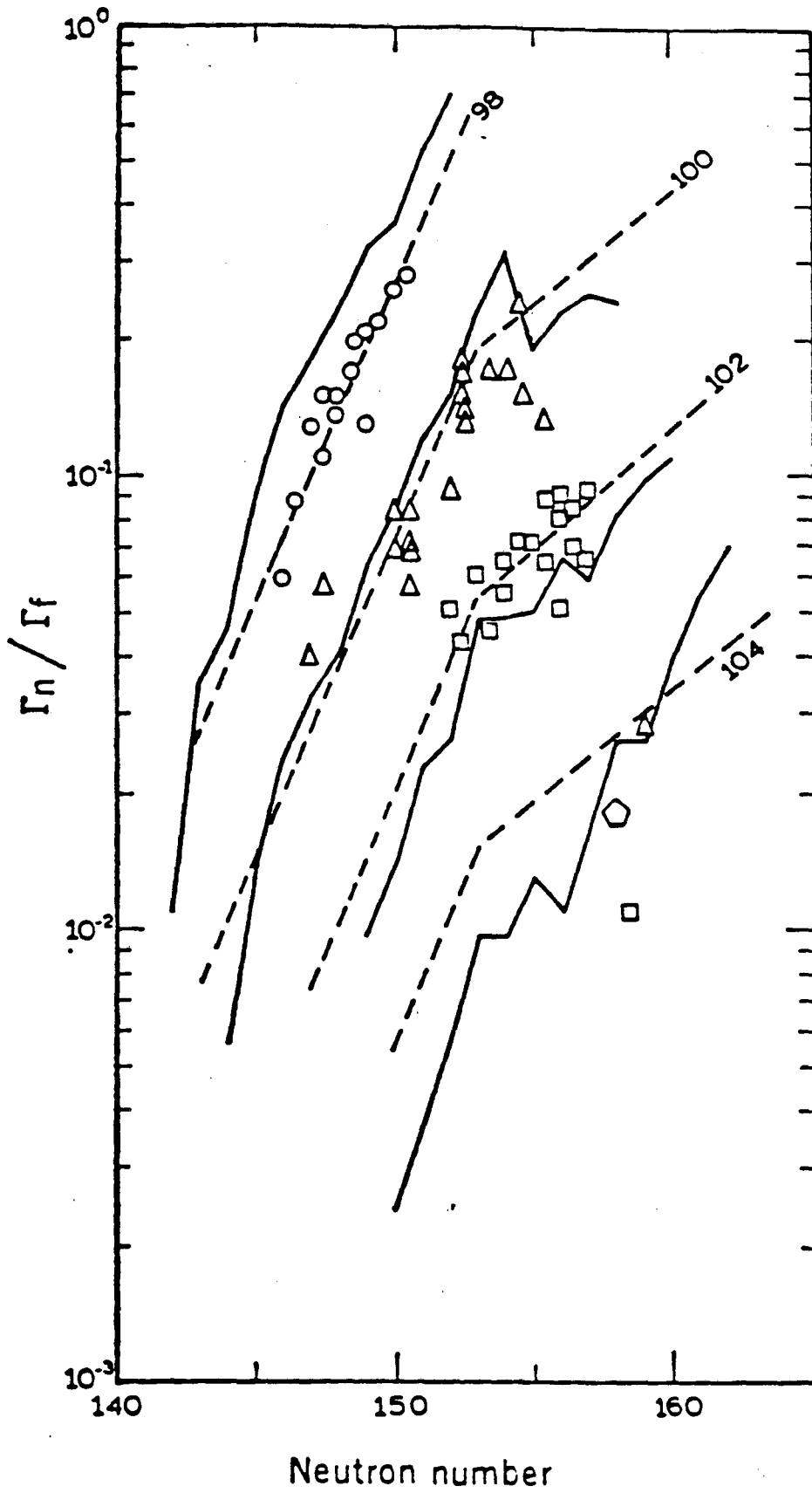


FIGURE 4



XBL 862-581

FIGURE 5

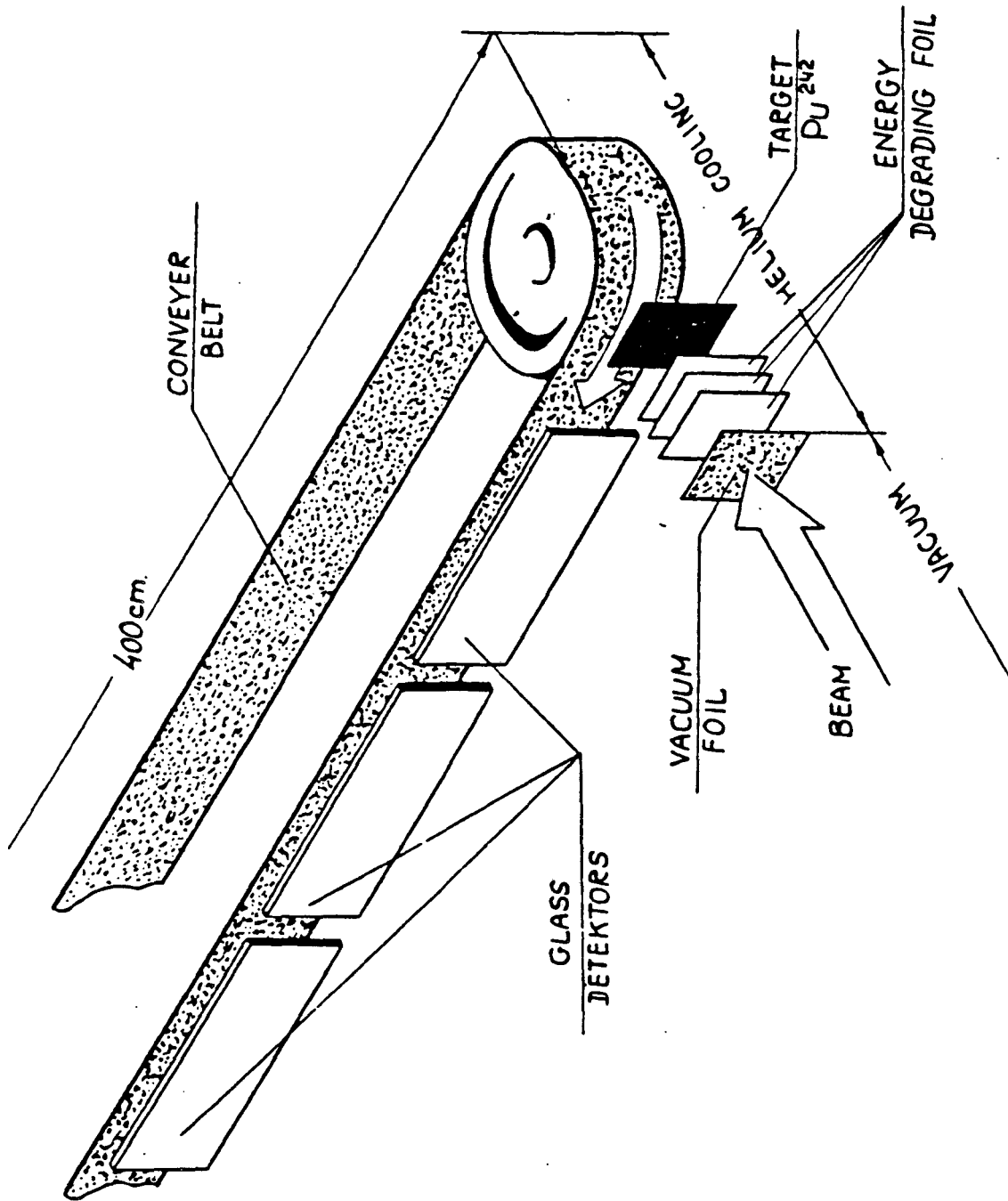
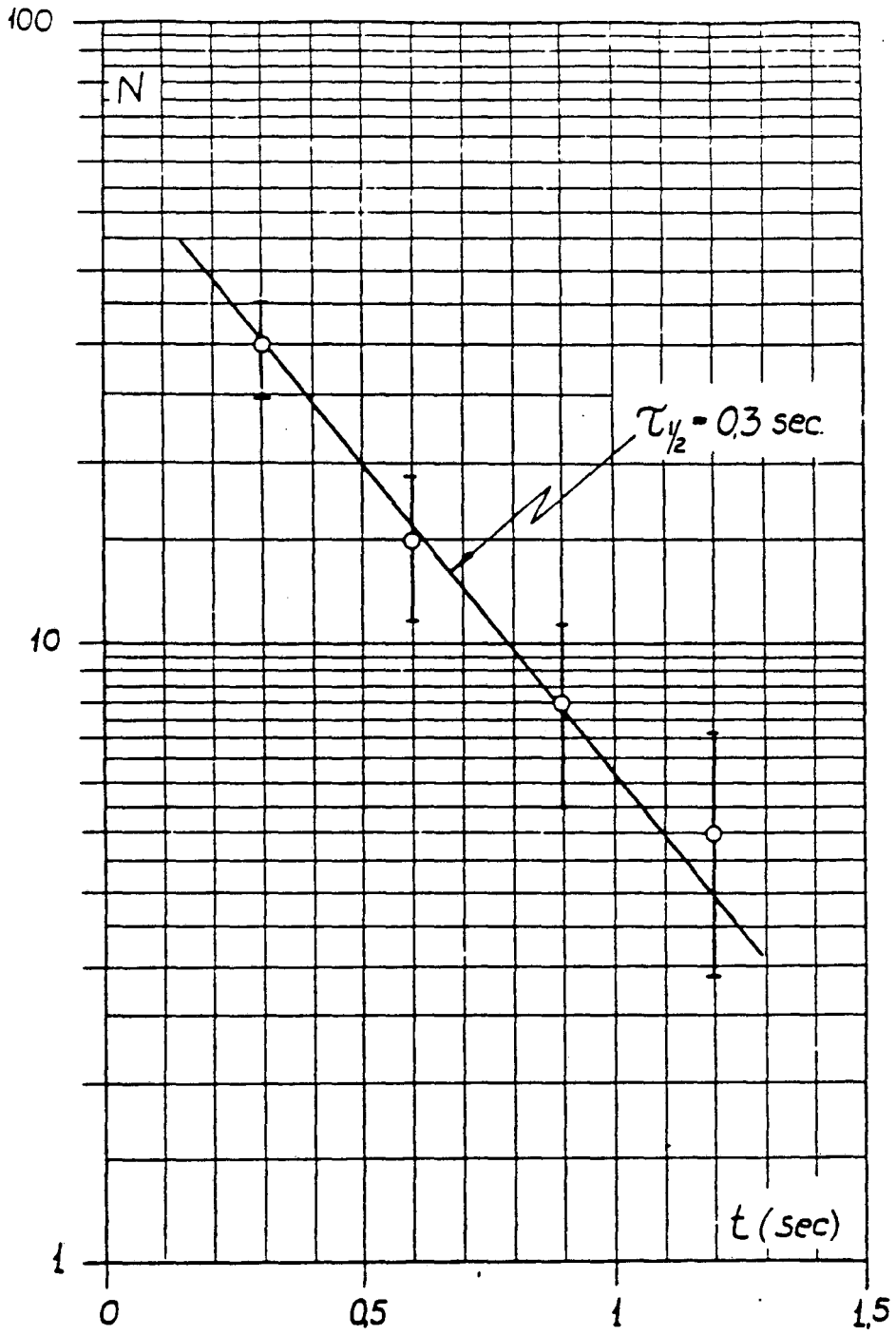
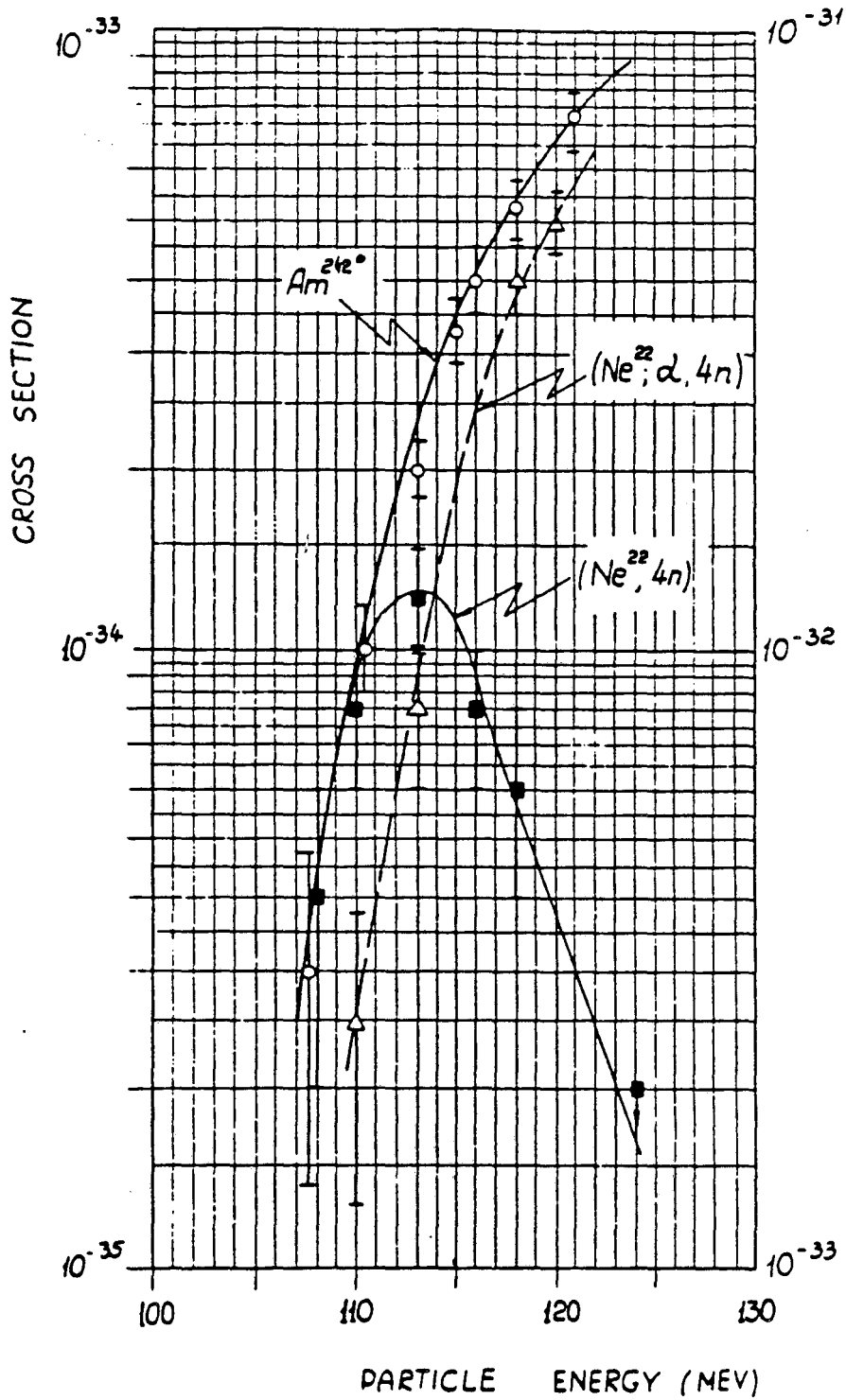


FIGURE 6



XBL 862-472

FIGURE 7



XBL 862-473

FIGURE 8

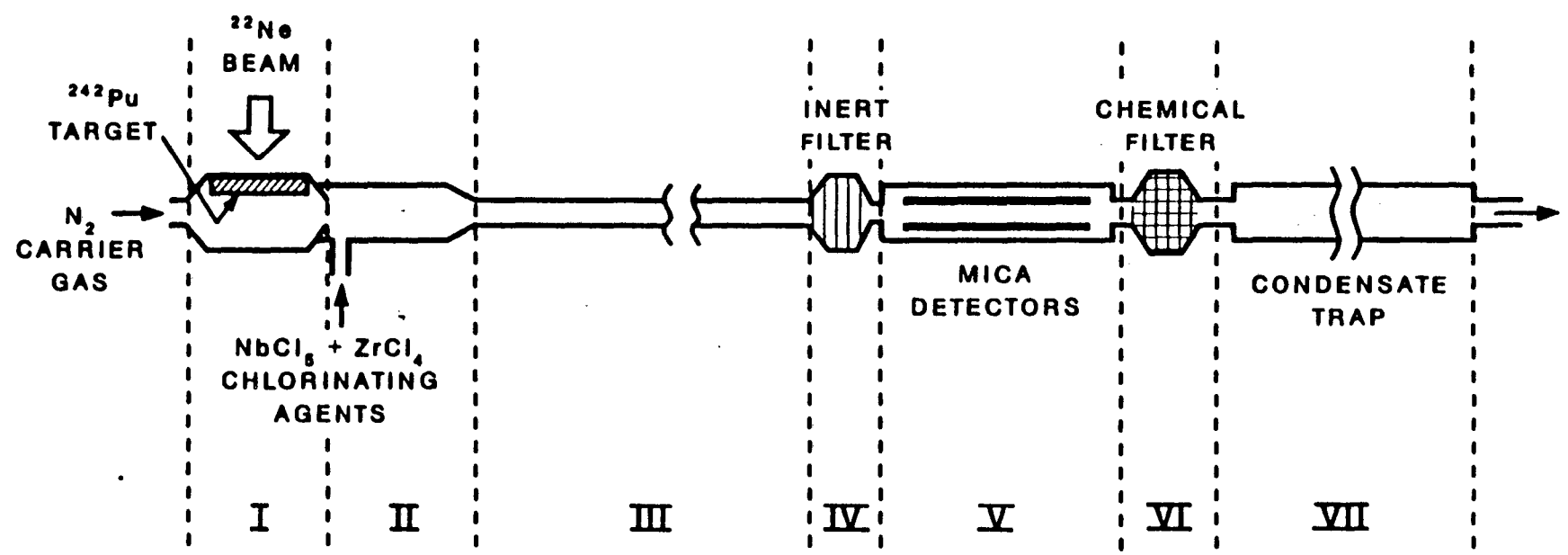


FIGURE 9

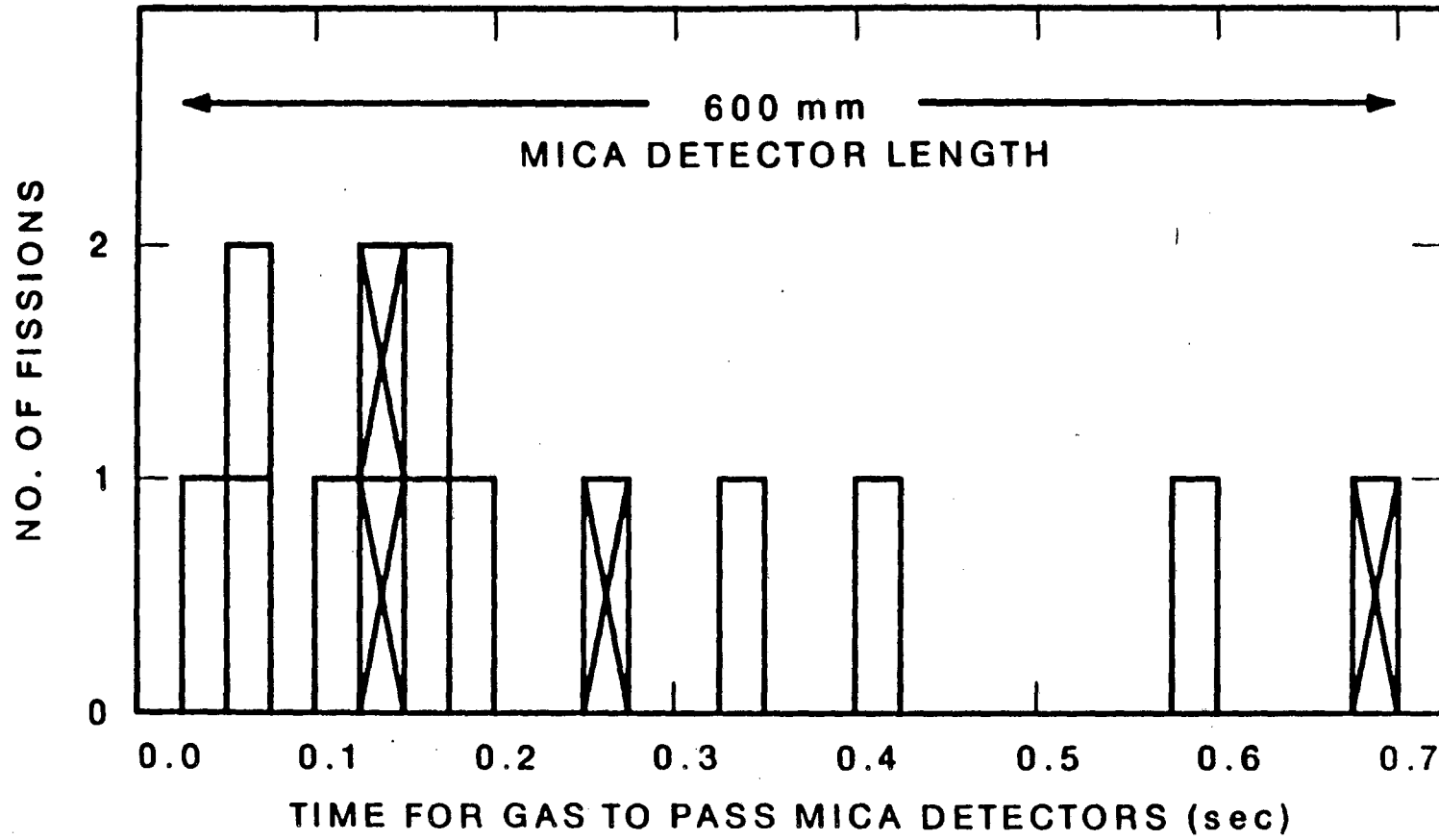


FIGURE 10

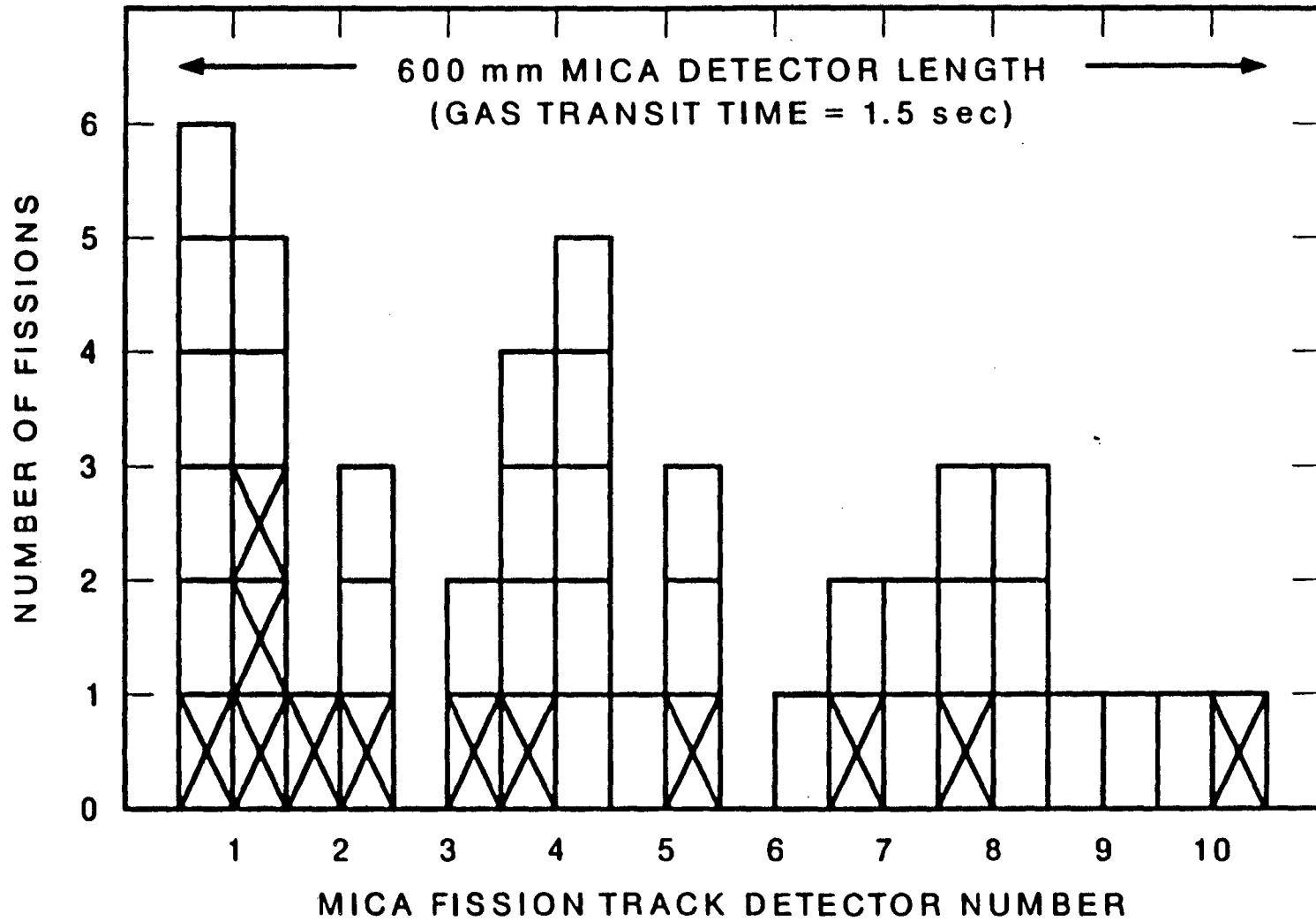


FIGURE 11

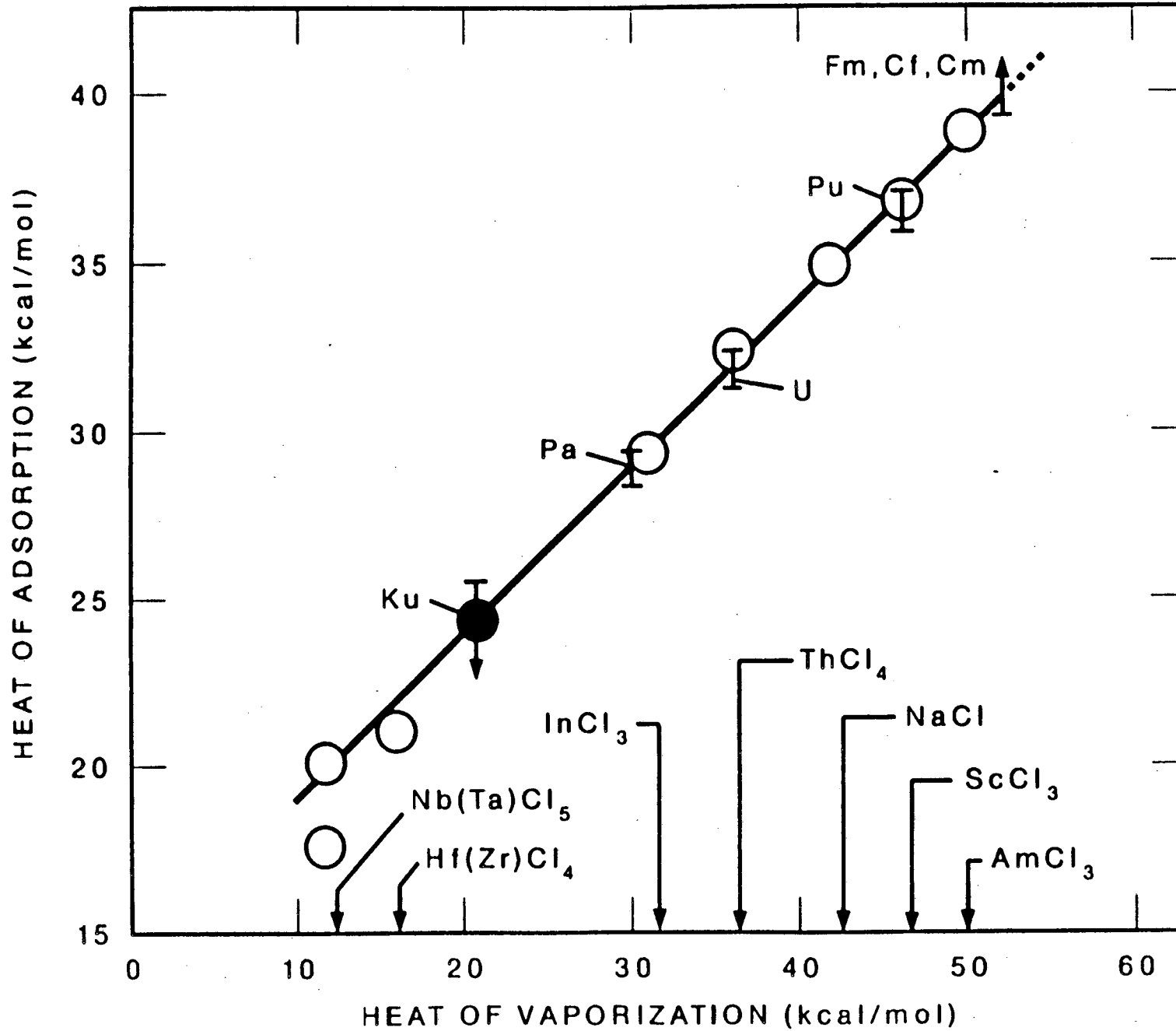


FIGURE 17

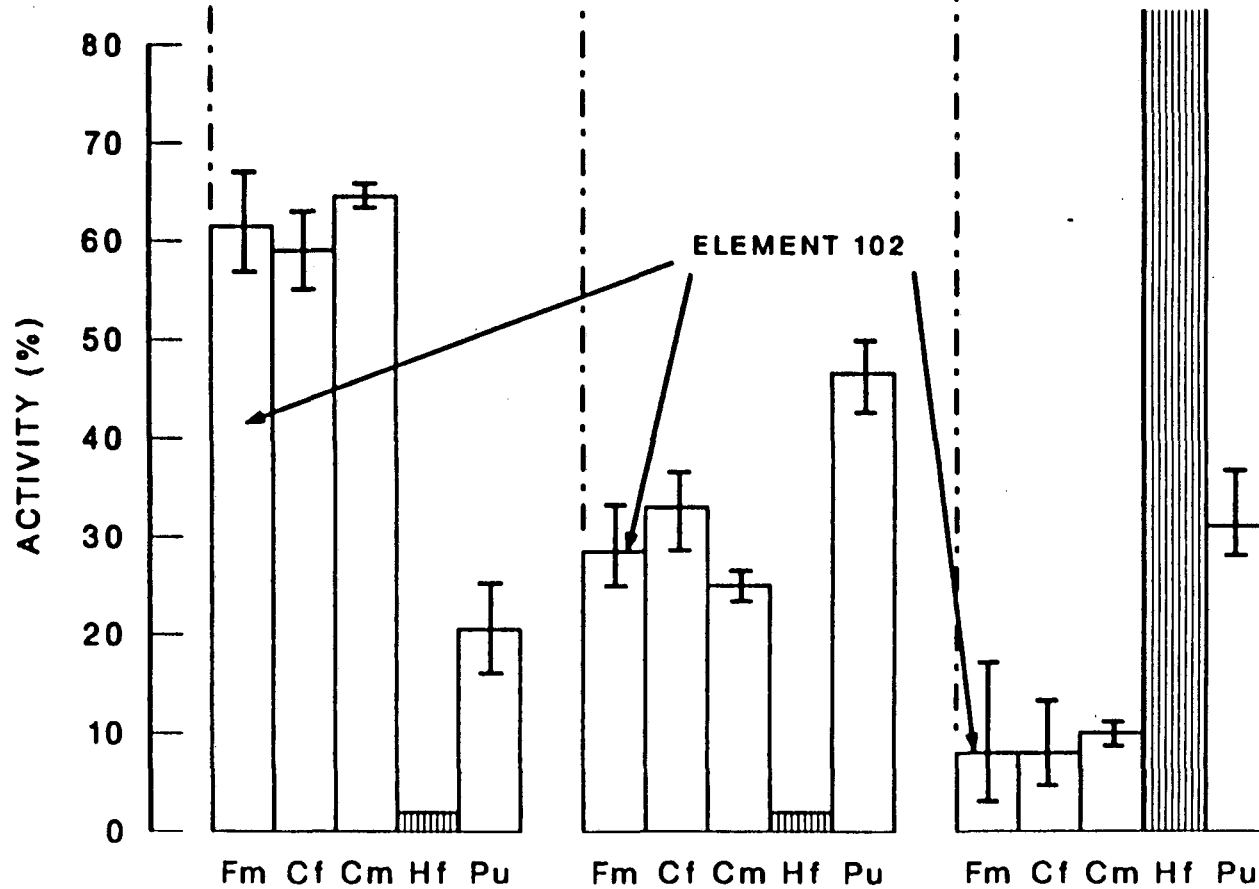
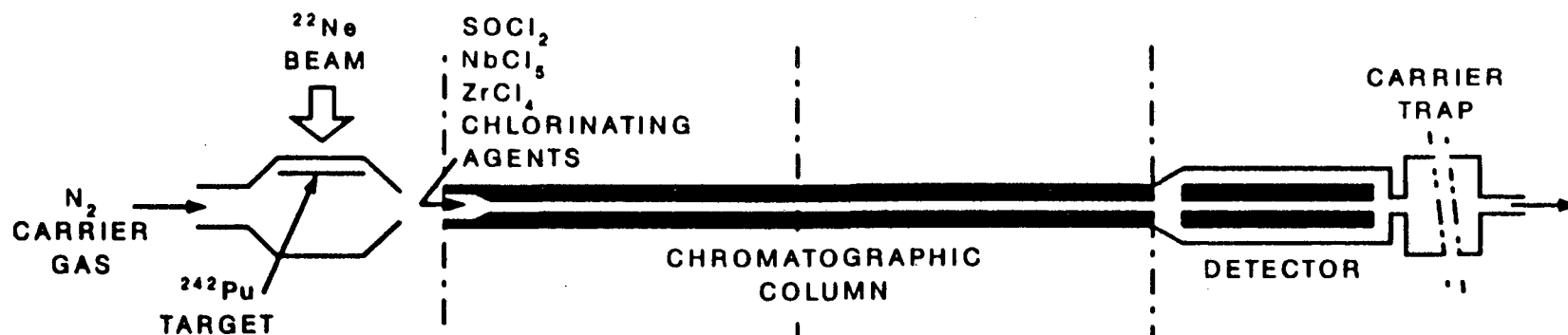


FIGURE 13

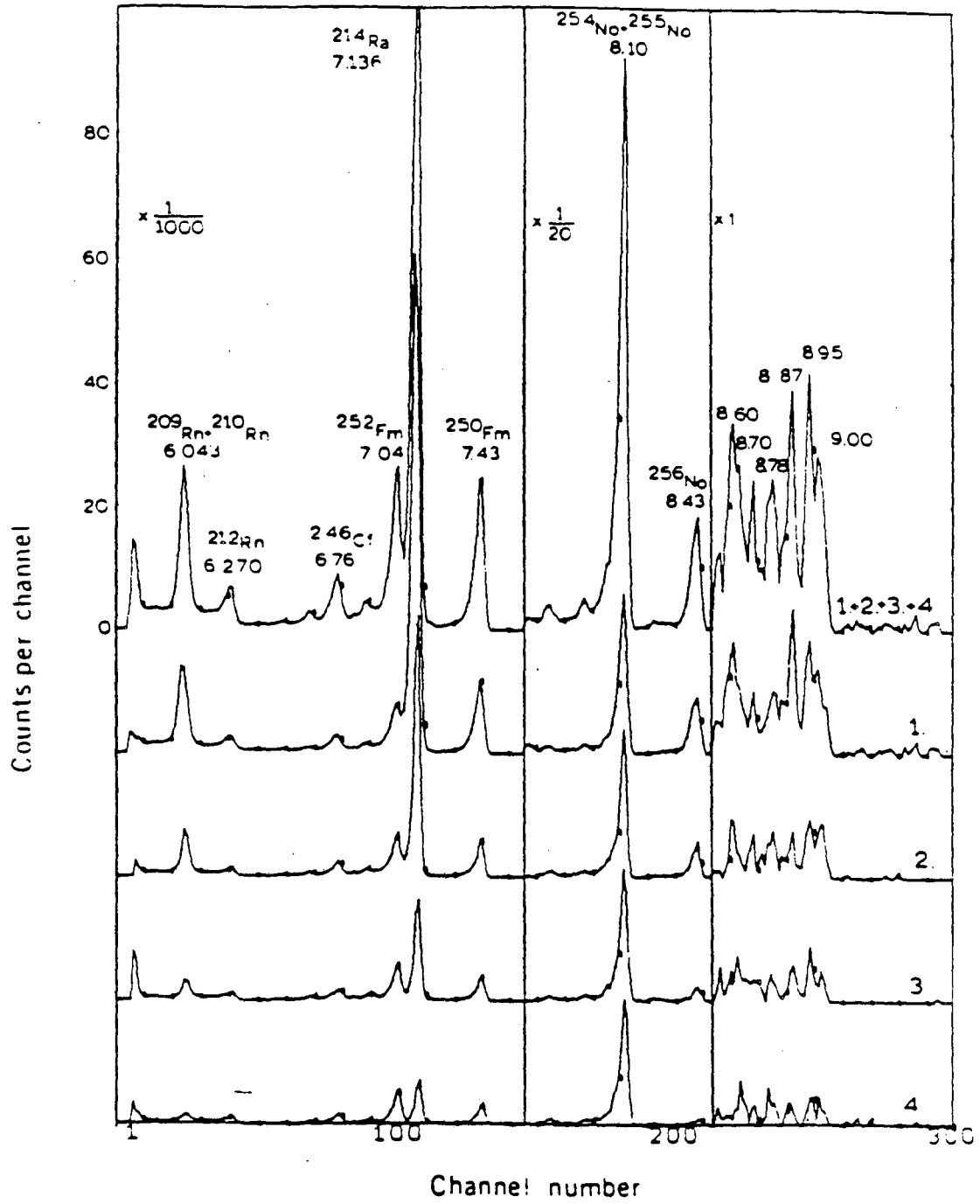
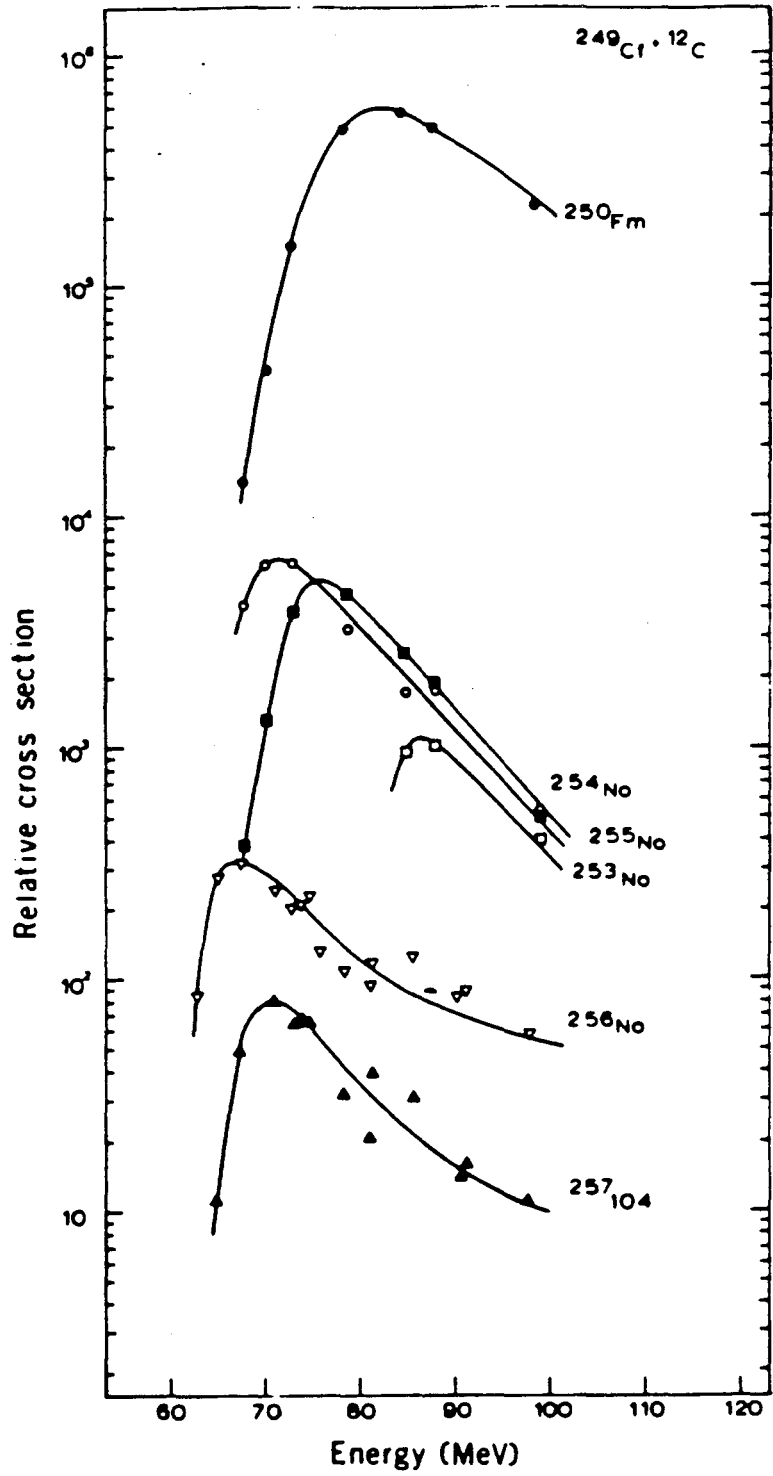
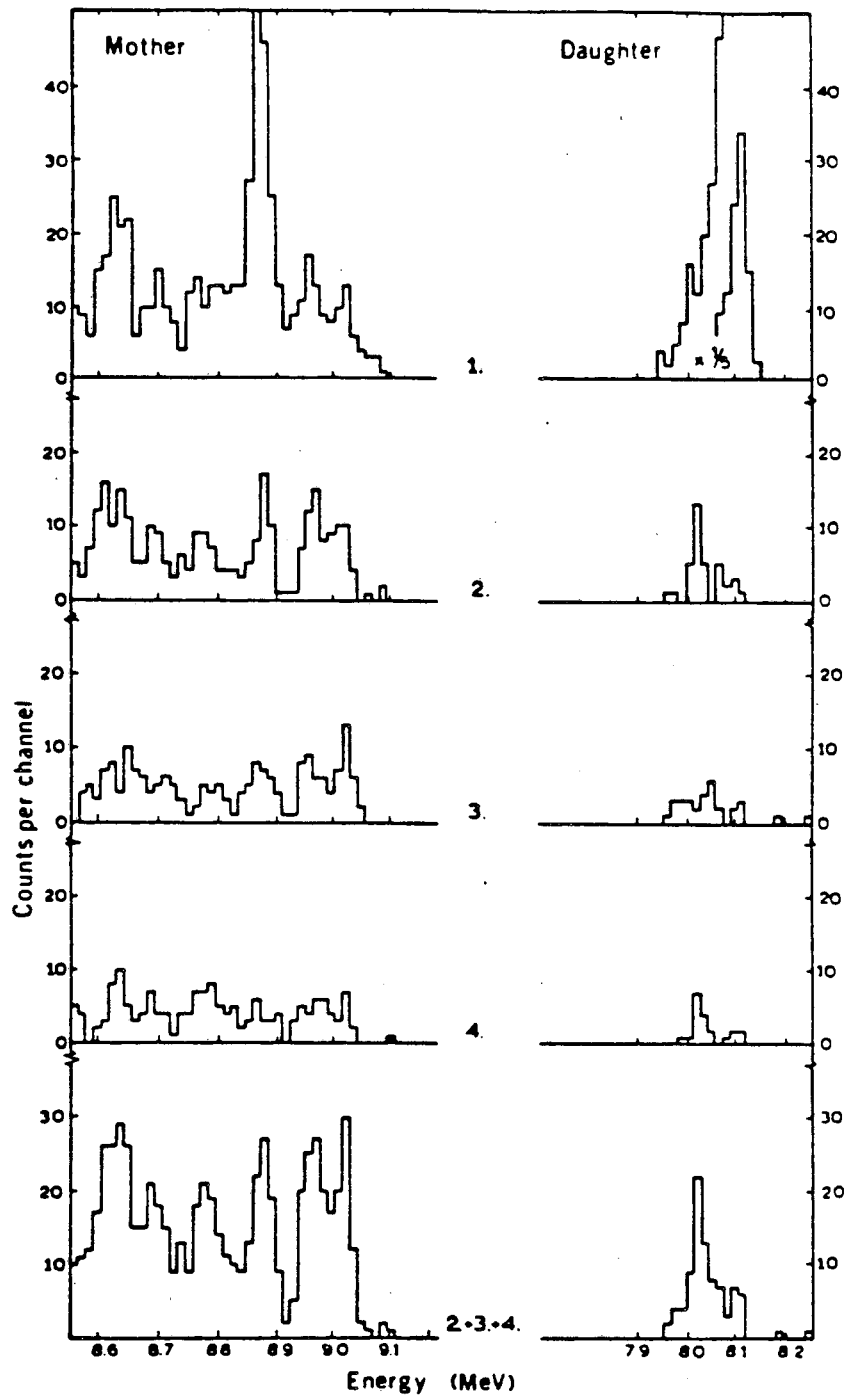


FIGURE 14



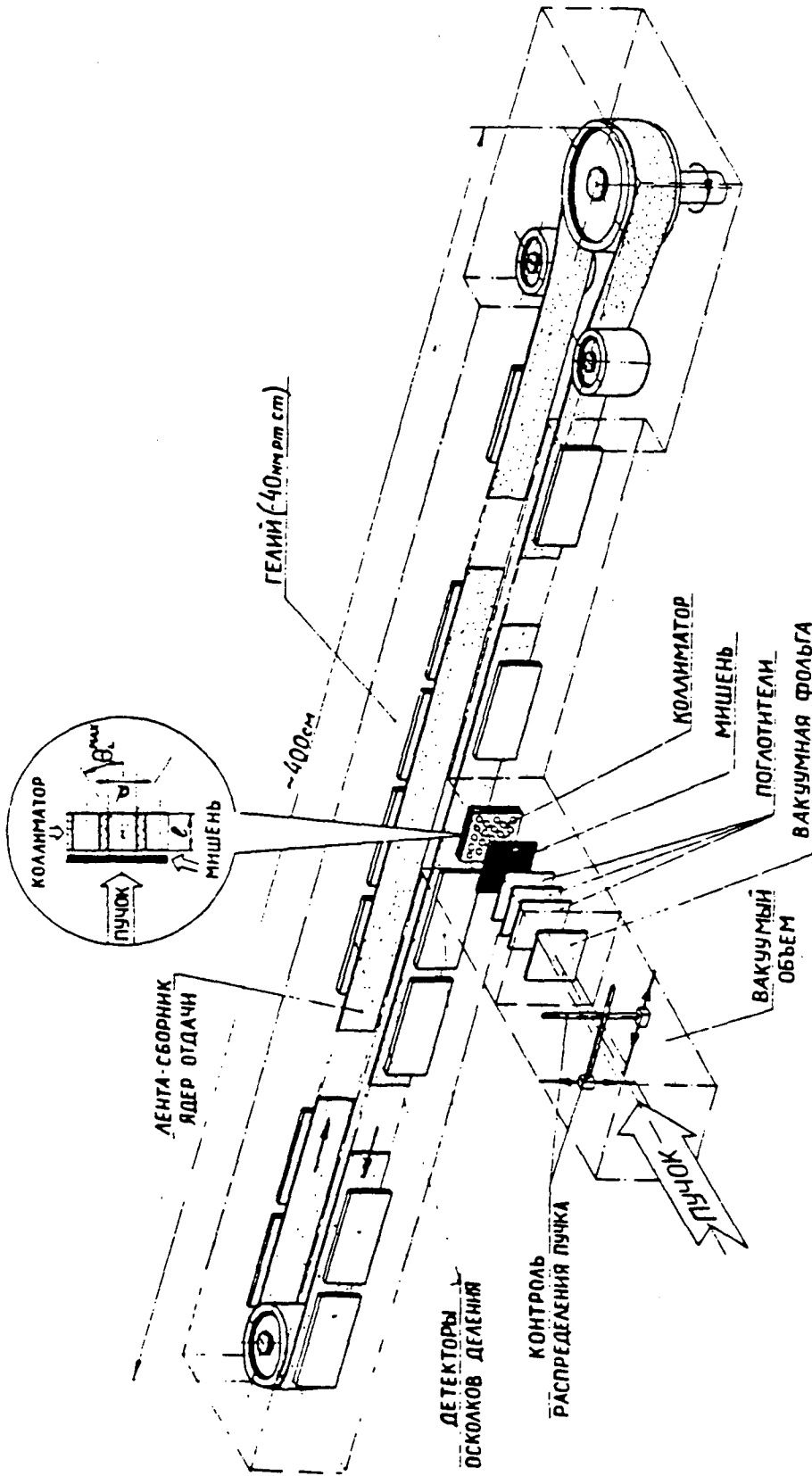
XBL 694-4815

FIGURE 15



XBL 862-475

FIGURE 16



XBL 864-1303

FIGURE 17

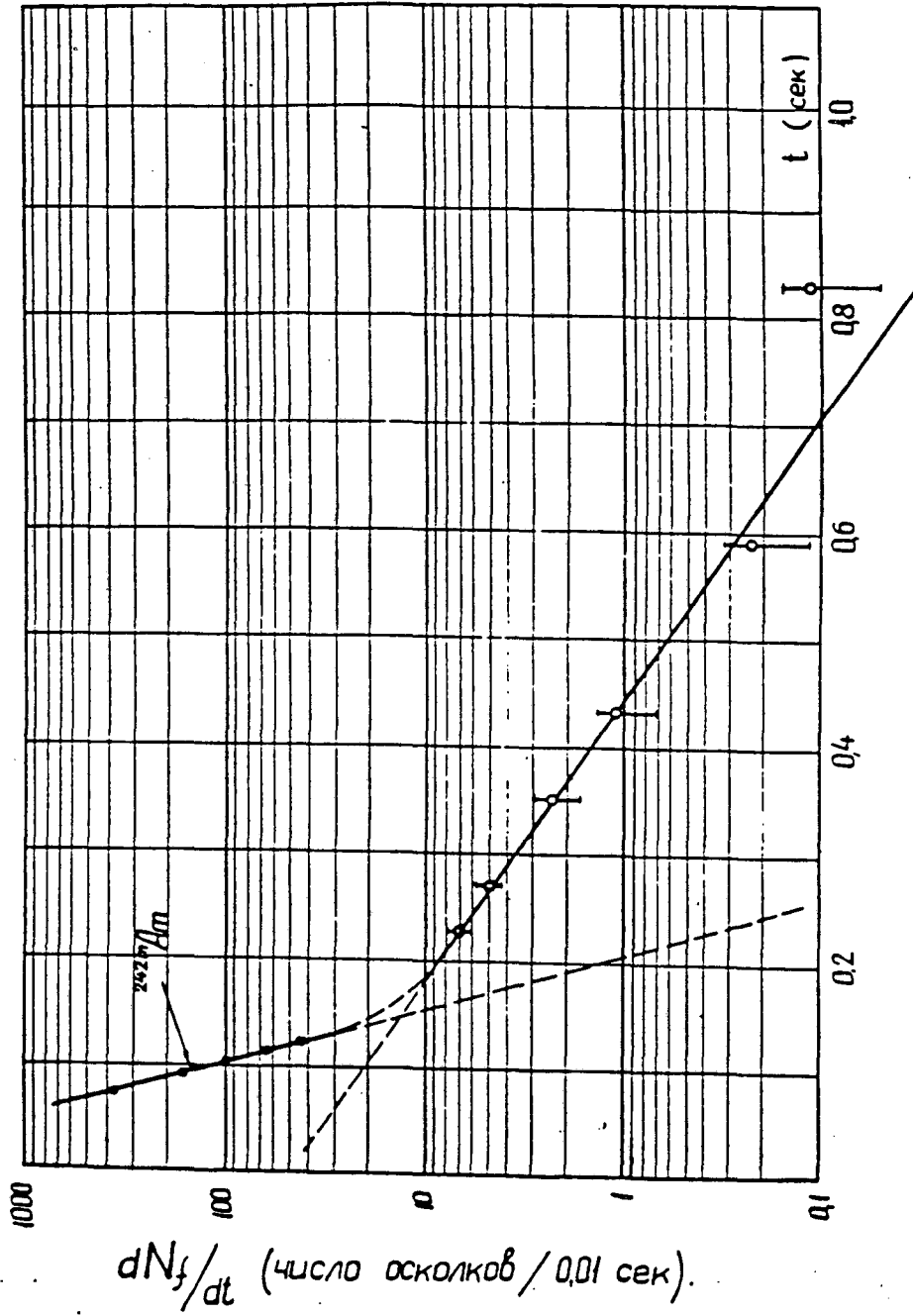


FIGURE 18

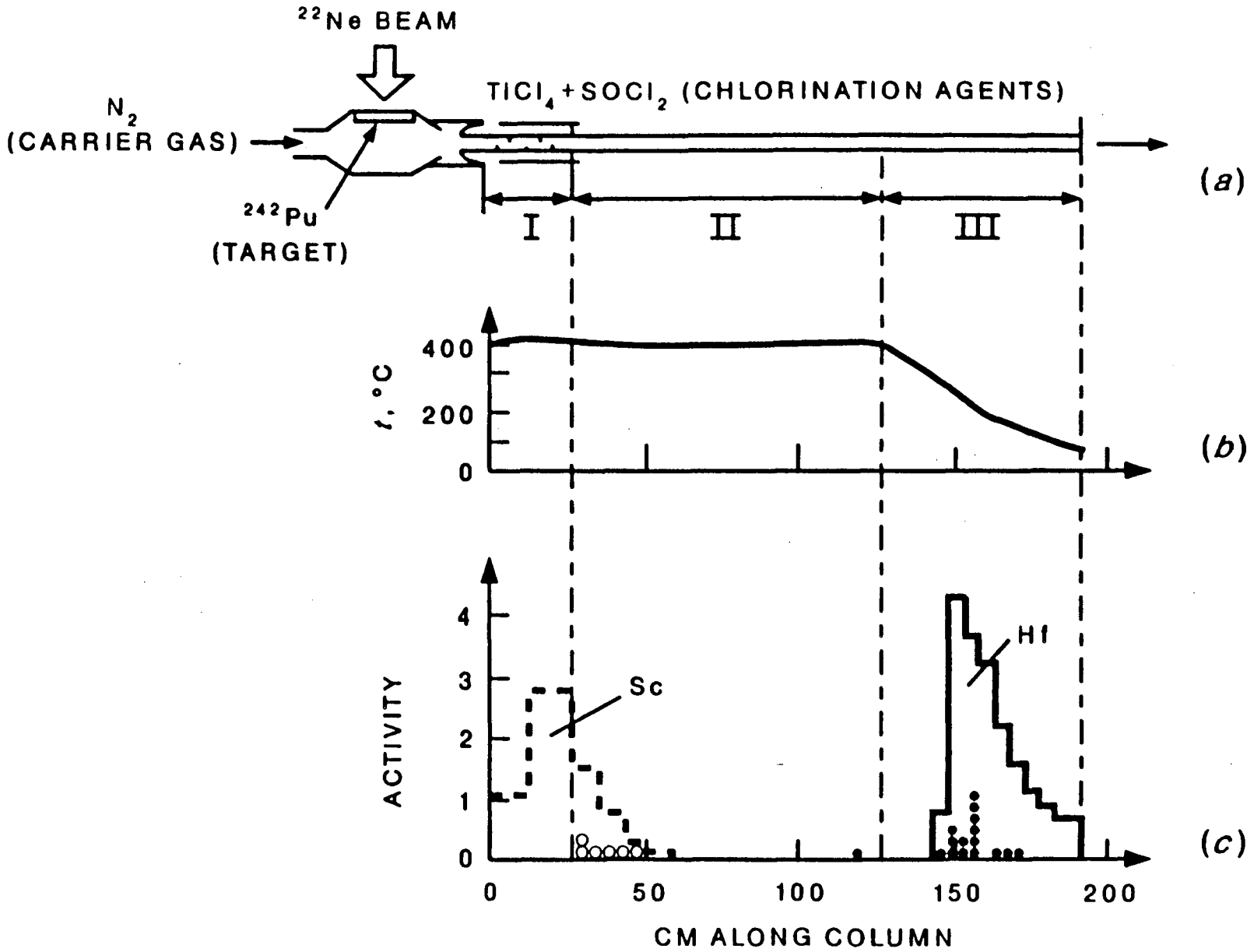


FIGURE 19

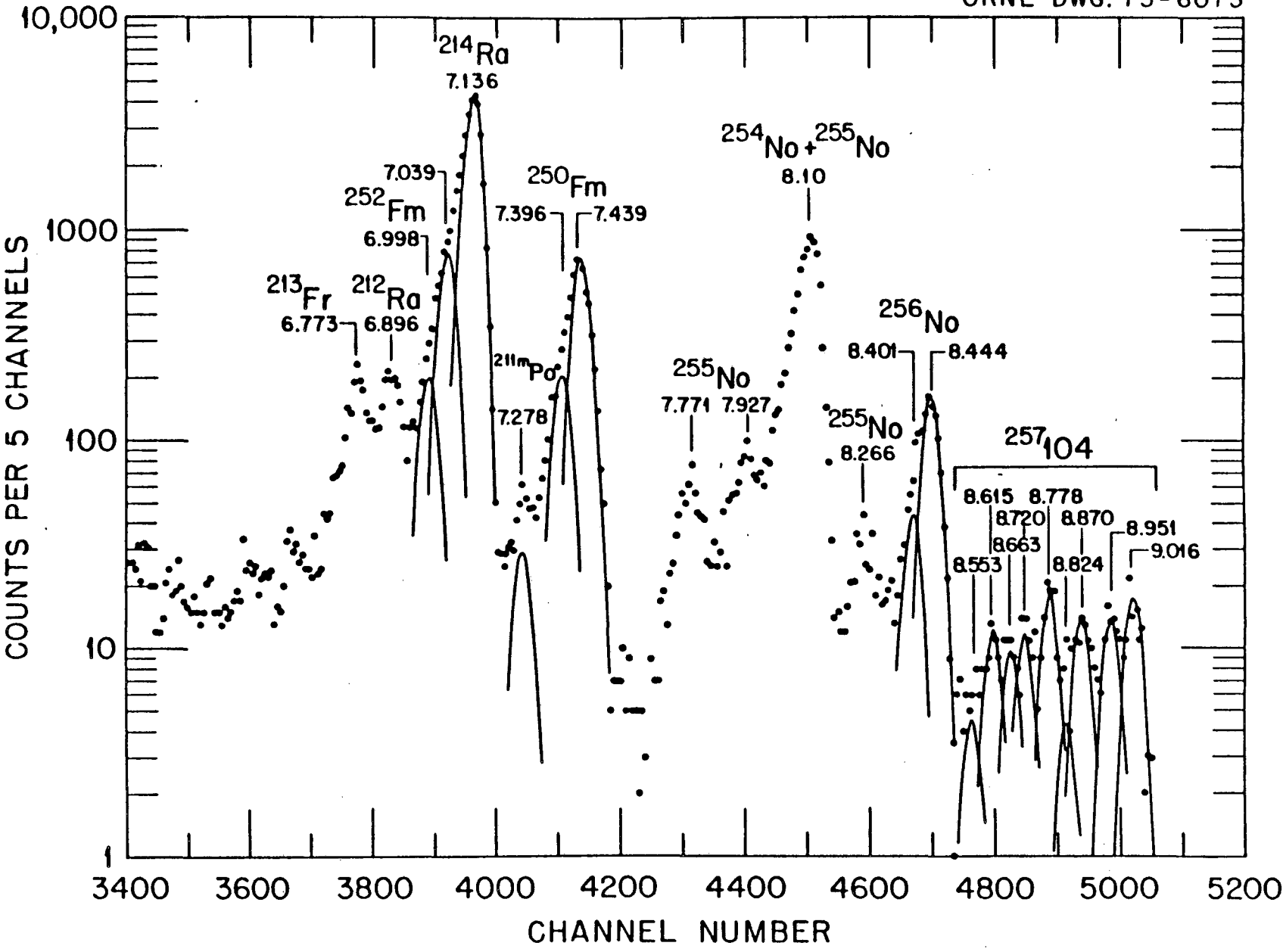


FIGURE 20

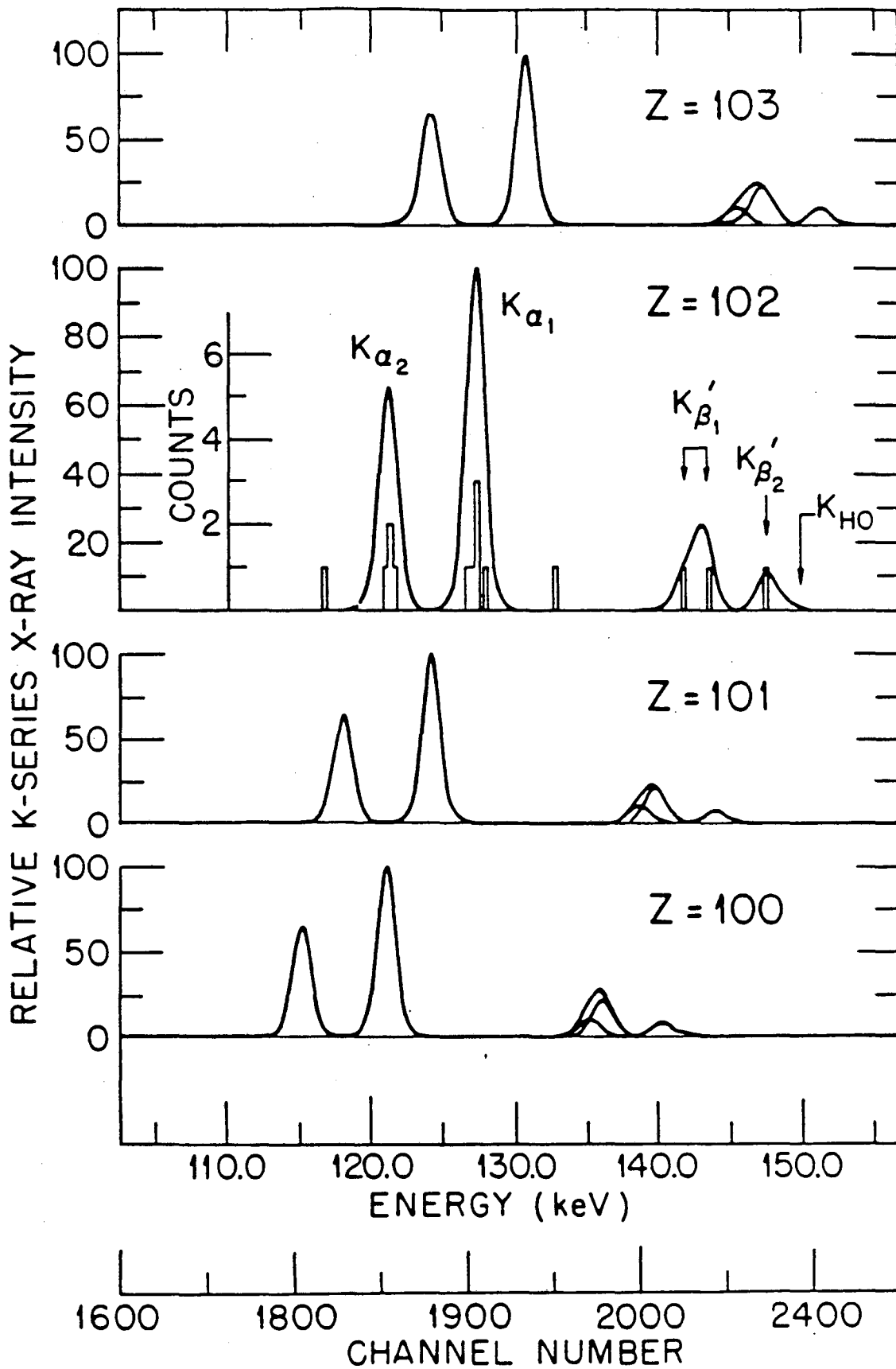


FIGURE 21

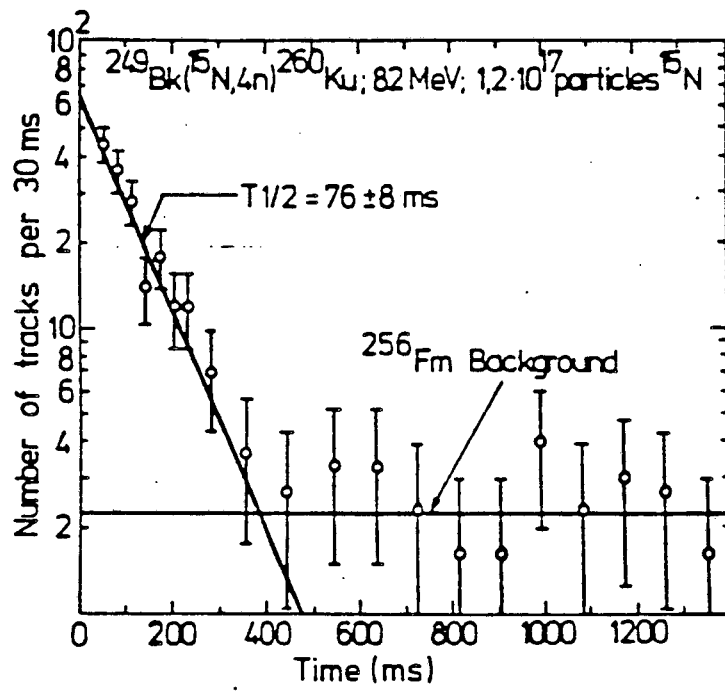
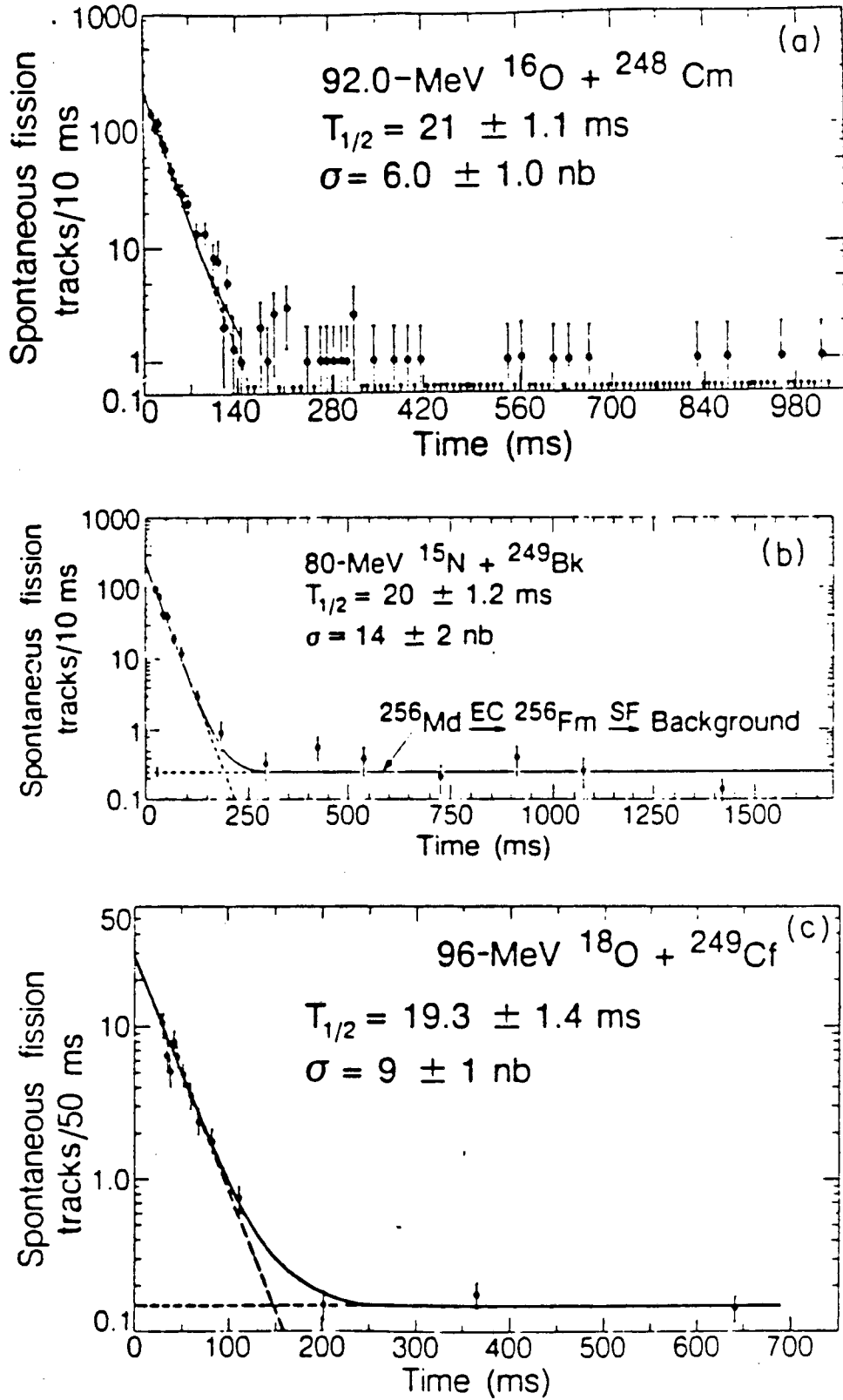


FIGURE 22



XBL 862-582

FIGURE 23

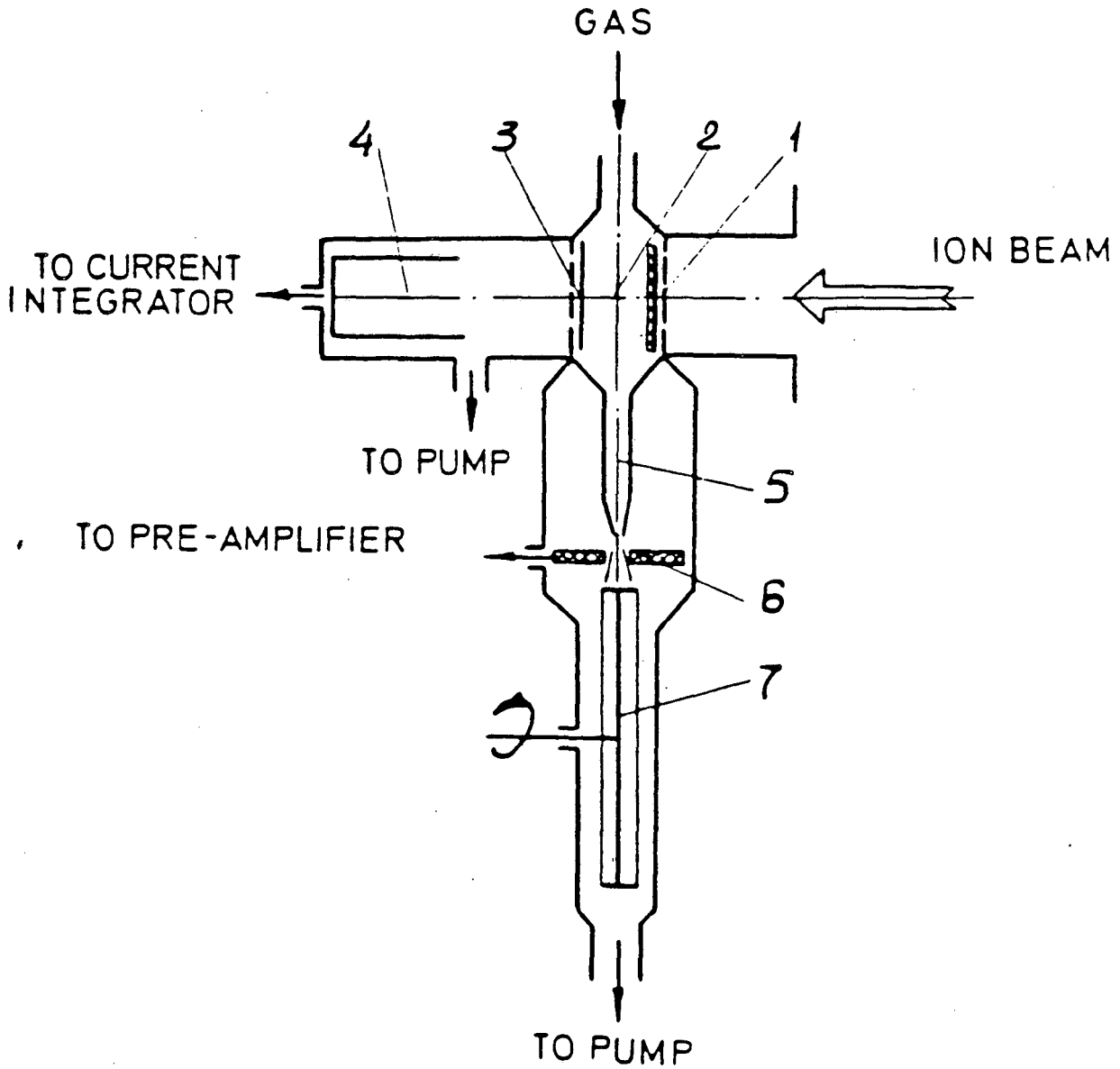


FIGURE 24

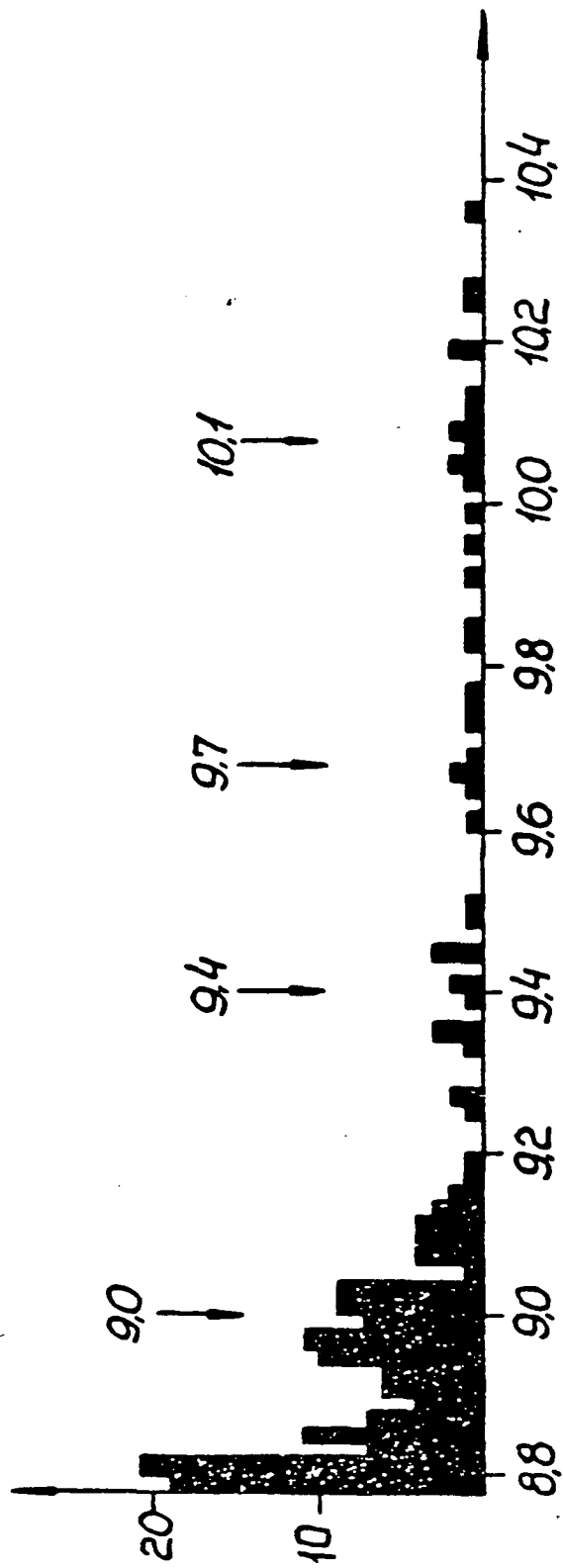


FIGURE 25

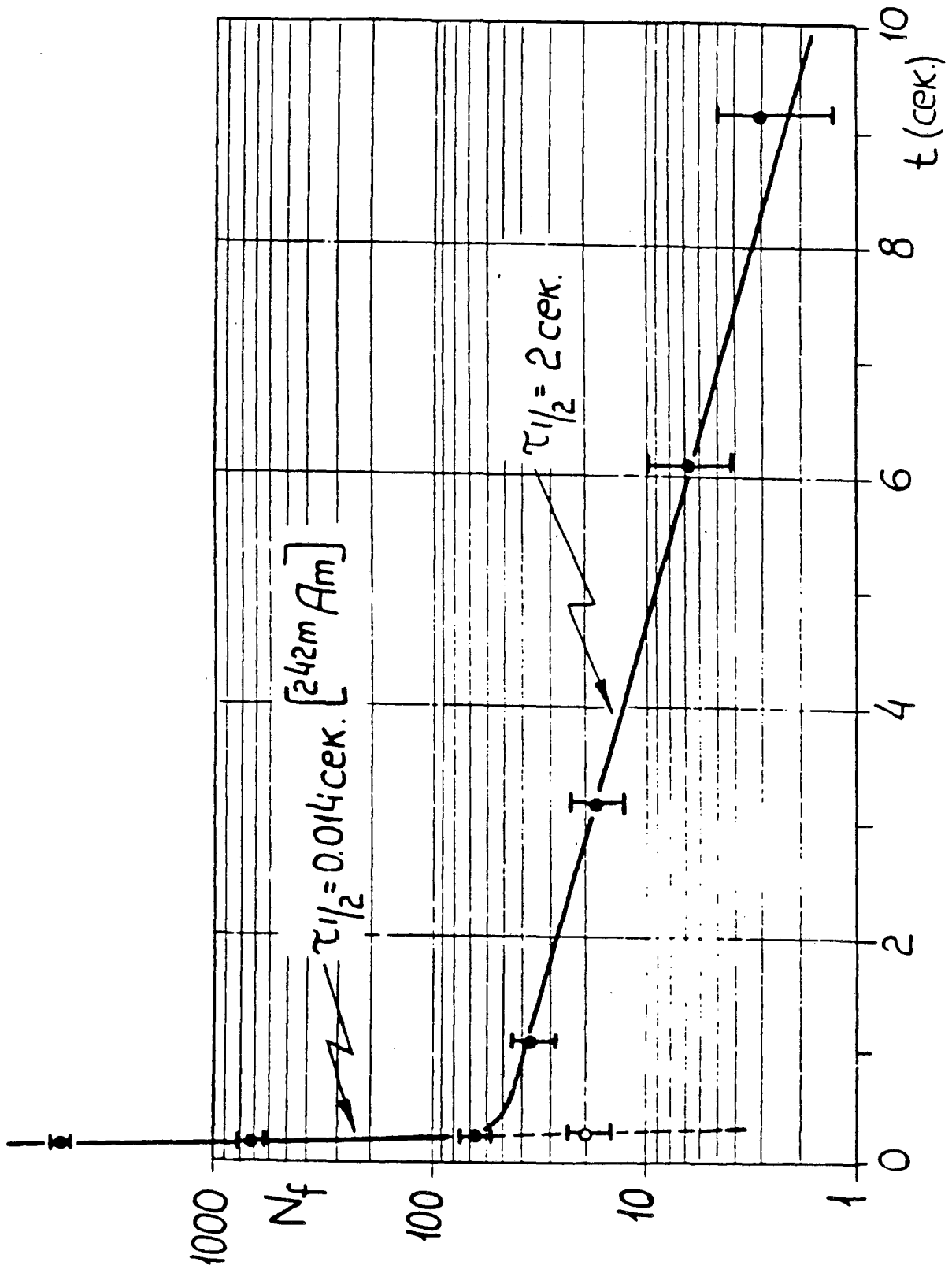
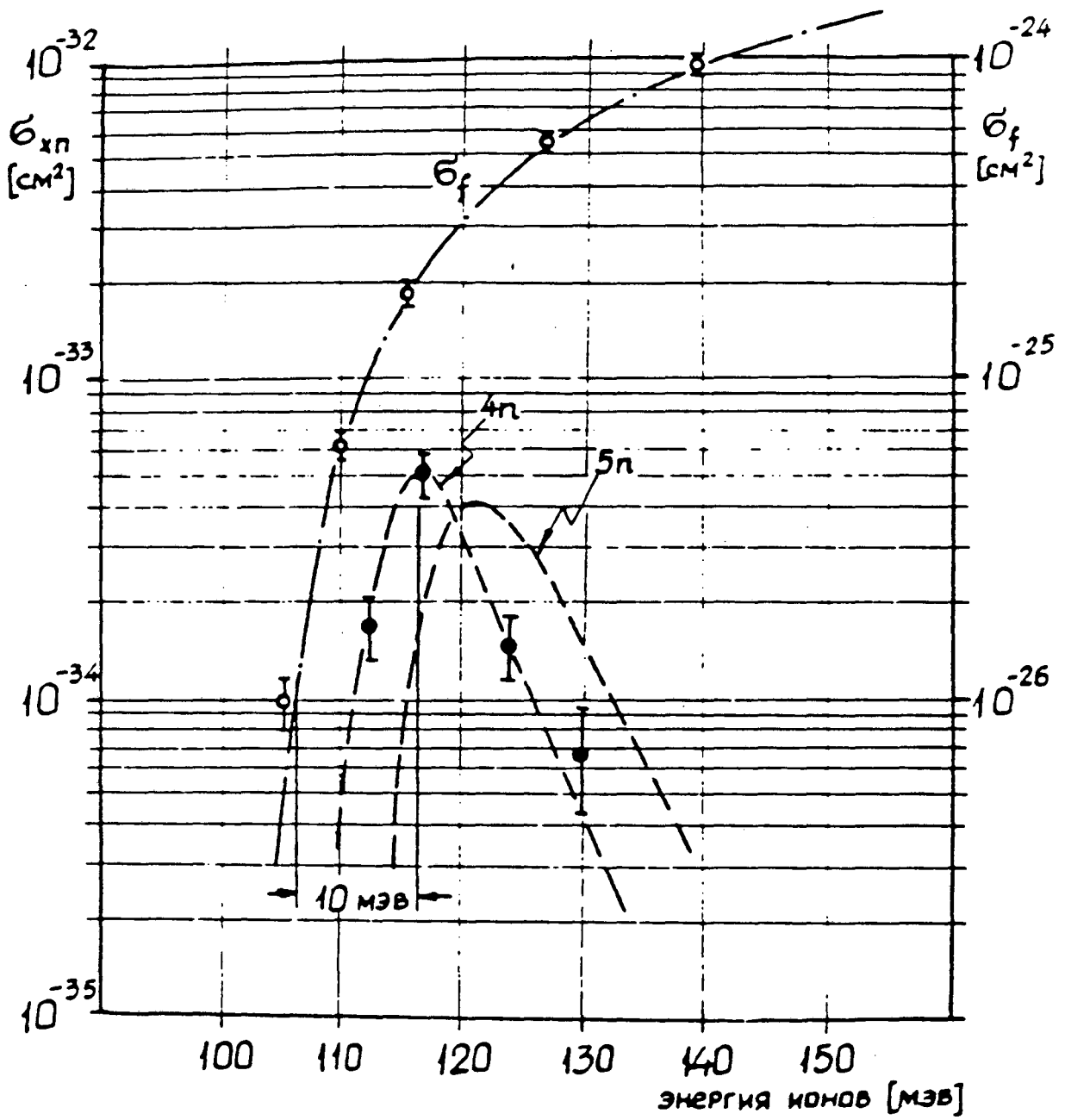


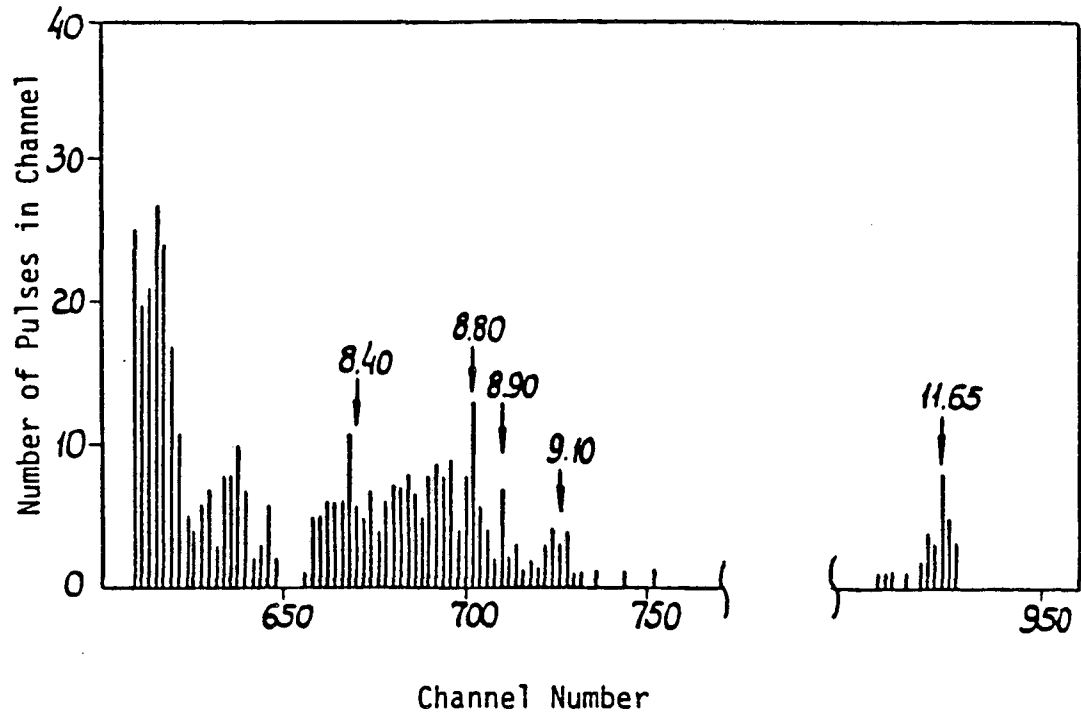
FIGURE 26

XBL 864-1304



XBL 864-1305

FIGURE 27



XBL 864-1341

FIGURE 28

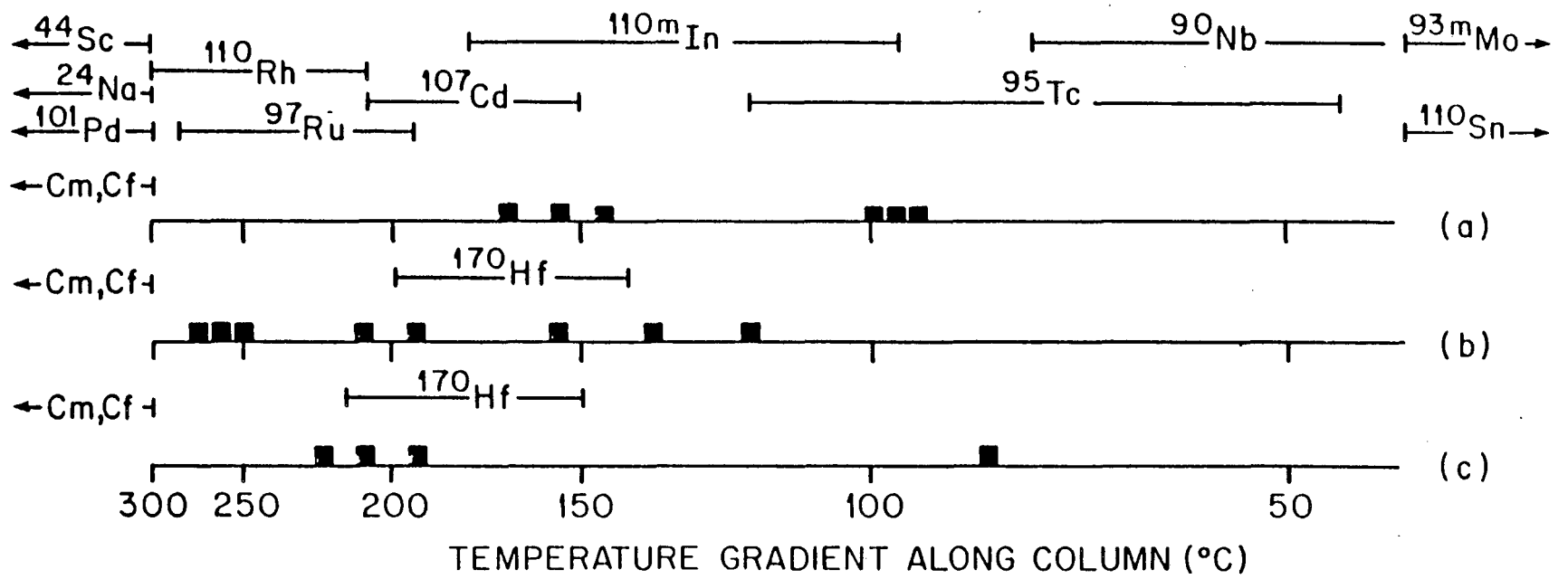


FIGURE 29

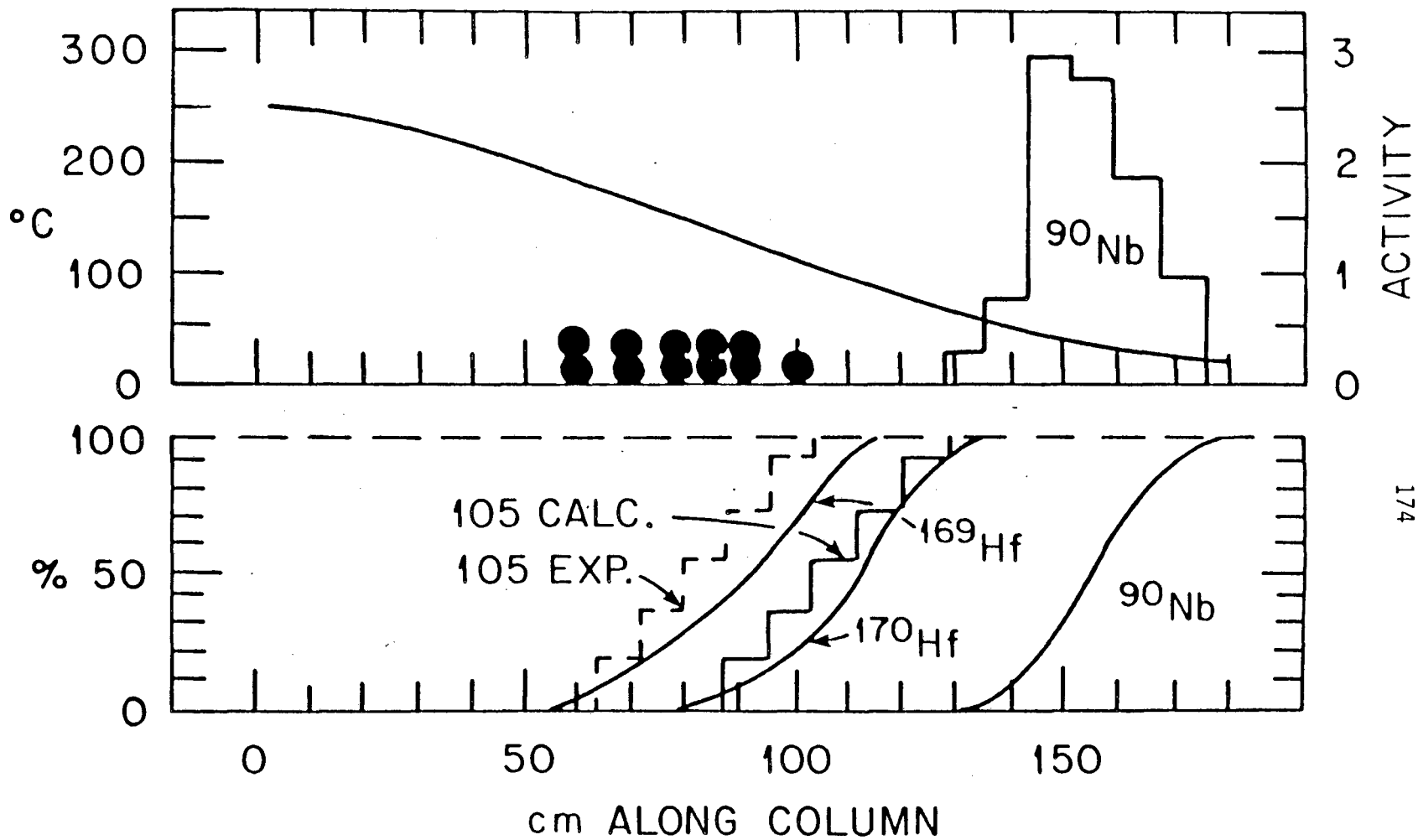
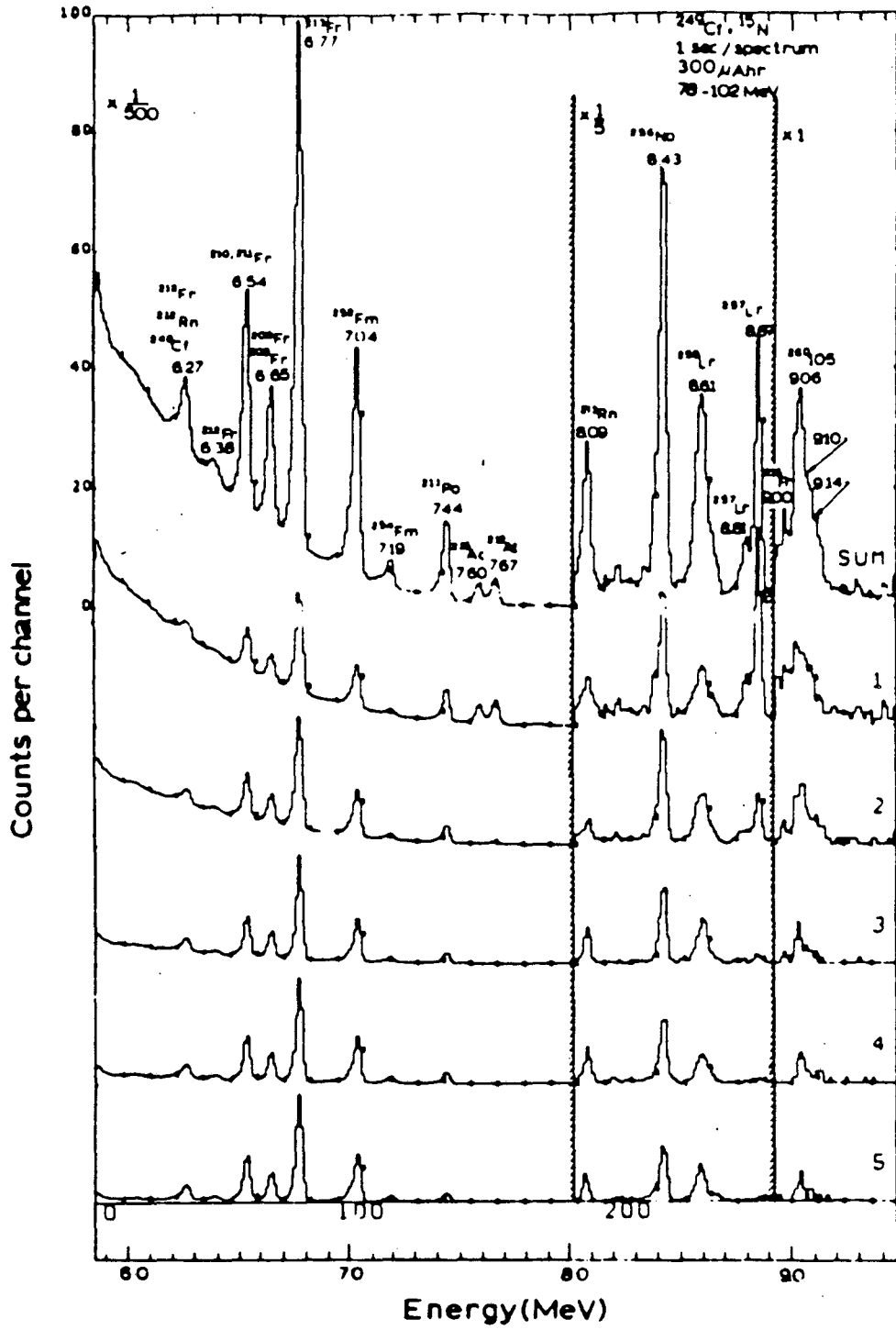
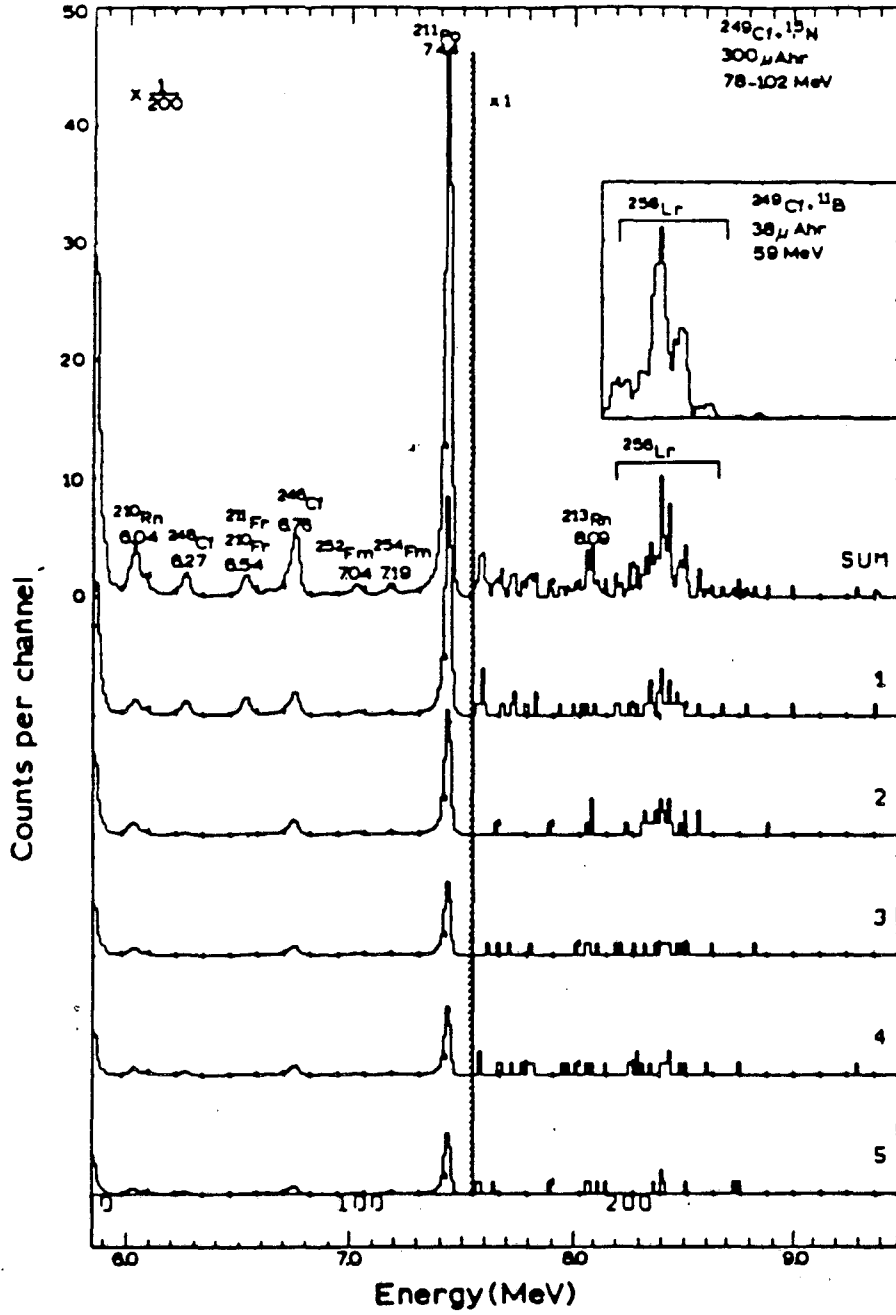


FIGURE 30



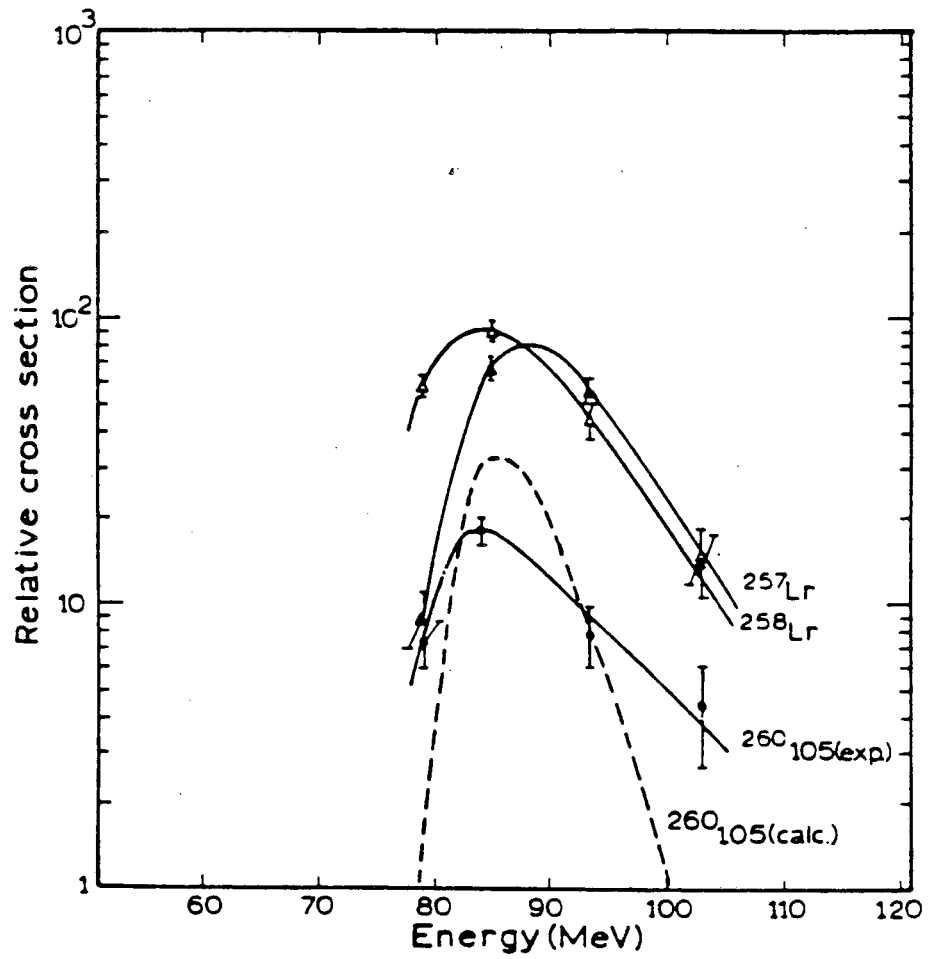
XBL 70-6171

FIGURE 31



XBL 704 6172

FIGURE 32



XBL 864-1310

FIGURE 33

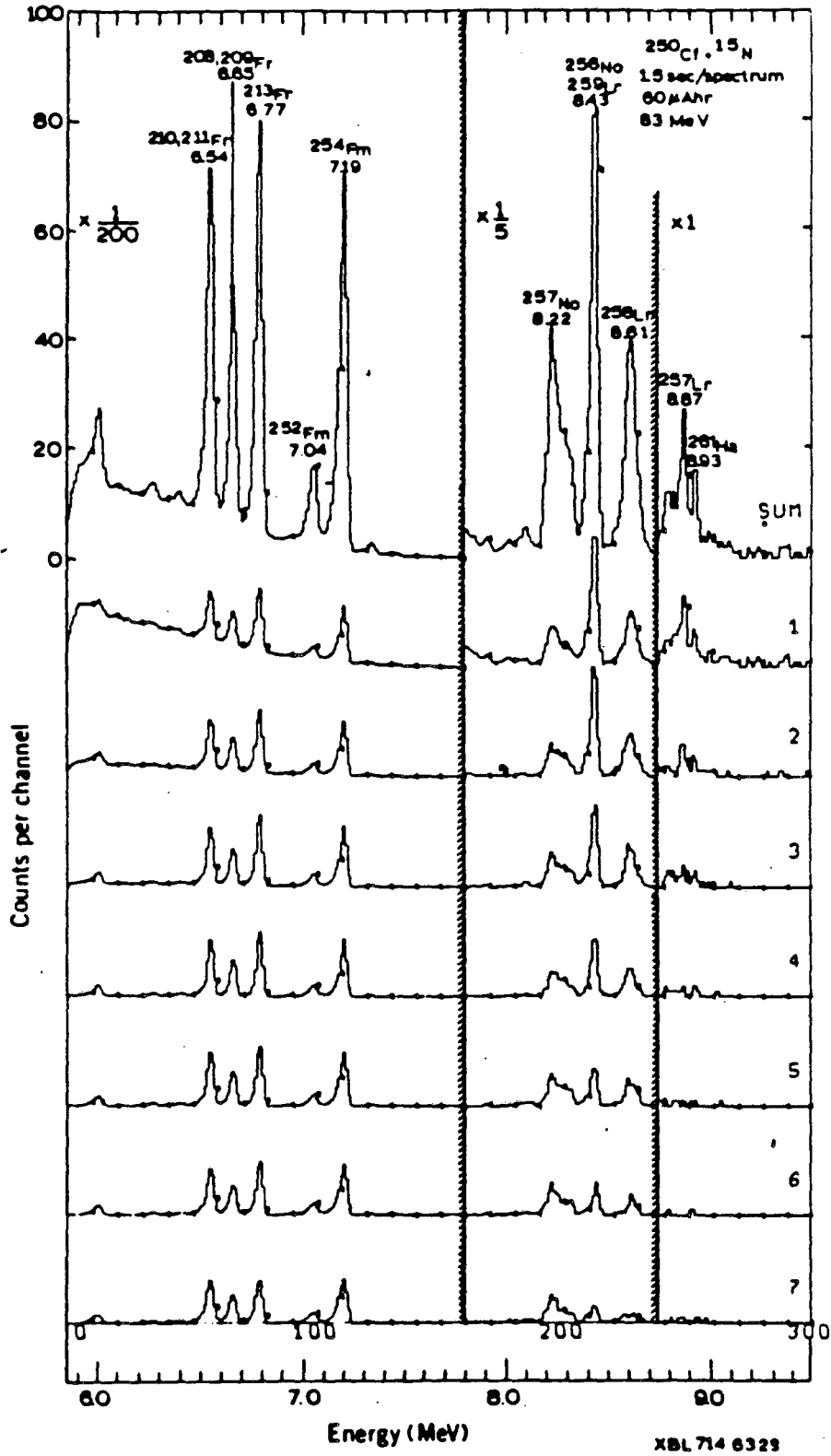


FIGURE 34

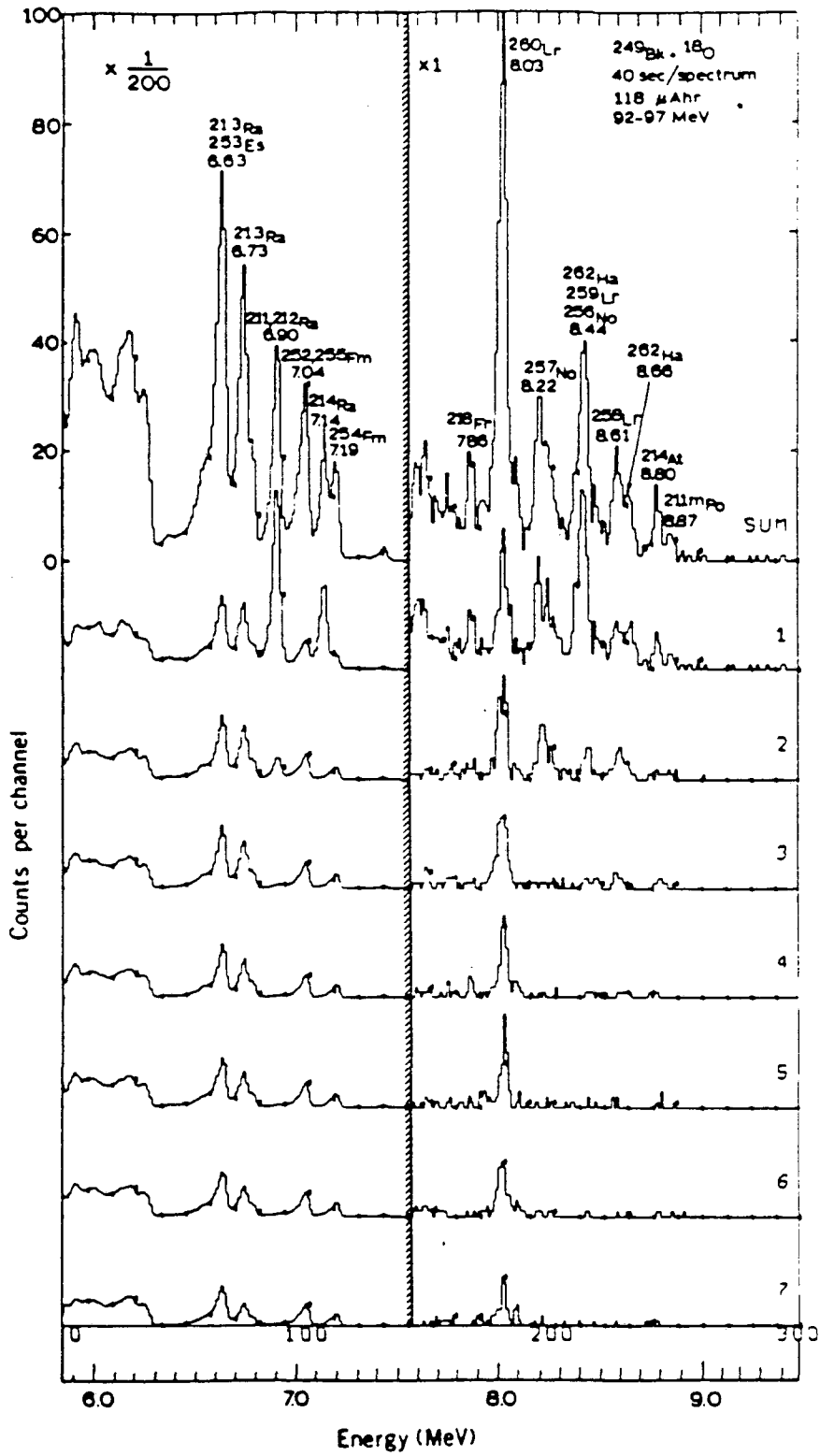


FIGURE 35

ORNL-DWG. 75-4008

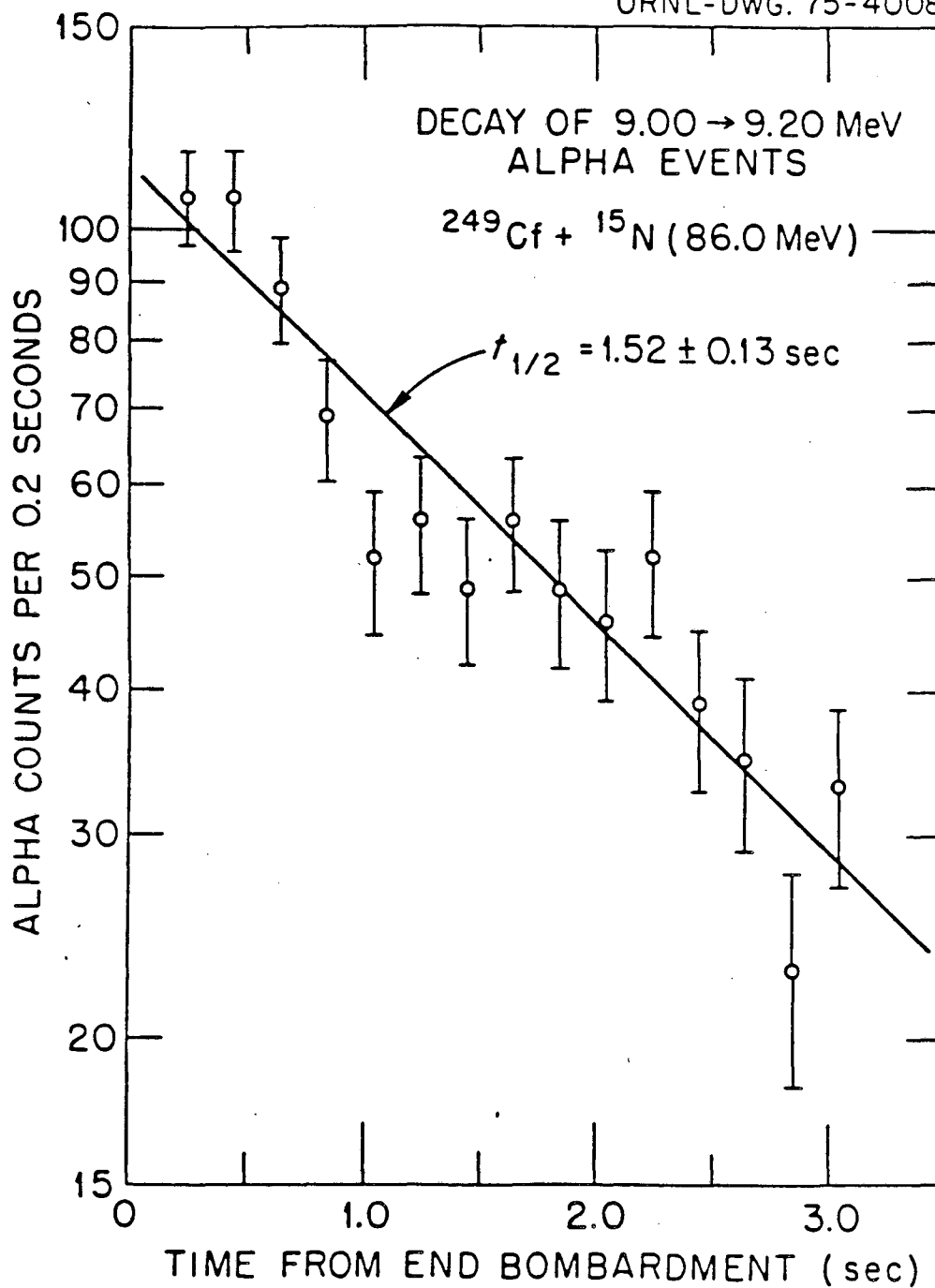


FIGURE 36

XBL 864-1317

ORNL-DWG 75-4183

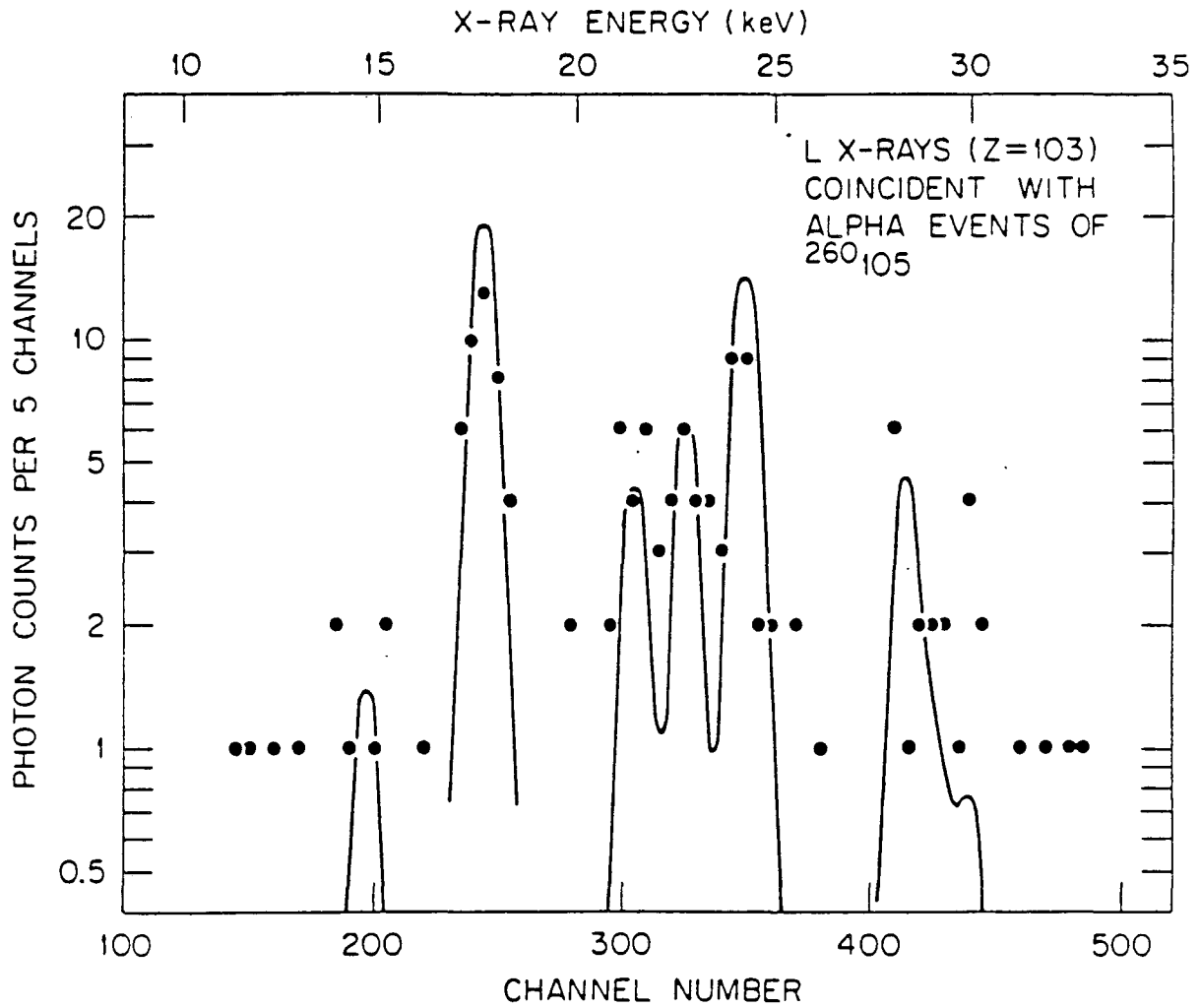


FIGURE 37

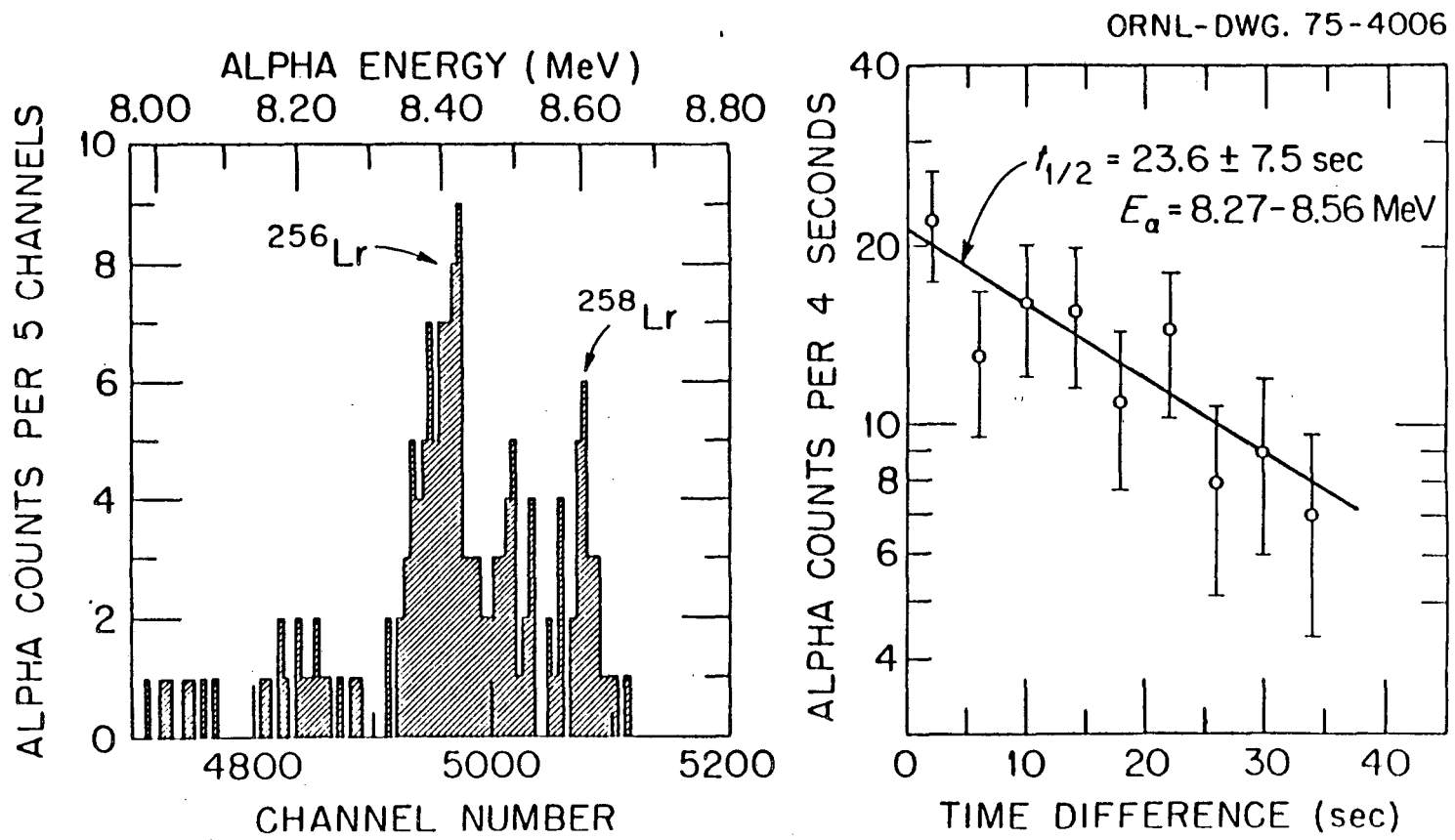


FIGURE 38

-- XBL 864-1316 --

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