of uranium was then calculated to be $2.01 \cdot 10^{14}$ atoms per gram of uranium using 1.868 for the $Kr^{38}/Kr^{85}_{(10.27 \text{ yr.})}$ ratio (corrected for decay) obtained by Wanless and Thode[8] for the thermal neutron fission of U^{235}.

The branching ratio of Kr^{85m} may now be calculated from the Rb^{85} and Kr^{85} yield data. From the rubidium yield data we have the equation

$$Y(1-X) + XY(1-e^{-\lambda t}) = 7.518 \cdot 10^{14} \,,$$

where Y is the total yield of the 85 mass chain in atoms per gram of uranium, λ is the decay constant for the 10.27-year Kr^{85}, t is 2.55 years and X and $(1-X)$ are the fractions of the Kr^{85m} decaying to the ground state of the Kr^{85} and directly to Rb^{85} respectively, and $X/(1-X)$ is taken as the branching ratio for Kr^{85m}.

Now from the krypton yield data we have

$$X\,Y = 2.01 \cdot 10^{14} \,.$$

Solving for the branching ratio we have

$$\frac{X}{1-X} = \frac{2.01 \cdot 10^{14}}{7.518 \cdot 10^{14} - 2.01 \cdot 10^{14}(1-e^{-\lambda t})} = 0.277 \,.$$

Since the flux in the larger 6.7 gram piece of uranium will be slightly lower than in the smaller 2.97 gram piece because of a slightly greater self-shielding, a small correction is necessary. The difference in flux in the two samples is calculated to be 1.7% assuming the two pieces to be spheroids[11]. Thus the Kr^{85} yield should be raised by 1.7% in comparison with the Rb^{85} yield and the final branching ratio then becomes 0.28. Since the individual yields are probably good to 2%, the branching ratio should be accurate to better than 4%.

This value is in excellent agreement with the earlier value (0.30) obtained by Bergstrom and is therefore confirmation of the decay scheme ascribed to Kr^{85m}. On the other hand, the agreement with the early mass spectrometer yield value (0.29) based on krypton isotope yields indicates that the total yield of the 85 mass chain falls on a smooth yield curve in this mass region for the thermal neutron fission of U^{235}, an assumption made in the early determination. It is of interest to note here that Wanless and Thode have recently reported extensive fine structure in the yield curve at mass 85 for the neutron fission of U^{238}. In this case, the low yield at mass 85 is accompanied by a high yield at mass 84.

If this fine structure is due to neutron emission in the 85 mass chain at, for example, As^{85} as has been suggested, then the extent of this chain branching will depend on the distribution of charge in fission. This will in turn vary considerably from U^{235} to U^{238} fission and will therefore account for the quite different results obtained in the two cases.

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Concerning the Masses of the Stable Zinc Isotopes

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To J. Mattauch for his 60th birthday

The masses of Zn^{64}, Zn^{66}, Zn^{67} and Zn^{68} have been studied mass spectroscopically by means of the O_2^{16}–$^1/_2\,Zn^{64}$, $^1/_2\,Xe^{132}$–Zn^{66}, $^1/_2\,Xe^{134}$–Zn^{67} and $^1/_2\,Xe^{136}$–Zn^{68} doublets. These studies suggest that the currently accepted masses of Zn^{64} and Zn^{66} are too large by $\sim$ 0.4 mMU. If these revisions are made several existing discrepancies between transmutation and mass data disappear.

1. Introduction

Since the war the precision with which atomic masses can be determined mass spectroscopically has greatly improved, as have also the techniques for studying the energy balance in nuclear reactions. As a result, it is frequently possible to make meaningful comparisons between mass spectroscopically-derived masses and those computed from transmutation Q-values. Among the lighter atoms such comparisons have been of great value in assessing the reliability of the mass spectroscopic work as, for example, in the case of C^{12}. Here, for a period of time, the transmutation-derived value was signifi-

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cantly lower than the most precise mass spectroscopic determinations, a situation which caused much concern. However, this discrepancy has been greatly reduced by the recent work of Mattauch and Bieri[1], which has since been confirmed, at least in a general way, by both Nier[2] and Smith[2].

2. Mass Differences Involving Stable Isotopes of Zinc

Although the amount of available transmutation data decreases rapidly with increasing atomic number, in certain heavier mass regions there is a good deal of information. Such is the case in the iron-nickel-zinc region, as can be seen in Tables 1 and 2.

Nuclides	Mass Differences (A.M.U.)		Discrepancy (10⁻⁵ A.M.U.)
	Transmutation	Mass Spectroscopic	
Ga^{71}—Ga^{69}	2.00004 ± 32	1.99974 ± 11	30 ± 33
Zn^{70}—Zn^{68}	2.00124 ± 26	2.00093 ± 9	31 ± 28
Zn^{68}—Zn^{67}	0.99877 ± 10	0.99871 ± 9	6 ± 14
Zn^{67}—Zn^{66}	1.00147 ± 21	1.00093 ± 8	54 ± 22
Zn^{66}—Zn^{64}	1.99748 ± 22	1.99767 ± 6	19 ± 23
Cu^{65}—Cu^{63}	1.99889 ± 5	1.99909 ± 8	20 ± 10
Ni^{64}—Ni^{62}	2.00044 ± 6	2.00074 ± 12	30 ± 13
Ni^{61}—Ni^{60}	0.99983 ± 1	0.99982 ± 26	1 ± 26
Ni^{60}—Ni^{58}	1.99606 ± 1	1.99576 ± 16	30 ± 16
Fe^{58}—Fe^{57}	0.99804 ± 4	0.99841 ± 41	33 ± 42
Fe^{57}—Fe^{56}	1.00078 ± 1	1.00085 ± 14	7 ± 14
Fe^{56}—Fe^{54}	1.99602 ± 3	1.99570 ± 10	32 ± 11
Cr^{54}—Cr^{53}	0.99855 ± 1	0.99858 ± 21	3 ± 21
Cr^{53}—Cr^{52}	1.00047 ± 1	1.00065 ± 12	18 ± 12

Table 1. A Comparison of Mass Differences, Derived from Transmutation and Mass Spectroscopic Data, between Nuclides of the Same Element in the Range $24 < Z < 32$.

Here a large number of mass differences have been calculated from both transmutation and mass data, those between stable nuclides of the same element appearing in Table 1, while those between stable nuclides of different *Z* are shown in Table 2. With one exception[3], the transmutation differences are based entirely upon the reaction data compiled by Van Patter and Whaling[4], and the total decay-energies given by King[5]. The mass spectroscopic differences are obtained from masses given by Collins, Nier and Johnson[6,7] and by Hogg and Duckworth[8]. Because of the large error associated with the mass of Fe^{58}, no differences involving this nuclide have been included in Table 2. The Ni^{58}–Fe^{57} transmutation difference has been calculated from the Ni^{58}–Ni^{59}–Co^{59}–Co^{58}–Fe^{58}–Fe^{57} chain, rather than by the more direct Ni^{59}–Ni^{57}–Co^{57}–Fe^{57} route, since the latter yields a highly discordant value, presumably indicating an error[9] in the listed value for the Co^{57}–Fe^{57} total decay energy.

Nuclides	Mass Differences (A.M.U.)		Discrepancy (10⁻⁵ A.M.U.)
	Transmutation	Mass Spectroscopic	
Ga^{71}—Ge^{70}	1.00103 ± 22	1.00115 ± 12	12 ± 25
Ge^{70}—Ga^{69}	0.99901 ± 22	0.99859 ± 9	42 ± 24
Zn^{70}—Ga^{69}	1.00006 ± 22	1.00001 ± 9	5 ± 24
Ga^{69}—Zn^{68}	1.00116 ± 16	1.00092 ± 9	24 ± 18
Zn^{66}—Cu^{65}	0.99845 ± 23	0.99887 ± 9	42 ± 25
Cu^{65}—Zn^{64}	0.99903 ± 5	0.99880 ± 6	23 ± 8
Cu^{65}—Ni^{64}	1.00007 ± 24	1.00080 ± 10	73 ± 26
Zn^{64}—Ni^{64}	0.001187 ± 3	0.00200 ± 7	81 ± 7
Zn^{64}—Cu^{63}	0.99986 ± 1	1.00029 ± 6	43 ± 6
Ni^{64}—Cu^{63}	0.99868 ± 1	0.99829 ± 9	39 ± 9
Cu^{63}—Ni^{62}	1.00176 ± 6	1.00245 ± 11	69 ± 12
Ni^{60}—Fe^{57}	2.99640 ± 22	2.99566 ± 16	74 ± 27
Ni^{58}—Fe^{57}	1.00034 ± 22	0.99990 ± 14	44 ± 26
Fe^{56}—Mn^{55}	0.99726 ± 3	0.99693 ± 14	33 ± 15
Mn^{55}—Fe^{54}	0.99876 ± 1	0.99877 ± 11	1 ± 11
Mn^{55}—Cr^{54}	0.99945 ± 15	0.99951 ± 23	6 ± 28
Fe^{54}—Cr^{53}	0.99877 ± 25	0.99932 ± 10	55 ± 27

Table 2. A Comparison of Mass Differences, Derived from Transmutation and Mass Spectroscopic Data, between Nuclides of Different Elements in the Range $24 < Z < 32$.

In Table 1 the only serious discrepancy exists in the case of the Zn^{67}–Zn^{66} mass difference.

In Table 2 the situation is much less satisfactory and has led both Wapstra[10] and us[11] to suggest that the nickel masses may all be too small by ∼ 0.6 mMU. This is an attractive hypothesis since it would account for the largest discrepancies, and we are currently attempting to put it to an experi-

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mental test. However, it may be that the inconsistencies cannot be resolved this simply.

In this paper we describe new mass comparisons involving Zn^{64}, Zn^{66}, Zn^{67} and Zn^{68}. The Zn^{64} work grew out of our concern for the Zn^{64}–Ni^{64} discrepancy (see Table 2), where the well-authenticated decay scheme of Cu^{64} discourages the hope that the transmutation value may be in error. The Zn^{66}, Zn^{67} and Zn^{68} work was expected to shed some light on the Zn^{67}–Zn^{66} discrepancy.

3. Experimental

These experiments were done with a Dempster-type double-focusing mass spectrograph[12] operating with a resolution of ~ 1 part in 7000. The ion source was a modified Shaw source[13], originally constructed by R. B. Shields. In this arrangement a metal, either in metallic form or as a salt, is located in a small crucible which is heated by bombardment with 500-volt electrons. These electrons serve the additional purpose of ionizing the vaporized material and, at the same time, ionize gaseous materials which happen to be present. The source, consequently, provides a convenient method for simultaneously obtaining metallic and gaseous ions, and was used by Shaw for this purpose.

The mass of Zn^{64} was determined by means of the O_2^{16}–$^1/_2\,Zn^{64}$ doublet at mass number 32, while the Zn^{66}, Zn^{67} and Zn^{68} masses were studied via the $^1/_2\,Xe^{132}$–Zn^{66}, $^1/_2\,Xe^{134}$–Zn^{67} and $^1/_2\,Xe^{136}$–Zn^{68} doublets at mass numbers 66, 67 and 68, respectively. The mass differences so found are given in Table 3.

Nuclides	Mass Differences in mMU
O_2^{16}—$^1/_2\,Zn^{64}$	25.45 ± 0.15
$^1/_2\,Xe^{132}$—Zn^{66}	25.61 ± 0.15
$^1/_2\,Xe^{134}$—Zn^{67}	25.25 ± 0.20
$^1/_2\,Xe^{136}$—Zn^{68}	27.20 ± 0.20

Table 3. List of New Atomic Mass Differences.

The O_2^{16}–$^1/_2\,Zn^{64}$ value is based on twelve doublet photographs, seven of which were taken in May, 1954, and the remainder in June and July, 1955. The $^1/_2$ Xe–Zn values are based on ten, nine and eight doublet photographs, respectively, for several of which the pressure in the mass spectrograph was deliberately and substantially increased, with no observable effect on the doublet spacing.

4. Discussion

Zn^{64}. The mass of Zn^{64} deduced from these experiments is 63.94909 ± 15 AMU, which should be compared to the value of 63.94955 ± 2 AMU obtained by Collins, Nier and Johnson. This new value for Zn^{64}, when combined with the Minnesota value for Ni^{64} (63.94755 ± 7 AMU) leads to the mass difference Zn^{64}–Ni^{64} = 1.54 ± 0.17 mMU = 1.43 ± 0.15 MeV. The value obtained from the decay scheme of Cu^{64} is 1.187 ± 0.003 mMU = 1.105 ± 0.003 MeV, whereas the Minnesota values for both Zn^{64} and Ni^{64} give Zn^{64}–Ni^{64} = 2.00 ± 0.08 mMU = 1.86 ± 0.08 MeV.

This new mass value for Zn^{64} carries with it a much larger statistical error than does that of Collins, Nier and Johnson. However, it is an independent determination which is tied directly to O^{16}, and has the virtue that it materially improves the agreement between transmutation and mass data at mass number 64. Further, as can be seen from Table 2, it also improves the agreement in both the Cu^{65}–Zn^{64} and Zn^{64}–Cu^{63} differences, particularly the latter.

Zn^{66}, Zn^{67} and Zn^{68}. The masses of Xe^{132}, Xe^{134} and Xe^{136} have been accurately determined by Halsted[14]. When these are recalculated on the basis of Mattauch and Bieri's mass of C^{12}, and combined with the $^1/_2$ Xe–Zn mass differences listed in Table 3, they lead to the values Zn^{66} = 65.94737 ± 15 AMU, Zn^{67} = 66.94857 ± 20 AMU and Zn^{68} = 67.94740 ± 20 AMU. It is instructive to calculate certain Zn–Zn mass differences using these new values, and to compare them with the corresponding differences in Table 1. This is done in Table 4.

Nuclides	Transmutation	Mass Spectroscopic (Minnesota)	Mass Spectroscopic (this paper)
Zn^{66}—Zn^{64}	1.99748 ± 22	1.99767 ± 6	1.99828 ± 20
Zn^{67}—Zn^{66}	1.00147 ± 21	1.00093 ± 8	1.00120 ± 25
Zn^{68}—Zn^{67}	0.99877 ± 10	0.99871 ± 9	0.9988 ± 3

Table 4. Comparison of New and Existing Mass Differences (in AMU).

In the first place, it is clear from the Zn^{66}–Zn^{64} mass difference that the xenon-derived masses are too large. This may be due either to incorrect xenon masses or to some systematic error in our own comparisons. We, naturally, prefer the former alternative.

In either event, we expect the Zn^{68}–Zn^{67} and Zn^{67}–Zn^{66} differences to be correct and it is, therefore, a matter of satisfaction to note that they agree

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with the transmutation values. For the Zn^{67}–Zn^{66} differences, the weighted mean of our value and the transmutation value is 1.00135 ± 16 AMU. If this be the correct value, it indicates that the Minnesota value for Zn^{66} is too large by ~ 0.4 mMU. Such a change would remove the Zn^{66}–Cu^{65} discrepancy and, in the light of the smaller Zn^{64} value would provide a concordant value for the Zn^{66}–Zn^{64} difference.

We appreciate the assistance of our colleague, J. G. V. Taylor, in the calculation of the transmutation mass differences. The work reported in this paper has been supported by the Office of Scientific Research, Air Research and Development Command, U.S. Air Force, the National Research Council of Canada, the Ontario Research Foundation and the Shell Oil Company of Canada.

Preliminary Report on a large Mass Spectrograph newly constructed at Osaka University

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To J. Mattauch for his 60th birthday

A new large mass spectrograph of double focusing type was constructed at Osaka University. The apparatus has a uniform magnetic field of $\pi/3$ sector type and $\sqrt{2}\,\pi/3$ cylindrical electric field. The radius of central ion beam in magnetic field is 1200 mm and that in electric field 1093 mm.

From the preliminary experiment, the dispersion coefficient was estimated to be about 9 cm for $1^0/_0$ mass difference and the maximum resolving power about 500,000. In the preliminary determination of N_2^{14}–$C^{12}O^{16}$ doublet mass difference, a value of 112.441 (10^{-4} amu) was obtained with the present condition.

In the past we have constructed a mass spectrograph of Bainbridge-Jordan type at Osaka University. With the mass spectrograph of a modified type we have achieved a maximum resolution of about 60,000 towards the end of 1950[1].

Since then, we were planning to construct a larger mass spectrograph of double focusing type in order to raise the resolving power and accuracy about one order of magnitude. To obtain such a large resolving power with the ordinary mass spetrograph the following two methods may be conceivable in general: one is that of reducing the width of mass spectrum line by improving the focusing characteristic and the other that of increasing the mass dispersion of the apparatus itself by enlarging the linear dimension. In order to raise the resolving power about one order magnitude with the first method, the line width must be made $< 1 - 2 \cdot 10^{-3}$ mm with our old mass spectrograph of the modified Baindridge-Jordan type. Moreover, with this method, there are some difficulties involved in the measurement of line separation due to the relatively larger grain size of our self-made Schumann plate, the limited accuracy of our comparator, the error due to shrinkage of the emulsion layer at the time of development, etc. Therefore, the authors decided to adopt the second method, that is to increase the linear dimension of the apparatus itself.

Even in the second method, it is naturally important to construct the apparatus with a good focusing characteristic, and for this purpose the Mattauch type mass spectrograph is best suited one. However, the Mattauch type needs a much larger magnet than other sector types for the same mean radius of ion beam, and consequently a highly accurate technique may be required for its construction. Mainly for economic reasons, the authors were obliged to adopt a 60° sector type magnet. However, the authors found it rather difficult to set the apparatus of the modified Bainbridge-Jordan type with the linear dimension required for their purpose because of the limited floor space of the laboratory.

In order to avoid such difficulties, a new type of double focusing mass spectrograph was constructed as described in this paper.

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