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The $Zr^{92}-Mo^{92}$ and $Zr^{94}-Mo^{94}$ Pairs and the $Zr^{96}-Mo^{96}-Ru^{96}$ Triplet

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Mass spectrographic measurements are reported of the following mass differences; $\frac{1}{2}W^{184}-Zr^{92}$, $\frac{1}{2}W^{184}-Mo^{92}$, $\frac{1}{2}Os^{188}-Zr^{94}$, $\frac{1}{2}Os^{188}-Mo^{94}$, $\frac{1}{2}Os^{192}-Zr^{96}$, $\frac{1}{2}Os^{192}-Mo^{96}$ and $\frac{1}{2}Os^{192}-Ru^{96}$. These results are used to check and supplement existing transmutation and disintegration data in the region of the $Zr^{92}-Mo^{92}$ and $Zr^{94}-Mo^{94}$ isobaric pairs and the $Zr^{96}-Mo^{96}-Ru^{96}$ isobaric triplet.

INTRODUCTION

THIS paper describes a mass spectrographic determination of the $Zr^{92}-Mo^{92}$, $Zr^{94}-Mo^{94}$, $Zr^{96}-Mo^{96}$, and $Ru^{96}-Mo^{96}$ mass differences. The information so obtained is useful in checking the correctness of certain transmutation and disintegration data and in estimating the energy available for unobserved reactions.

EXPERIMENTAL

The measurements herein reported were made by the doublet method using our large Dempster-type mass spectrograph.[1] The ion source was a high frequency spark. One electrode of the spark consisted of a thin-walled nickel tube which was packed with the elements to be studied. Exposure times ranged from 5–10 minutes The mass differences obtained in this way are shown in Table I.

The $Zr^{92}-Mo^{92}$ Stable Isobaric Pair

Nb^{92} has been found to decay to both Zr^{92} and Mo^{92}, in the former case by K-capture. There is a 0.93-Mev gamma-ray in the K-capture branch,[2] which represents all but 0.05 percent of the transitions, while the end point of the negatron group leading to Mo^{92} has been reported[3,4] to be 1.38 Mev.

It is possible to compute the energy available for the K-capture decay in two ways using existing transmutation data. The first calculation is based on the $Nb^{93}(\gamma,n)$ threshold[5] of 8.7±0.2 Mev, a Q[6] of 4.33±0.2 Mev for the $Zr^{92}(d,p)$ reaction, and values of 0.19[7] and 0.06[8] Mev for the end point of the Zr^{93} negatrons, *viz.*:

$$Nb^{93}-Nb^{92}=0.99964\pm21 \text{ amu};$$
$$Zr^{93}-Zr^{92}=1.00193\pm21 \text{ amu};$$
$$Zr^{93}-Nb^{93}=0.00013\pm\ 7 \text{ amu}.$$

Therefore,

$$Nb^{92}-Zr^{92}=0.00216\pm30 \text{ amu}=2.0\pm0.3 \text{ Mev}.$$

The second calculation is based on 2.5±0.2 Mev for the $Zr^{92}(p,n)Nb^{92}$ threshold,[9] and gives the value $Nb^{92}-Zr^{92}=1.7\pm0.2$ Mev. These two figures are in satisfactory agreement and appear consistent with a K-capture branch containing a 0.93-Mev gamma-ray. We shall assume a value of 1.8±0.2 Mev for the $Nb^{92}-Zr^{92}$ mass difference.

It is likewise possible to compute a value for the $Nb^{92}-Mo^{92}$ decay energy. The pertinent data are 8.7±0.2 Mev[5] for the $Nb^{93}(\gamma,n)$ threshold, 6.08±0.2 Mev[6] for the Q of the $Mo^{92}(d,p)$ reaction, 3.7±0.2 Mev[9] for the $Nb^{93}(p,n)Mo^{93m}$ (6.75 hr) threshold, and 2.5–2.7 Mev[10,11] for the $Mo^{93m}\rightarrow Mo^{93}$ transition. Thus,

$$Nb^{93}-Nb^{92}=0.99964\pm21 \text{ amu};$$
$$Mo^{93}-Mo^{92}=1.00005\pm21 \text{ amu};$$
$$Mo^{93m}-Nb^{93}=0.00309\pm21 \text{ amu};$$
$$Mo^{93m}-Mo^{93}=0.00279\pm10 \text{ amu}.$$

TABLE I. Mass spectrographic mass differences.

Nuclides	Mass difference mMU	Mev	Previous measurements
$\frac{1}{2}W^{184}-Zr^{92}$	69.79±0.14		74.1±1.8[a] 69.3±0.4[b]
$\frac{1}{2}W^{184}-Mo^{92}$	68.45±0.22		——
$Mo^{92}-Zr^{92}$ [c]	1.34±0.26	1.25±0.25	
$\frac{1}{2}Os^{188}-Zr^{94}$	71.34±0.12		——
$\frac{1}{2}Os^{188}-Mo^{94}$	72.56±0.16		73.1±2.8[a]
$Zr^{94}-Mo^{94}$ [c]	1.22±0.20	1.14±0.2	
$\frac{1}{2}Os^{192}-Zr^{96}$	71.83±0.24		——
$\frac{1}{2}Os^{192}-Mo^{96}$	75.46±0.14		72.8±2.9[a]
$\frac{1}{2}Os^{192}-Ru^{96}$	72.44±0.17		75.9±1.9[a] 73.4±1.3[d]
$Zr^{96}-Mo^{96}$ [c]	3.63±0.28	3.4 ±0.3	
$Ru^{96}-Mo^{96}$ [c]	3.02±0.22	2.8 ±0.2	

[a] A. J. Dempster, Phys. Rev. **53**, 64 (1938).
[b] Duckworth, Kegley, Olson, and Stanford, Phys. Rev. **83**, 1114 (1951).
[c] These values are computed from the other values which are the experimentally determined ones.
[d] A. C. Graves, Phys. Rev. **55**, 863 (1939).

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‡ Holder of a Research Council of Ontario Scholarship.
[1] H. E. Duckworth, Rev. Sci. Instr. **21**, 54 (1950).
[2] P. Preiswerk and P. Stähelin, Helv. Phys. Acta **24**, 300 (1951).
[3] Sagane, Kojima, Niyamoto, and Ikawa, Proc. Phys. Math. Japan **22**, 174 (1940).
[4] D. N. Kundu and M. L. Pool, Phys. Rev. **71**, 140 (1947).
[5] Sher, Halpern, and Mann, Phys. Rev. **84**, 387 (1951).
[6] J. A. Harvey, Phys. Rev. **81**, 353 (1951).
[7] G. E. Boyd and Q. V. Larson, Oak Ridge National Laboratory Report ORNL-685 (1950) (unpublished).
[8] E. P. Steinberg and L. E. Glendenin, Phys. Rev. **78**, 624 (1950).
[9] Blaser, Boehm, Marmier, and Scherrer, Helv. Phys. Acta **24**, 441 (1951).
[10] *Nuclear Data*, National Bureau of Standards Circular 499 (1950), p. 100.
[11] L. Ruby and J. R. Richardson, Phys. Rev. **83**, 698 (1951).

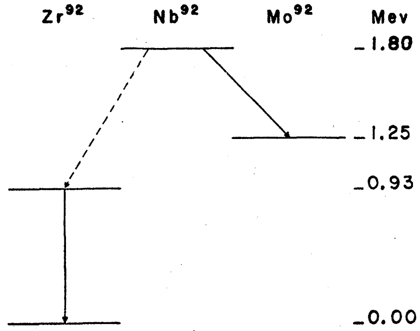

FIG. 1. Energetics at mass 92.

Therefore,

$$Nb^{92}-Mo^{92}=0.00011\pm38 \text{ amu}=0.1\pm0.4 \text{ Mev.}§$$

It appears from this result that the 1.38-Mev beta-ray is not a transition from the ground state of Nb^{92} to the ground state of Mo^{92}.

These calculations indicate that the $Mo^{92}-Zr^{92}$ mass difference is $(1.8\pm0.2)-(0.1\pm0.4)=1.7\pm0.4$ Mev. From our experiments, as seen from Table I, this difference has been found to be 1.25 ± 0.25 Mev. These results are in satisfactory agreement, especially when one considers that the computed values are based on a chain of several measurements, each of which is subject to an error of 0.1–0.2 Mev. The energy relationships between Zr^{92}, Nb^{92}, and Mo^{92} are shown in Fig. 1. No attempt has been made to fit the 0.1 ± 0.4 Mev $Nb^{92}-Mo^{92}$ mass difference into this figure.

The $Zr^{94}-Mo^{94}$ Isobaric Pair

The state of knowledge of Nb^{94} is shown in Fig. 2.[10] It is not possible to compute the $Zr^{94}-Mo^{94}$ mass difference from disintegration or transmutation data. From Table I it is seen that the measured $Zr^{94}-Mo^{94}$ mass difference is 1.14 ± 0.2 Mev. This leaves 0.12 Mev of energy available for the *K*-capture mode of decay from the ground state of Nb^{94}, a figure which explains the lack of success[12] with which experiments to detect it have met.

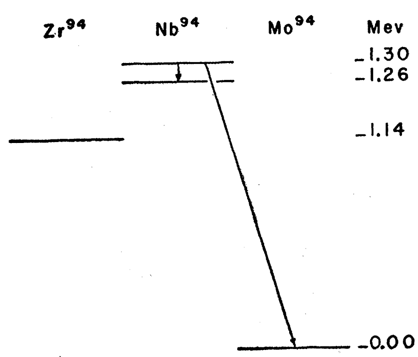

FIG. 2. Energetics at mass 94.

§ *Note added in proof:* Blaser *et al.* (see reference 9) have also made a calculation of this mass difference and have obtained substantially the same result.

[12] Hein, Fowler, and McFarlane, Phys. Rev. **85**, 138 (1952).

The $Zr^{96}-Mo^{96}-Ru^{96}$ Isobaric Triplet

Some knowledge of the $Zr^{96}-Mo^{96}$ mass difference may be obtained from the $Zr^{96}(p,n)$ threshold plus the various studies of the decay scheme of Nb^{96}.

Regarding the $Zr^{96}(p,n)$ threshold, this has been found by Blaser *et al.*[9] to be 2.6 ± 0.2 Mev. When this is compared to 2.5 Mev for the $Zr^{92}(p,n)$ threshold, obtained in the same laboratory, it is difficult to believe that both can be correct. One would expect the figure for Zr^{96} to be much lower than that for Zr^{92}. The latter has been seen above to be consistent with the other data at mass number 92. Consequently, we are assuming that the reported $Zr^{96}(p,n)$ value is to an excited state of Nb^{96}.

Concerning the $Nb^{96}-Mo^{96}$ decay, three values have been reported for the total decay energy: these are 3.14 Mev,[10] 3.16 Mev,[2] and 1.98 Mev.[13] Our value for the $Zr^{96}-Mo^{96}$ mass difference is 3.4 ± 0.25 Mev, which is larger than any of the figures for the $Nb^{96}-Mo^{96}$

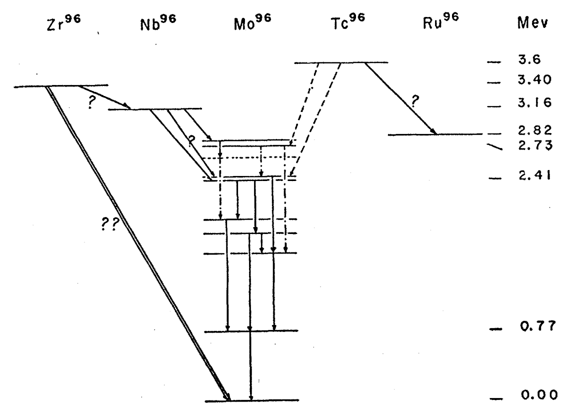

FIG. 3. Energetics at mass 96.

difference. This implies either that there is more energy in the Nb^{96} decay than is presently realized, or that Zr^{96} is unstable against beta-decay to Nb^{96}. The former possibility does not seem very likely since two recent independent measurements agree closely on the total decay energy for Nb^{96}. The second possibility, which is the more likely, is analogous to the case of Ca^{48} which does not decay to Sc^{48}, although energetically possible, because of the large spin change involved.[14] Further, it is interesting to note that McCarthy[15] has obtained preliminary evidence for an activity of 3.3–4.3 Mev in Zr^{96}. This activity, if authentic, would represent the double beta-decay of Zr^{96} to Mo^{96}.

Turning now to the $Ru^{96}-Mo^{96}$ pair, it is known that Tc^{96} decays to Mo^{96} by *K*-capture and possibly to Ru^{96} by negatron emission. In the former case, 2.73

[13] M. L. Pool (private communication).

[14] T. P. Kohman, Phys. Rev. **73**, 16 (1948); T. P. Kohman, Phys. Rev. **73**, 1223 (1948); J. W. Jones and T. P. Kohman, Phys. Rev. **85**, 941 (1952); D. Kurath, Phys. Rev. **87**, 528 (1952); Hammermesh, Hummel, Goodman, and Engelkemeir, Phys. Rev. **87**, 528 (1952).

[15] J. A. McCarthy (private communication, August 10, 1952).

Mev[16] of gamma-rays follow the K-capture event, while the negatron group, if it exists, has an end point of $\sim$0.8 Mev.[17,18] Both these data are consistent with our measurement of 2.8±0.2 Mev for the $Ru^{96}-Mo^{96}$ mass difference. Figure 3 shows the level schemes at mass number 96.

It is interesting to use our values for the $Zr^{96}-Mo^{96}$ and $Ru^{96}-Mo^{96}$ mass differences to construct at mass 96 the even-even parabola, which is predicted by the semi-empirical mass formula. This is done in Fig. 4, where it is compared to the parabola resulting from the use of the computed masses (with suitable vertical displacement) of Metropolis and Reitwiesner.[19]

The experimental parabola is seen to be wider than the predicted one. This also follows from the experiments of the Columbia group[20] who have found the sides of the valley of stability at constant Z (for $Z=32$ and $Z=34$) to be less steep than given by the computed masses.

Discussion of the Mo−Zr Mass Differences

In some cases at odd mass number, the Mo−Zr mass differences have been found by studying the decay schemes involved, *viz*: $Mo^{91}-Zr^{91}\geqslant 3.6$ Mev,[21]‖ $Zr^{95}-Mo^{95}=2.04$ Mev,[22] and $Zr^{97}-Mo^{97}=4.59$ Mev.[23] In Fig. 5, where these values are plotted together with our $Mo^{92}-Zr^{92}$, $Zr^{94}-Mo^{94}$, and $Zr^{96}-Mo^{96}$ results, the mass difference appears to be a linear function of the mass number.

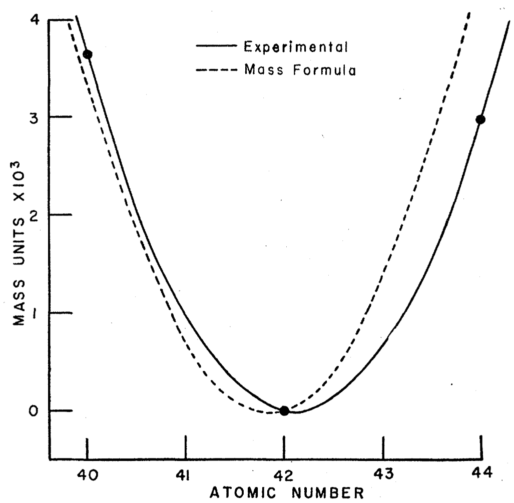

FIG. 4. The experimental and theoretical parabolae at mass 96. Assuming a form $({}_{Z}M^{A}-{}_{Z_0}M^{A})=\frac{1}{2}B(Z-Z_0)^2$, the parameters in the experimental case are $B=1.55$, $Z_0=42.1$, and in the theoretical case $B=1.94$, $Z_0=41.8$. *Note added in proof:* Charles D. Coryell, in a paper entitled "Beta Decay Energetics," to be published in Vol. II of *Annual Reviews of Nuclear Science*, gives in the region $70<A<90$, a curve of B *versus* A, derived from decay energies. This curve, when extrapolated to $A=96$, gives the value $B\sim1.5$, in good agreement with our experimental one.

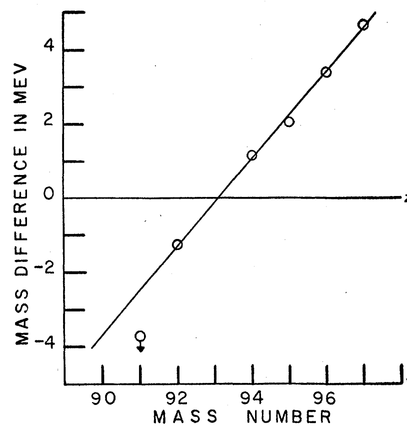

FIG. 5. A plot of the isobaric Zr−Mo mass differences *vs* mass number.

The widespread existence of linear relationships of this type has been pointed out to us by Dr. Katharine Way and Miss Marion Wood, and will, we understand, be described in detail by them in a future publication. The semi-empirical mass formula also predicts an approximately linear curve. We, therefore, regard Fig. 5 as an indication of the general correctness of our results.¶ The departure of the point at mass number 91 from the straight line curve is adequately explained by the extra energy involved in the $Mo^{91}-Nb^{91}$ transition, owing to the ease with which Mo^{91} is transformed into the 50 neutron configuration, Nb^{91}.

These results indicate that Zr^{93} and Mo^{93} are practically equal in mass. This is compatible with the fact that Zr^{93} has a half-life of $>4\times10^{6}$ years and Mo^{93} decays by K-capture with presumably a long lifetime.

The authors are grateful to Dr. Katherine Way and her colleague, Miss Marion Wood, both for sending us helpful preliminary reports on their beta-ray systematics in the mass 90–100 region, and for drawing to our attention pertinent experimental work. We also appreciate receiving prepublication accounts from John A. McCarthy of his studies of double-beta decay in Zr^{96}. These experiments have been generously supported by the National Research Council of Canada, the Research Council of Ontario, and the Research Corporation of New York.

[16] Medicus, Preiswerk, and Scherrer, Helv. Phys. Acta **23**, 299 (1950).
[17] Medicus, Mukerji, Preiswerk, and de Saussure, Phys. Rev. **74**, 839 (1948).
[18] J. E. Edwards and M. L. Pool, Phys. Rev. **72**, 384 (1947).
[19] N. Metropolis and G. Reitwiesner, *Table of Atomic Masses* (Argonne National Laboratory, Chicago, 1950).
[20] Geschwind, Minden, and Townes, Phys. Rev. **78**, 174 (1950), and S. Geschwind and R. Gunther-Mohr, Phys. Rev. **81**, 882 (1951).
[21] R. B. Duffield and J. D. Knight, Phys. Rev. **76**, 573 (1949).
‖ *Note added in proof:* A recent study of the 15.5-min Mo^{91} positrons has given an end point of 3.32±0.05 Mev (Leon Katz, private communication, September 13, 1952). The $Mo^{91}-Zr^{91}$ mass difference is, therefore, $\geqslant$4.4 Mev.
[22] J. S. Levinger, *Radiochemical Studies: The Fission Products* (McGraw-Hill Book Company, Inc., New York, 1951), Paper No. 94, National Nuclear Energy Series, Plutonium Project Record, Vol. 9, Div. IV; and C. Y. Fay, Phys. Rev. **81**, 300 (1951).
[23] Burgus, Knight, and Prestwood, Phys. Rev. **79**, 104 (1950).
¶ *Note added in proof:* In current mass spectrographic investigations of the Na−Sm isobaric differences at mass numbers 144, 148, and 150, we have found that a similar linear relationship appears to exist in this heavier region.