NEW DETERMINATIONS OF THE ATOMIC MASSES of Zn<sup>66</sup>, Zn<sup>67</sup>, Zn<sup>68</sup> and Ni<sup>58</sup> NEW DETERMINATIONS OF THE ATOMIC MASSES of Zn<sup>66</sup>, Zn<sup>67</sup>, Zn<sup>68</sup> and Ni<sup>58</sup>

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## A Thesis

Submitted to the Faculty of Arts and Sciences in Partial Fulfilment of the Requirements

for the Degree

Master of Science

McMaster University

October 1955

MASTER OF SCIENCE (1955) (Physics) McMASTER UNIVERSITY Hamilton, Onterio

TITLE: New Determinations of the Atomic Masses of Zn<sup>66</sup>, Zn<sup>67</sup>, Zn<sup>68</sup> and Ni<sup>58</sup>

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SCOPE AND CONTENTS:

Masses have been determined by means of new mass spectrographic comparisons for the nuclides Zn<sup>66</sup>, Zn<sup>67</sup>, Zn<sup>68</sup> and Ni<sup>58</sup>. The values obtained are compared with earlier ones from another laboratory. Mass differences have been obtained, which, together with those from nuclear reaction experiments, provide a basis for the adjustment of former mass values. These adjustments have lead to better agreement between mass determinations and nuclear disintegration data.

### ACKNOWLEDGMENT

I am very grateful to Professor H. E. Duckworth, who gave very helpful encouragement and advice during the course of this investigation and in the preparation of this thesis. I also thank Professor A. B. McLay for suggesting improvements in the manuscript. John T. Kerr gave much helpful advice concerning the experimental work, and Philip C. Eastman is thanked for his help in the measurement of doublet photographs.

Financial assistance in the form of a scholarship from the Shell Oil Company of Canada, and a summer research stipend from the National Research Council of Canada are gratefully acknowledged.

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### INTRODUCTION

F. W. Aston first pointed out the fact (1) that atomic masses are not integrally related. This was attributed to differences in nuclear binding energies of different nuclei. This binding energy is the energy which would be required to split the nucleus into its components, if such a process were possible. Since a nucleus possesses less mass than its individual nucleons, by determining its atomic mass, its binding energy can be found. This gives a measure of the stability of the nucleus.

In the study of nuclear properties which depend upon stability, mass determinations are very useful. When nuclear reactions take place, energy is lost or gained. A knowledge of the mass difference between the original and final nuclei, together with a knowledge of the particles emitted or absorbed, gives the energy release, or Q-value of the reaction. Predictions based on mass values can be made regarding the probability of certain reactions taking place. The mass difference between neighbouring isobars, giving as it does the total decay energy, is useful in establishing the details of a decay scheme. Mass differences are also useful in the study (2) of double beta-decay. This simultaneous emission of two beta-particles has been predicted for otherwise stable isobars, which are separated by two atomic number units. In these cases, the intervening nuclide is heavier than either.

Mass determinations support the theory (3) of nuclear shell structure. This theory attributes greater stability to nuclides possessing

certain numbers of protons and/or neutrons. Mass determinations show that the masses of these nuclides are lower (4) (5) than normal, and also that discontinuities (6) exist in the slope of the binding energy curve at points corresponding to these nuclides.

### MASS DETERMINATIONS AND TRANSMUTATION DATA

The earliest and most obvious way to determine the mass of an atom is to weigh it. Whereas objects on a macroscopic scale are weighed by observing the effect of the earth's gravitational field, ionized atoms are weighed by observing the effects of electric and magnetic fields. The mass spectroscopist causes ions to move in special fields, and compares the deflections which they experience with the deflection of a standard ion.

At the present time, accurate Q-values are available from studies of the energetics of nuclear decay and transmutation. As stated above, but conversely, energy released or absorbed in a transmutation gives a measure of the mass change. Thus atomic mass differences can be determined by nuclear reaction experiments. These determinations must of course be referred to standards to obtain atomic masses.

Mass spectroscopic determinations and reaction determinations do not always agree. There has been, until very recently (7), a bothersome discrepancy (8) between mass spectroscopic and transmutation masses for  $c^{12}$ . This has been quite serious, since  $c^{12}$  is widely used as a substandard in mass spectroscopy. Also, in the iron-nickel-zinc region, objectionable differences (9) have shown up between the masses determined by the two methods. This has brought suspicion upon mass spectroscopic data for these elements. With this problem in mind, the author has undertaken to redetermine some of the atomic masses in this region by means of new comparison ions. It was hoped that some of the discordant mass differences could be rectified.

#### EXPERIMENTAL

I. Mass spectrograph. The instrument used is a large Dempster type (10) mass spectrograph. It consists of a 90° electrostatic analyser, and a 180° magnetic analyser. It is shown schematically in figure 1. The symbols used are those of Herzog (11). The separation of the electrostatic analyser plates is 0.200 inches. Their mean radius of curvature is 7.9 inches. A potential difference between them of about 700 volts was maintained by batteries.

The magnetic field is located in a 0.125 inches gap between pole faces. The ions enter this field as a narrow beam of small energy spread, having been selected by the slit  $S_3$ , which follows the electrostatic analyser. This beam is focussed, after being deflected through  $180^\circ$ , onto a photographic plate. The radius of curvature of the path of the best focussed ions is 9.1 inches.

The accelerating voltage used in this investigation was about 14,000 volts. The magnetic field current was well stabilized electronically. A field of about 6000 cersteds was used. Two diffusion pumps backed by mechanical pumps, maintained the vacuum. The source and analyser regions were pumped separately. The usual operating pressure in the analyser was  $5 \times 10^{-6}$  mm. Hg. In some instances, this was substantially increased, in order to observe any effect produced.

The principle slit, S2, of the collimating system was adjusted to 0.001 inches. This corresponds to a resolution of 1 part in 7000.

II. Ion source. A small version of the type of ion source described by Shaw (12) was used to produce ions. It is illustrated in figure 2. It consists of a tubular crucible surrounded concentrically by a tungsten filament. The plane of the filament is slightly behind the open end of the crucible. The housing consists of a cylinder with a cap. There is a small hole in the cap directly in front of the open end of the crucible. The solid to be ionized is placed in the crucible. Gases can be ionized by leading them into the crucible region.

When the filament is hot, electrons of 500 volts energy bombard the crucible. With a current of a few milliamperes, sufficient heat is produced to volatilize many metals and salts. The electrons also ionize the vapor so formed, and any other gas in the vicinity.

The electron bombardment voltage also serves as a drawing-out voltage for the ions. The hole in the cap is lined up with the collimating slits for maximum ion current. The space in which the ions receive their acceleration is about 0.5 inches.

<u>III.</u> Doublet formation. A doublet is a pair of lines obtained on a photographic plate, which is due to two ion types of slightly different e/nratios. Thus, a singly charged atom of a certain mass will form a doublet with a doubly charged atom of double its mass number. The spacing of the doublet depends upon the packing fraction<sup> $\hat{n}$ </sup> difference between the two ion types. Since it is not difficult to achieve uniform dispersion over short distances, doublets are used to determine mass differences.

<sup>th</sup>The packing fraction of an atom is defined as  $\frac{M-A}{A} \ge 10^4$  where M is its atomic mass and A, the nearest integer to M, is the mass number.

With the Shaw-type source described, doublets could be obtained quite readily. The number of ions formed of any element depends upon the amount of that element present in the gaseous state, and the electron current used. Thus, if a gas at a certain pressure is introduced, the ion current should vary almost linearly with the electron current.

In the case of ions produced from a solid in the crucible, the ion current depends upon a higher power of the electron current. The electrons must provide energy to vaporize the solid before it can be ionized. Thus, the ion current does not begin until the electron current is appreciable, and then rises more sharply than with gases. It is obvious that a condition can be reached when the "gaseous" ion current, and the "solid" ion current are equal in intensity.

Individual doublets with solid and gaseous elements can be matched in intensity in this way. This is very important, especially when photographic recording is used. The peak intensity of a line may not exactly correspond to its center, and errors in measurement of the doublet spacing could result.

#### THE DOUBLETS

## I. Mass 66

At mass 66 a doublet was formed with singly charged Zn<sup>66</sup> and doubly charged Xe<sup>132</sup>. From many doublet photographs, eight were chosen as being suitable for measurement. The reference distance used to determine the mass dispersion was the distance between the Xe<sup>132</sup> and Xe<sup>134</sup>lines. II. Mass 67

Singly ionized Zn<sup>67</sup> and doubly ionized Xe<sup>134</sup> form a doublet at mass 67. Ten doublet photographs were measured. The reference distance was the same as that taken for the doublet at mass 66.

#### III. Mass 68

Singly charged Zn<sup>68</sup> and doubly charged Xe<sup>136</sup> form a doublet at mass 68. Nine doublet photographs were found satisfactory and were measured. The reference distance taken was that between the Xe<sup>134</sup> and Xe<sup>136</sup> lines.

## IV. Mass 58

Singly charged Ni<sup>58</sup> formed a doublet with the fragment C<sub>3</sub>H<sub>6</sub>O. This was quite a broad doublet. The dispersion distance used was that between the C<sub>3</sub>H<sub>6</sub>O and C<sub>4</sub>H<sub>9</sub> lines. Twelve doublet photographs were measured. Some exposures were made at higher than normal pressure, in order to determine whether or not this factor would affect the doublet spacing.

#### MASS DIFFERENCES

Table I gives the mass differences determined in the course of this investigation.

| Mass Differ                         | rences           | (millimass units) |      |
|-------------------------------------|------------------|-------------------|------|
| ixe <sup>132</sup> - ₂              | 66<br>Zn         | 25.61 ±           | 0.15 |
| ixe <sup>134</sup> - 2              | 2n <sup>67</sup> | 25.25 ±           | 0.20 |
| ixe <sup>136</sup> - 2              | 3n <sup>68</sup> | 27.70 ±           | 0.20 |
| C <sub>3</sub> H <sub>6</sub> 0 - 1 | 11 58            | 106.23 ±          | 0.20 |

TABLE I

The xenon masses in the calculation of these differences are based on xenon-hydrocarbon differences by Halsted (13), and recent carbon and hydrogen values by Mattauch and Bieri (14). The xenon masses calculated and used are  $Xe^{132} = 131.94605 \pm 15$ ,  $Xe^{134} = 133.94778 \pm 12$ and  $Xe^{136} = 135.95020 \pm 10$  A.M.U. The new masses calculated are listed in table II.

## TABLE II

NEW MASS VALUES (Atomic mass units)

| Zn <sup>66</sup> | 65.94737 | +   | 15 |
|------------------|----------|-----|----|
| Zn <sup>67</sup> | 66.94857 | + 8 | 20 |
| Zn <sup>68</sup> | 67.94740 | + - | 20 |
| N1 <sup>58</sup> | 57.95411 | + + | 20 |

### DISCUSSION OF DATA

The values obtained for the nuclides studied can be compared with those based on differences obtained by Collins et al. (15). This is done in Table III. Mattauch and Bieri's new carbon and hydrogen masses were used to recalculate Collins' zinc and nickel masses.

## TABLE III

#### COMPARISON OF MASS VALUES

| Nuclide          | Collins et al. | Author        | Discrepancy x 10 <sup>5</sup> |
|------------------|----------------|---------------|-------------------------------|
| Zn <sup>66</sup> | 65.94712 ± 5   | 65.94737 ± 15 | + 25 ± 16                     |
| Zn <sup>67</sup> | 66.94806 ± 6   | 66.94857 ± 20 | + 51 ± 21                     |
| Zn <sup>68</sup> | 67.94677 ± 6   | 67.94740 ± 20 | + 63 ± 21                     |
| N1 <sup>58</sup> | 57.95337 ± 9   | 57.95411 ± 20 | + 74 ± 22                     |

The discrepancies in the zinc masses might be due to an error occurring systematically throughout the xenon determinations of Halsted, or a systematic error in this investigation. In either case, it is expected that the zinc-zinc differences should be satisfactory.

The mass differences between the zinc isotopes are given accurately by  $\gamma$ , n (16) and n,  $\gamma$  (17) reactions. These may be compared with differences between mass values. Table IV shows this comparison.

The mass difference found for  $2n^{68} - 2n^{67}$  agrees with both nuclear data and previous mass determinations. The value found for the  $2n^{67}$  -  $2n^{66}$  difference lies midway between nuclear data and the determinations

of Collins. However, the nuclear data involved here are considered quite reliable, so that it is satisfying to see that this investigation has yielded better agreement in the case of the  $2n^{67} - 2n^{66}$  difference than did earlier work.

#### TABLE IV

#### COMPARISON OF MASS DIFFERENCES

#### WITH TRANSMUTATION DATA

| Nuclides                            | Trans. Data  | Collins et al. | Author       |
|-------------------------------------|--------------|----------------|--------------|
| $2n^{67} - 2n^{66}$                 | 1.00147 ± 21 | 1.00094 ± 8    | 1.00120 ± 22 |
| Zn <sup>68</sup> - Zn <sup>67</sup> | 0.99877 ± 10 | 0.99871 ± 9    | 0.99883 ± 24 |

The weighted mean of the nuclear value and the result of this work is 1.00135  $\pm$  16 A.M.U. for the  $2n^{67} - 2n^{66}$  difference, indicating that the Collins value for either  $2n^{66}$  or  $2n^{67}$  must be adjusted by about 0.4 m.M.U. Since the Collins' value for  $2n^{68} - 2n^{67}$  gives good agreement, it is suggested that the  $2n^{66}$  value should be reduced by 0.4 m.M.U. This change has the merit that it makes all the discrepancies between the masses of Collins and the author approximately equal, which would be the case if the errors were really systematic. Also, the disagreement (9) with nuclear data in the case of the  $2n^{66} - Cu^{65}$  difference would be corrected.

A serious discrepancy also exists between the nickel masses of Collins and the author. This is so far unexplained. However there is reason to believe (9) (18) that the accepted masses of all the isotopes of nickel have been too low by about 0.6 millimass units.

Some attempts were made to determine whether or not the operating

pressure in the mass spectroscope could influence doublet spacings and hence mass determinations. No effect was observed in the case of the Zn - Xe doublets, but there was some indication that the nickel doublet might suffer from such an effect. Table V illustrates the results of this part of the investigation. The numbers given are the ratios of the doublet spacings to the dispersion distances for the different pressure ranges.

## TABLE V

## RESULTS OF PRESSURE VARIATIONS

(pressure in units of 10<sup>-6</sup> mm.Hg.)

| Doublet  | C <sub>3</sub> H <sub>6</sub> 0 − Ni <sup>58</sup> |                                    | <sup>132</sup> - 2n <sup>66</sup> |                                   |
|----------|--|------------------------------------|-----------------------------------|-----------------------------------|
| pressure | (5 <b>&lt;</b> p <b>&lt;</b> 15)                   | ( 50 <b>&lt;</b> p <b>&lt;</b> 60) | (3 <b>&lt;</b> p <b>&lt;</b> 6)   | (10 <b>&lt;</b> p <b>&lt;</b> 14) |
| mean     | 0.10911+15   | 0.10763±22                         | 0.02576_8                         | 0.02570111                        |
| extremes | 0.10825  | 0.10627                            | 0.02518                           | 0.02494                           |
|          | 0.11025  | 0.10908                            | 0.02631                           | 0.02640                           |

The errors quoted in this table are purely statistical in nature, and do not include any systematic errors. They are therefore smaller than those quoted in Table I.

At present, work is continuing along this line in order to provide more conclusive results.

# BIBLIOGRAPHY

| 1.  | F. W. ASTON, Phil. Mag. 45, 941 (1923).   |
|-----|---|
| 2.  | B. G. HOGG and H. E. DUCKWORTH, Phys. Rev. <u>86</u> , 567 (1952).                                    |
| 3.  | J. M. BLATT and V. F. WEISSKOPF, Theoretical Nuclear Physics,<br>John Wiley and Sons, New York, 1952. |
| 4.  | B. G. HOGG and H. E. DUCKWORTH, Can. J. Phys. 31, 942 (1953).   |
| 5.  | B. G. HOGG and H. E. DUCKWORTH, Can. J. Phys. 32, 65 (1954).  |
| 6.  | H. E. DUCKWORTH, Nature, 170, 158 (1952).   |
| 7.  | Third Annual Conference in Mass Spectrometry, San Francisco,<br>May 22-27, 1955.                      |
| 8.  | H. E. DUCKWORTH, B. G. HOGG and E. M. PENNINGTON, Rev. Mod. Phy.<br>26, 463 (1954).                   |
| 9.  | J. T. KERR, J. G. V. TAYLOR and H. E. DUCKWORTH, (in press).  |
| 10. | H. E. DUCKWORTH, Rev. Sci. Instr. 21, 54 (1950).  |
| 11. | R. HERZOG, Zeits. f. Physik, 89, 447 (1934).  |
| 12. | A. E. SHAW, Phys. Rev. <u>75</u> , 1011 (1949).   |
| 13. | R. E. HALSTED, Phys. Rev. 88, 666 (1952).   |
| 14. | J. MATTAUCH and R. BIERI, Z. Naturforsch, 9a, 303 (1954).   |
| 15. | COLLINS, NIER and JOHNSON, Phys. Rev. 86, 408 (1952).   |
| 16. | SHER, HALPERN and MANN, Phys. Rev. 84, 387 (1951).  |
| 17. | B. B. KINSEY and BARTHOLOMEW, Can. J. Phys. 31, 901 (1953).   |
| 18. | A. H. WAPSTRA, Physica, 21, 385 (1955).   |



FIG. I.



Legend

- c crucible
- f filament
- g gas inlet
- h housing i insulator
- k-cap
- I leads
- p-focussing plate
- s crucible support t glass tube

FIG. 2.