JULY 1 AND 15, 1942 PHYSICAL REVIEW VOLUME 62

New Packing Fractions and the Packing Fraction Curve

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(Received May 15, 1942)

Descriptions and data are given of a number of mass comparisons (Ti-O, Cu-O, Cu-Ti, Cr-Ag, Mn-Ag, Fe-Ag, Cu-Zn, Cu-Mo, Cu-Pt-Ir, Cu-Au, Zn-Au, Pt-Au). These, together with other recent data, are critically analyzed in order to obtain values for the packing fraction curve. A curve is shown in which packing fractions are plotted against atomic number (instead of mass number), and the rule is suggested that the packing fraction of an element with odd atomic number is algebraically larger than the average of the two adjacent elements with even atomic number. This implies that elements with even atomic number are more stable than odd-numbered elements and, also, supports the theory that protons in the nucleus are associated together in groups of two.

INTRODUCTION

IN 1935 Professor A. J. Dempster described a double-focusing mass spectrograph[1] in which the ion source consisted of a high frequency spark between metallic electrodes.[1,2] With this type of source, in 1938, he was able to measure the packing fractions of many of the heavier elements[3,4] which had hitherto not been analyzed in a mass spectrograph. The data obtained in these investigations filled in many gaps in Aston's packing fraction curve[5] and suggested the general shape of the whole curve.[4] In 1939, Graves[6] used the same apparatus to examine the mass defects of an additional group of elements. The writer has also employed Dempster's mass spectrograph in further measurements of packing fractions among the heavier isotopes.

EXPERIMENTAL DETAILS

I. The Ion Source

It was discovered by Dempster[1,2] and further demonstrated by Sheng-Lin Ch'u[7] that a high frequency spark between metal electrodes is an abundant source of singly- and multiply-charged ions. It is thus a simple matter to obtain positive ions of any metal provided a rod composed of the desired metallic element is used as one of the electrodes. It will also frequently happen that a non-metallic ion can be obtained from an electrode in which it is present as an impurity. For example, a certain sample of electrolytic calcium has been found to provide a satisfactory source of chlorine. In dealing with other non-metallic elements, the oxide of the desired element has been packed with a suitable reducing agent in a small nickel tube which serves as the electrode. There is also ample evidence that gases present in the spark chamber are ionized by the spark and contribute positive ions to the total ion beam.[3,7] For many elements it has been found that a rod of the pure element is not as satisfactory for electrodes as is an alloy. The noble metals, gold and silver, produce a steady spark with little sputtering and by heating with an oxygen flame can be persuaded to "dissolve" suitable amounts of the desired element. If moderate quantities of another metal are added to gold the alloy spark apparently retains the desirable features of the gold spark.

The electrodes were adjusted by means of sylphons and were arranged so that four different combinations of electrodes could be employed without opening the vacuum system. Since it only took a couple of minutes to adjust the electrodes so that the spark occurred between a different pair, the ion intensity from two alloys containing differing amounts of the desired element could be compared under identical pressure and field conditions.

The ion after emission in the spark passed

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[1] A. J. Dempster, Proc. Am. Phil. Soc. **75**, 762 (1935).

[2] A. J. Dempster, Rev. Sci. Inst. **7**, 46 (1936).

[3] A. J. Dempster, Phys. Rev. **53**, 64 (1938).

[4] A. J. Dempster, Phys. Rev. **53**, 869 (1938).

[5] F. W. Aston, *Mass Spectra and Isotopes* (Longmans, Green and Co., 1933), p. 167.

[6] Alvin C. Graves, Phys. Rev. **55**, 863 (1939).

[7] Sheng-Lin Ch'u, Phys. Rev. **50**, 212 (1936).

through a small aperture in a diaphragm which was at high positive potential. It was then accelerated for a distance of about 3 mm before entering the collimating slit system.

II. The Mass Spectrograph

A complete description of the mass spectrograph has been given by Dempster.[1, 8] In the present investigation the ion beam was collimated by two slits of width 0.14 mm and 0.04 mm, the first being approximately 6 mm from the ion source and the second 20 mm behind the first. After a deflection of 90° in an electrostatic field between quadrant condenser plates, the collimated beam enters a magnetic field where it is bent through 180° and the ions possessing the same values of e/m are brought to a focus. This focusing for a small range of e/m permits mass comparisons of ions whose values of e/m are nearly the same. In some experiments a slit of 1.8 mm was inserted between the electrostatic and magnetic fields in order to select a fairly narrow energy range; in other trials the deflecting field was reduced until most of the ions struck the deflecting condenser plate and only those with least energy passed along the quadrant path. This had the same effect as inserting a slit and worked very well.

III. Current and Voltage Sources

The source of high frequency current needed for the ion source was a simple oscillator with frequency of about 10^6 oscillations per second. The intensity of the spark was controlled by varying the current in the primary side of the transformer. The accelerating potential of 4000–6000 volts was obtained from a transformer and rectified by a kenotron and condenser. The electrostatic deflecting potential of 600–800 volts was supplied by comparatively new B-batteries. The magnetic field of 4000–9000 oersteds required a current of 2–8 amperes which was obtained from lead storage cells.

IV. Sharpness of Lines

Since the accuracy of the measurements depends directly upon the sharpness and symmetry of the mass spectrum lines, it is pertinent to mention the various factors which determine these qualities. Most obvious of these factors is the stability of the electrostatic and magnetic fields. Since the B-batteries were in good condition and the charged plate was connected to one side of a 2-microfarad condenser, the other side being grounded as was the uncharged plate, the electrostatic field due to the batteries was probably quite steady. On the other hand the magnetic current slowly dropped, thereby setting a premium on short exposures. For this reason the magnetic field was generally set at a near-saturation value so that a slight decrement in the field current would produce no appreciable change in the field strength. In order to permit short exposures, as required by the decreasing magnetic field, a fairly large spark current was employed and consequently a strong ion beam obtained. When this beam entered the space between the condenser plates it probably distorted the field somewhat, the distortion depending upon the number of ions constituting the beam. Since the spark current is not altogether steady there is a continual variation in the intensity of the ion beam and hence a continual changing of the distorted electrostatic field. From the point of view of constant electrostatic field, then, the ion current should be kept low, while from magnetic field considerations the ion current should be large: clearly a compromise was necessary. Furthermore, under the impact of positive rays, the deflecting plates may become coated with non-conducting layers and local surface charges collect,[3, 9] while dust particles lodged on the plates may likewise create local field distortions.[3]

Another important factor is the type of plates used for photographing the lines. During the early part of the investigation Eastman I. O. U. V. Sens. plates were used and some of the measurements were made from these. Later, an order from Adam Hilger, Ltd., London, England, for Schumann plates was unexpectedly filled and they were thereafter used exclusively for final measurements. They are undoubtedly finer-grained than the Eastman plates and considerably more sensitive, enabling shorter exposures to be made.

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[8] A. J. Dempster, Phys. Rev. **51**, 67 (1937).

[9] R. Lariviere Stewart, Phys. Rev. **45**, 488–90 (1934).

The pressure conditions necessary for satisfactory photographs are happily, not too exacting. As previously reported by Dempster,[3] a pressure of 2×10^{-5} mm of mercury has usually been sufficient to obtain good lines.

The lines were measured with a Gaertner comparator whose accuracy had been checked.[6] The scale on the instrument measured directly to thousandths of a millimeter and could be estimated to ten-thousandths. Since the average result of five measurements of a line was used in the calculations, and settings could generally be reproduced to within 0.001 mm, errors introduced in the measuring process can be considered negligible as compared to other experimental errors.

THEORY OF CALCULATIONS

Dempster has shown[3] that the following relations exist. The position of the mass spectrum line on the photographic plate is given by $M^{\frac{1}{2}}=a+cx$, where M is the mass of the ion divided by the number of charges it carries, x is the distance on the plate measured from some fiducial point, while a and c are constants. In practice the constant c is obtained by using two of the heavier isotopes as a mass scale and assuming that their difference is an integral number of mass units. If one knows the dispersion constant c, other isotopes having approximately the same values of e/m can be compared with the two standards. Thus if M_1 and M_2, isotopes two units apart, be used as the mass scale, an unknown mass M, appearing on the mass spectrum somewhere between M_1 and M_2 or even a short distance outside this space, may be compared with the standards by the relation—

$$\Delta f=\frac{[(I_1)^{\frac{1}{2}}+a]^2-I}{I},$$

where

$$a=\frac{x-x_1}{x_2-x_1}[(I_2)^{\frac{1}{2}}-(I_1)^{\frac{1}{2}}]=\frac{x-x_1}{x_2-x_1}[(I_1+2)^{\frac{1}{2}}-(I_1)^{\frac{1}{2}}],$$

I, I_1, and I_2 being the integers corresponding to M, M_1, and M_2, and Δf being the difference between the packing fraction of M and a suitable average of M_1 and M_2. For example, $\Delta f=f-\frac{1}{2}(f_1+f_2)$, $f-\frac{1}{3}(f_1+2f_2)$, or $f-\frac{1}{4}(f_1+3f_2)$ depending upon whether the unknown line M is one-half, two-thirds, or three-quarters of the distance from M_1 to M_2.

As Dempster has pointed out,[3] the error in the value of f introduced by assuming integral spacing of the heavier isotopes will be much less than 0.1×10^{-4} unless there is a considerable departure from the integral spacing assumption. Since the two isotopes constituting the mass scale will generally be isotopes of the same element it is probable that the mass scale error will be well within the experimental error. Some values of the departure from integral spacing necessary to produce an error of 0.1×10^{-4} in the packing fraction will be given.

DESCRIPTION OF COMPARISONS

Titanium-Oxygen

With two titanium electrodes and also with one titanium and one copper electrode, doublets of O^{16} and triply-charged Ti^{48} were obtained with exposure times of 40–120 seconds. A much shorter exposure time would have been sufficient had it not been necessary to use Ti^{47} and Ti^{49} as the mass scale: Ti^{47}, the more abundant of the pair, is about one-tenth as abundant as Ti^{48}. Altogether thirteen photographs of the doublet were obtained and from this number five were selected as being superior to the remainder. Measurement of these five gave an average value for the packing fraction of Ti^{48} of -7.60 ± 0.07,[10] corresponding to mass 47.9635 ± 0.0004. The average value of the thirteen doublets was -7.71 ± 0.15, in good agreement with the value obtained from the best five. From these same plates it was possible to measure the difference between the packing fraction of oxygen and the average packing fraction of Ti^{47} and Ti^{49}. The five best photographs gave for this pair a value of $\bar{f}=-7.37\pm 0.15$.

These results agree well with those obtained last year by T. Okuda and K. Ogata[11] who obtained a discharge through titantetrabromide and measured the weights of the five titanium isotopes with a Bainbridge-Jordan type mass spectrograph. They report packing fractions

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[10] Throughout the paper the factor $\times 10^{-4}$ will be understood and not written.

[11] T. Okuda and K. Ogata, Phys. Rev. **60**, 690 (1941).

of $\mathrm{Ti}^{46} = -7.36 \pm 0.21$, $\mathrm{Ti}^{47} = -7.50 \pm 0.20$, $\mathrm{Ti}^{48} = -7.64 \pm 0.10$, $\mathrm{Ti}^{49} = -7.19 \pm 0.11$, and $\mathrm{Ti}^{50} = -7.54 \pm 0.08$, from which we can calculate $\bar{f}$ of Ti^{47} and Ti^{49} as -7.34 ± 0.16.

The assumption that Ti^{47} and Ti^{49} are two mass units apart is here clearly justified: it would require a departure of 0.025 mass unit from this supposition to introduce an error of 0.1×10^{-4} in the packing fraction while the divergence from integral spacing is only 0.004 mass unit between Ti^{46} and Ti^{50}!

Copper-Oxygen

This is a very important comparison because copper serves as one of the links between the standard O^{16} and the heavier isotopes. Two copper rods were used as electrodes and photographs obtained in which quadruply-charged copper ions at mass 15.75 and 16.25 bracketed the singly-charged O^{16} atom. For other photographs a copper electrode was used with a titanium electrode. Exposure times varied from 90–240 seconds for the sixteen measurable plates which were obtained. Unfortunately all these photographs were second-rate—either the copper lines were too faint or the oxygen line was over-exposed. Taking the mean of these sixteen we get the average packing fraction of Cu^{63} and Cu^{65} to be $\bar{f} = -7.67 \pm 0.35$: individual values ranged from a high of -6.68 to a low of -8.87. The only previous measurement of copper was made by Dempster,[3] who found $\bar{f} = -6.9 \pm 0.2$ for three photographs.

Unless a very high vacuum is obtained the oxygen line will probably always be more intense than the quadruply-charged copper lines. It is possible that a suitable source of molecular oxygen ions may be found, in which case doubly-charged copper ions can be employed with shorter exposure time. A calcium electrode which had been exposed to air for three weeks, thereby accumulating a layer of CaO on its surface, has proved to be a promising source of molecular oxygen and may make this comparison possible.

Copper-Titanium

With copper and titanium electrodes a comparison was sought between triply-charged titanium ions and quadruply-charged copper ions, the titanium ions appearing at mass 15.67, 16.00, 16.33 and the copper at 15.75 and 16.25. However, the titanium vaporized so rapidly that it soon coated the copper and the spark yielded only titanium ions. In spite of the faintness of the copper ions, the best photograph was measured and it gave $\bar{f}$ of $\mathrm{Ti}^{47}-\mathrm{Ti}^{48}$ greater than the packing fraction of Cu^{63} by 0.4 and $\bar{f}$ of $\mathrm{Ti}^{48}-\mathrm{Ti}^{49}$ greater than the packing fraction of Cu^{65} by 0.9. The amounts of these differences are not very significant but it is important to note that according to these results copper lies lower on the packing curve than does titanium. More will be said about this later.

Chromium-Silver

With one electrode of nichrome and the other of silver, with exposures of one hundred seconds, doubly-charged silver ions at mass 53.5 and 54.5 were compared with singly-charged chromium ions at 53 and 54. Here it was possible both to use the chromium isotopes as the mass scale and to regard the silver isotopes as the unknowns, and to employ the separation between the silver lines as the mass scale and calculate the chromium values. From seven photographs, selected from a total of fourteen, the packing fraction of Ag^{107} was calculated to be 2.35 ± 0.10 greater than $\bar{f}$ of $\mathrm{Cr}^{53}-\mathrm{Cr}^{54}$, and $\bar{f}$ of $\mathrm{Ag}^{107}-\mathrm{Ag}^{109}$ greater than f of Cr^{54} by 2.38 ± 0.05. Using Dempster's[3] value for silver, $\bar{f} = -4.95 \pm 0.5$, we calculate the packing fraction of Cr^{54} to be -7.33 ± 0.55. Since nichrome contains 20 percent iron and 15 percent chromium, it might be suspected that the line at mass 54 is due to both Cr^{54} and Fe^{54}. However, the faintness of the iron isotope at mass 56, which is fourteen times as abundant as Fe^{54}, indicated that the effect of this latter isotope could be disregarded.

Iron-Silver

With exposures ranging from twenty to one hundred seconds, a spark between one electrode of shim steel and the other of silver yielded satisfactory doubly-charged silver lines at mass 53.5 and 54.5, bracketing an iron isotope at mass 54. Out of 22 photographs the nine best were selected for measurement, giving $\bar{f}$ of

$Ag^{107}-Ag^{109}$ greater than the packing fraction of Fe^{54} by 2.42±0.05. With Dempster's value for silver we find the packing fraction of Fe^{54} is -7.37 ± 0.55. This comparison has been previously made by Dempster[3] who obtained -7.5 ± 0.8 for two photographs.

Manganese-Silver

In the chromium-silver comparison, on a number of plates, the manganese line at mass 55 was sharp enough to measure with respect to the doubly-charged silver lines. Five photographs were measured and an average packing fraction difference of 2.03±0.10 found. Combining this with Dempster's $\bar{f}$ of silver, one finds the packing fraction of Mn^{55} to be -6.98 ± 0.60.

Copper-Zinc

It will be recalled that copper has two isotopes, of mass number 63 and 65, which alternate with two of the more abundant zinc isotopes at 64 and 66. Since both these metals vaporize rapidly, especially the zinc, the ion emission is unsteady and diffuse lines result if the pure metals are used as electrodes. It was found expedient to make alloys of gold, copper, and zinc which were predominantly gold and hence possessed those merits of gold electrodes which have been previously mentioned. With exposures of twenty to fifty seconds, sometimes between two such alloys electrodes and other times between one alloy and platinum, several photographs were secured in which Zn^{64} could be measured in terms of $\bar{f}$ of $Cu^{63}-Cu^{65}$, and Cu^{65} could be measured in terms of f of $Zn^{64}-Zn^{66}$. As a rule the copper lines were over-exposed, particularly the more abundant Cu^{63}. Consequently, only one photograph of the $Cu^{63}-Zn^{64}-Cu^{65}$ group was used while four measurements were made of the $Zn^{64}-Cu^{65}-Zn^{66}$ comparison. In the first case the packing fraction of Zn^{64} was 1.01±0.4 smaller than $\bar{f}$ of $Cu^{63}-Cu^{65}$, and in the second case f of Cu^{65} was 1.20±0.15 greater than $\bar{f}$ of $Zn^{64}-Zn^{66}$.

Copper-Molybdenum

An unsuccessful attempt was made to compare singly-charged silicon ions with triply-charged zirconium by using silicon metal in a nickel tube as one electrode and a mixture of zirconium oxide and calcium in a nickel tube as the other. With exposures of two to four minutes, however, triply-charged molybdenum ions appeared at mass 30.67, 31.33, 31.67, 32.0, 32.33, 32.67, and 33.33, and doubly-charged copper ions of similar intensity at mass 31.5 and 32.5. It is probable that the molybdenum was present as an impurity in the zirconium oxide and the copper was an impurity in the nickel tube. Since zirconium and molybdenum both possess isotopes with mass number 92, 94, and 96, the lines corresponding to those mass numbers were attributed to both elements and were not used for measurement. With three photographs Cu^{63} was compared to $\bar{f}$ of $Mo^{95}-Mo^{97}$ and on four photographs Cu^{63} was measured in relation to $Mo^{95}-Mo^{98}$. The first set of measurements gave the packing fraction of Cu^{63} as 2.14±0.4 less than $\bar{f}$ of $Mo^{95}-Mo^{97}$, and the second set indicated f of Cu^{63} was 2.31±0.4 less than f of $Mo^{95}-Mo^{98}$. These $\bar{f}$'s of molybdenum are clearly not arithmetic means but possess similar values since the divergence from integral spacing is almost certainly small.

Copper-Platinum-Iridium

Platinum electrodes were made by tightly rolling platinum foil and fitting the roll into a nickel tube of small diameter. When two such tubes were used as electrodes, with exposures of forty to ninety seconds, sharp lines were obtained of singly-charged copper and triply-charged platinum and iridium. As can be seen in Plate I of Fig. 1, the more abundant platinum isotopes were more intense than the mass scale copper lines, while the less abundant platinum and the two iridium isotopes were approximately equal in intensity to the copper. The doublet at mass 65 due to Cu^{65} and Pt^{195} was clearly resolved and easily measured. From twelve photographs the packing fraction of Pt^{195} was calculated to be 10.74±0.20 greater than f of copper. Other measurements from these plates were: Ir^{191}—$\Delta f=10.55\pm0.05$ (14 photographs), Ir^{193}—$\Delta f=10.58\pm0.05$ (14 photographs), Pt^{192}—$\Delta f=10.53\pm0.15$ (6 photographs), Pt^{194}—$\Delta f=10.49\pm0.15$ (12 photographs), and Pt^{196}—$\Delta f=10.80\pm0.15$ (8 photographs).

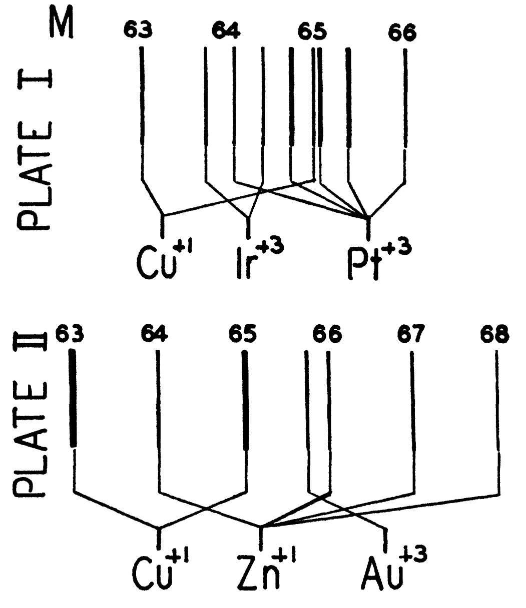

FIG. 1. Plate I shows a typical photograph of the Cu-Pt-Ir comparison. Plate II is a sample plate of the Zn-Au comparison: the two copper lines are present but are too broad to measure.

The previous value of Δf for $\mathrm{Cu}^{65}-\mathrm{Pt}^{195}$, published by Dempster[3] in 1938, was 8.93±0.1. In his photographs the copper lines are much heavier than the Pt^{195}, the opposite of the situation existing in this comparison. Perhaps an error in measurement, introduced by the extra intensity of the Pt^{195} line, was responsible for the Δf of the $\mathrm{Cu}^{65}-\mathrm{Pt}^{195}$ comparison being larger than the Δf between copper and the average of the remaining platinum isotopes. Since poor copper lines could invalidate all the measurements, it is important to observe in Plate I that they are perhaps the sharpest of the group.

In order to check these values, Cu^{63} and Cu^{65} were measured individually, by using the separation between Ir^{191} and Ir^{193} as the mass scale. Eight measurements showed f of Cu^{63} to be 10.49±0.1 lower than f of $\mathrm{Ir}^{191}-\mathrm{Ir}^{193}$, and f of Cu^{65} 10.75±0.1 lower than f of $\mathrm{Ir}^{191}-\mathrm{Ir}^{193}$. In addition, Cu^{65} was measured on eight plates, with Pt^{194} and Pt^{196} as the mass scale. It was found that f of Cu^{65} was 10.62±0.1 lower than the average of the packing fractions of those two isotopes. The consistency of these measurements suggested that the copper lines were being accurately located with the measuring instrument, thereby eliminating the most serious possibility of error.

Copper-Gold

Triply-charged gold ions at mass 65.67 appeared with singly-charged copper ions at mass 63 and 65 when the gold-copper-zinc alloy was used with a platinum electrode. Eight photographs, with exposures of forty to sixty seconds, gave f of gold to be 9.82±0.4 greater than f of $\mathrm{Cu}^{63}-\mathrm{Cu}^{65}$. The copper lines were somewhat diffuse and it is for this reason that the probable error has been given a large value.

Zinc-Gold

For this comparison both electrodes were made of the gold-copper-zinc alloy and exposure times ranged from forty to sixty-five seconds. The gold line at 65.67 and the zinc lines at 64, 66, 67, and 68 possessed comparable intensities so that by successively using $\mathrm{Zn}^{64}-\mathrm{Zn}^{66}$, $\mathrm{Zn}^{64}-\mathrm{Zn}^{67}$, and $\mathrm{Zn}^{64}-\mathrm{Zn}^{68}$ as the mass scale it was possible to determine something of the relation between the packing fractions of the zinc isotopes. Eleven good photographs showed the packing fraction of gold was 11.36±0.5 greater than $\bar{f}$ of $\mathrm{Zn}^{64}-\mathrm{Zn}^{66}$, two photographs gave f of gold 11.50±0.15 greater than $\bar{f}$ of $\mathrm{Zn}^{64}-\mathrm{Zn}^{67}$, and seven photographs indicated f of gold was 11.34±0.10 greater than $\bar{f}$ of $\mathrm{Zn}^{64}-\mathrm{Zn}^{68}$. The uniformity of these results is strong evidence that the packing fractions of Zn^{64}, Zn^{66}, Zn^{67}, and Zn^{68} have practically the same values.

Platinum-Gold

In the copper-gold comparison suitable platinum lines appeared on three of the photographs. On these the separation between Pt^{195} and Pt^{196} served as the mass scale and the measurement showed f of gold to be smaller than f of the platinum by 0.23±0.1. This similarity in packing fraction values was to be expected and too much emphasis should not be placed on the small Δf since the results of the copper-platinum comparison assign larger f's to Pt^{195} and Pt^{196} than to the remaining platinum isotopes.

CONCLUSIONS

I. Copper and Zinc Packing Fractions

From Dempster's[3] value for Δf between Au^{197} and $Ti^{49}-Ti^{50}$ of 9.36 ± 0.2, and $\bar{f}$ of $Ti^{49}-Ti^{50}=-7.28\pm0.10$, where Okuda and Ogata's values are averaged in the manner mentioned above, the packing fraction of gold is 2.08 ± 0.30. This value and the packing fraction differences between gold-zinc and zinc-copper enable us to calculate the packing fraction of copper, while the copper-gold comparison gives a second value for copper. A third value for copper can be obtained by employing Graves'[6] values for a gold-molybdenum comparison together with copper-molybdenum values. A fourth value for copper is derived from the less accurate copper-titanium comparison and a fifth is based on the rather unsatisfactory copper-oxygen-copper triplet. The results of these calculations and a mean value for copper based upon them are given in Table I.

The number in brackets following the packing fraction values gives the number of photographs upon which the value is based.

By means of the zinc-gold comparison, the calculated $\bar{f}$ of $Zn^{64}-Zn^{66}=-9.28\pm0.35$. It is surprising to find such a difference between the packing fractions of copper and zinc since they are adjacent in the periodic table and actually overlap each other in their isotopic masses. For this reason it is reassuring to note that the direct copper-zinc comparison yielded a similar result. Using Aston's values for the relative abundance of the zinc isotopes,[12] and assuming that all the zinc isotopes have the same packing fraction of -9.28 ± 0.35, one finds the chemical atomic weight to be 65.36, in reasonable agreement with the value of 65.38 obtained by chemical methods.

II. Discussion of the Packing Fraction Curve

Recent measurements including those reported in this paper, suggest changes in the packing fraction curve.[3] A critical survey of these measurements will follow.

a. *Titanium*—Previous values of the packing fraction of Ti^{48} were -7.22 ± 0.10 (Dempster[3]), -6.97 ± 0.20 (Aston,[13] recalculated by using $C^{12}=12.003871$[14]), and -7.64 ± 0.10 (Okuda and Ogata[11]). The agreement between the last of these and the present value of -7.60 ± 0.07 is a strong argument for accepting a value of -7.62 ± 0.08. Also, the closeness of the values for $\bar{f}$ of $Ti^{47}-Ti^{49}$, namely, -7.34 ± 0.16 (Okuda and Ogata) and -7.37 ± 0.15 (in this paper), is good justification for adopting the Okuda and Ogata values for the remaining titanium isotopes.

b. *Chromium*—Aston reported a value for the packing fraction of Cr^{52} in 1938[12] of -8.15 ± 0.33 (recalculated by using $C^{12}=12.003871$) which is to be compared with the present value for Cr^{54} of -7.33 ± 0.55. Since this difference is not likely real but due largely to experimental error, a mean value of -7.74 ± 0.45 should be a fair approximation for all chromium isotopes.

c. *Manganese*—The present measurement of the packing fraction of manganese is the only one that has been made.

d. *Iron*—Dempster obtained[3] packing fractions of $Fe^{54}=-7.5\pm0.8$ and $Fe^{56}=-7.0\pm0.4$ while in this paper f of $Fe^{54}=-7.37\pm0.55$. Therefore, for plotting on a packing fraction curve, the iron isotopes may be represented by -7.22 ± 0.3.

e. *Nickel*—The only reported value of a packing fraction for nickel has been given by Aston[13] for $Ni^{58}=-8.35\pm0.35$.

f. *Copper*—The present value of $\bar{f}$ for $Cu^{63}-Cu^{65}$ of -7.92 ± 0.25 is to be preferred to Dempster's previous value[3] of -6.9 ± 0.2 which was based on only two photographs.

g. *Zinc*—Aston has obtained a provisional value for the packing fraction of $Zn^{64}=-9.9\pm3.0$,[12] and Dempster has reported[3] for Zn^{64}

TABLE I.

	f of Cu^{63}	f of Cu^{65}	$\bar{f}$ of Cu
Cu-Zn		-8.07 ± 0.50 (4)	-8.27 ± 0.75 (1)
Cu-Mo	-8.10 ± 0.85 (3) -8.27 ± 0.85 (4)		
Cu-Au			-7.74 ± 0.70 (8)
Cu-O			-7.67 ± 0.35 (16)
Cu-Ti	-7.95 ± 0.55 (1)	-8.30 ± 0.50 (1)	
		Weighted Mean	-7.92 ± 0.25

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[12] F. W. Aston, *Mass Spectra and Isotopes* (Longmans, Green, and Co., 1933), p. 120.

[13] F. W. Aston, Nature **141**, 1096 (1938).

[14] K. Ogata, Proc. Phys. Math. Soc. Japan **22**, 486 (1940).

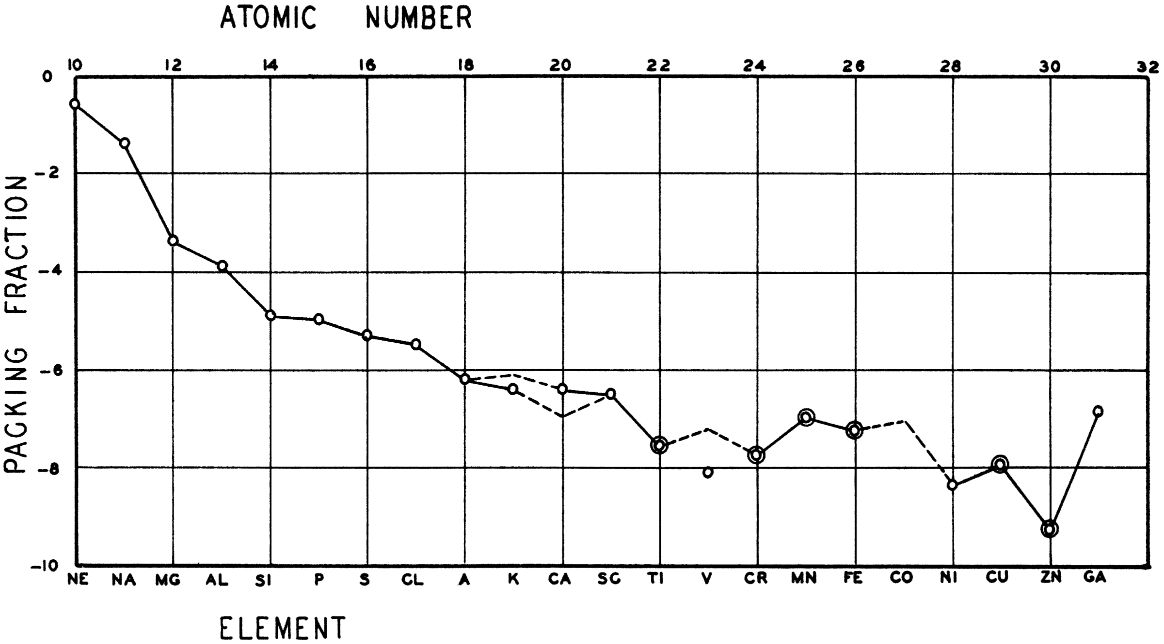

FIG. 2. Curve showing the variation of the packing fraction with increasing atomic number. The values marked by double-circles are reported in this paper, the value for gallium is Dempster's, the value for nickel is Aston's, and the remainder have been obtained from Pollard's list (reference 15).

$f=-6.9\pm0.8$, for Zn^{66} $f=-7.4\pm0.8$, and for $Zn^{68}-Zn^{70}$ $\bar{f}=-6.6\pm0.6$. The present experiments show the packing fractions of all the zinc isotopes (excepting Zn^{70}) to be approximately equal to -9.28 ± 0.35. This value is believed to be fairly reliable.

h. *Gallium*—The only value for gallium has been obtained by Dempster[3] who was able to compare it with palladium and showed that its packing fraction was 1.50 ± 0.20 smaller than that of palladium. This can be used with a palladium-aluminum comparison to calculate the packing fraction of gallium $=-6.85\pm0.9$. Dempster cautions that the palladium-aluminum comparison was not very satisfactory.

When these values, together with values for the lighter elements published by Pollard,[15] were plotted on the usual type of packing fraction curve, it was impossible to draw a reasonable curve which recognized most of the points. To solve this difficulty the packing fraction was plotted against atomic number instead of against mass number. The interesting result is shown in Fig. 2. With two exceptions, plus a possible exception due to the gap at cobalt (atomic number 27), *the packing fraction of an element with odd atomic number is algebraically larger than the average of the two adjacent even-numbered elements.* The two exceptions involve transmutation data which Pollard admits are inaccurate—a slightly smaller packing fraction for calcium and a somewhat larger packing fraction for vanadium would remove these discrepancies. With elements of atomic number less than 10 the adherence to the above rule is very marked and has only been omitted from this graph in order to avoid using too small a scale. Since experimental data above atomic number 32 are incomplete, there has been no attempt to extend this curve to higher elements. However, values for heavier elements published by Aston[13,16] (Br, Kr, Xe), Mattauch[17] (Sr), Dempster[3,4] (Rh, Pd, Ag, etc.), and Graves[6] (Mo, Ru, etc.) seem to follow the same rule although the variations become smaller with increasing atomic number. The fact that the packing fraction of N^{14} is greater than the arithmetic mean of C^{12}

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[15] Ernest Pollard, Phys. Rev. **57**, 1186 (1940).

[16] F. W. Aston, Nature **140**, 149 (1937).

[17] J. Mattauch, Naturwiss. **25**, 170 (1937).

and O^{16} has been pointed out by Weizsacker.[18] He suggested in this connection that the packing fraction of an isotope with odd atomic number will be greater than the mean of the neighboring isotopes with the *same* number of neutrons and one proton more or less.

This relation between odd- and even-numbered elements implies that *those with even atomic number are more stable than the odd-numbered elements.* Consequently, elements with an even atomic number should be more abundant in nature—a fact which has long been recognized. In 1917 Harkins[19] pointed out that elements of even atomic number are predominantly abundant both in the earth's crust and in meteors. This rule has been confirmed by x-ray analysis of terrestrial and stellar matter[20, 21] and is splendidly illustrated by the relative abundances of the rare earths.[22] A recent estimate states that elements with even atomic number constitute 87 percent of the earth's crust.[23]

Furthermore, since the general form of the packing fraction curve is well-known, it is possible to make a further generalization regarding the relative abundances of the elements. On the whole, elements with algebraically small packing fractions, being very stable, should be more abundant than those with larger packing fractions. This state of affairs is also well-recognized—the elements which comprise most of the earth's crust are those with negative packing fractions, while the very light and the very heavy elements, whose packing fractions are positive, are less abundant.

This rule may be interpreted as confirmation of the theory that protons in the nucleus are associated together in pairs. Starting with an even-numbered element, the addition of one proton produces an odd-numbered element which may or may not be more stable than its predecessor. However, the addition of a second proton results in an even-numbered element and, invariably (excepting the two afore-mentioned cases), in an increase in stability. We might suspect from this that the two protons have united with two neutrons to form the stable He^{++} unit which then forms part of the conveniently pictured "core" of the nucleus.[24] At any rate, whether neutrons are associated with them in the union or not, it appears as if the protons in the nucleus are grouped in pairs, each pair existing in or as a very stable union. In other words, the interaction between two protons in the same or adjacent quantum states adds greatly to the stability.[25] There is also considerable evidence for neutron-neutron interaction in the table of masses of stable nuclei published by Pollard.[15] For example, the packing fractions of Si^{28} (14 neutrons), Si^{29} (15 neutrons), and Si^{30} (16 neutrons) are -4.86, -4.54, -5.79, respectively, and those for S^{32} (16 neutrons), S^{33} (17 neutrons), and S^{34} (18 neutrons) are -5.30, -5.28, and -5.96. In these cases, as well as for the trios $O^{16}-O^{17}-O^{18}$, $Ne^{20}-Ne^{21}-Ne^{22}$, and $Mg^{24}-Mg^{25}-Mg^{26}$, the stability shows a marked increase with the addition of the second neutron. Perhaps neutrons obey a rule similar to the one suggested above for protons.

Using the same ordinates and abscissae as in the shown graph, and extending the range of Z so as to include the elements below $Z=10$, we have drawn two curves, one passing through the points representing the packing fractions of the odd-numbered and the other through the points corresponding to the even-numbered elements. If allowances for the inaccuracy of the data are made, the curves were reasonable, one lying above the other, but it was felt that such curves are of dubious value until the points are located with greater accuracy.

The results of the platinum-gold and copper-platinum-iridium comparisons show that platinum, atomic number 78, has a packing fraction larger than the average of iridium (77) and gold (79). This is in a region where the differences are slight and the apparent contradiction of the

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[18] C. F. v. Weizsacker, Zeits. f. Physik **96**, 431 (1935).

[19] W. D. Harkins, J. Am. Chem. Soc. **39**, 856 (1917).

[20] Georg von Hevesy, *Chemical Analysis by X-Rays and Its Application* (McGraw-Hill Book Co., 1932).

[21] V. M. Goldschmidt, *Geochemische Verteilungsgesetze der Elemente*, IX, *Die Mengenverhaltnisse der Elemente und der Atom-Arten* (Oslo, 1939).

[22] V. M. Goldschmidt and L. Thomassen, Videnskaps Skrift. 1, Mat.-Naturv. Kl., No. 5 (1924).

[23] Franco Rasetti, *Elements of Nuclear Physics* (Prentice-Hall, Inc., 1936), p. 160.

[24] W. H. Barkas, Phys. Rev. **55**, 691 (1939).

[25] H. A. Bethe and R. F. Bacher, Rev. Mod. Phys. **8**, 100 ff (1936).

TABLE II. Summary of comparisons.

Mass	Ions	$\Delta f \times 10^4$	Number of Photographs	Assume	Deduce
16	$Ti^{48}-O^{16}$	$-\ 7.60\pm0.07$	5	$O^{16}: f=0$	$Ti^{48}: f=-7.60\pm0.07$
16	$Ti^{47}-O^{16}-Ti^{49}$	$-\ 7.37\pm0.15$	5	$O^{16}: f=0$	$Ti^{47}-Ti^{49}: \bar{f}=-7.37\pm0.15$
16	$Cu^{63}-O^{16}-Cu^{65}$	$-\ 7.67\pm0.35$	16	$O^{16}: f=0$	$Cu^{63}-Cu^{65}: \bar{f}=-7.67\pm0.35$
15.75	$Ti^{47}-Cu^{63}-Ti^{48}$	$0.4\ \pm0.4$	1	$Ti^{47}-Ti^{48}: \bar{f}=-7.55\pm0.15$	$Cu^{63}: f=-7.95\pm0.55$
16.25	$Ti^{48}-Cu^{65}-Ti^{49}$	$0.9\ \pm0.4$	1	$Ti^{48}-Ti^{49}: \bar{f}=-7.40\pm0.10$	$Cu^{65}: f=-8.30\pm0.50$
53.5	$Cr^{53}-Ag^{107}-Cr^{54}$	$-\ 2.35\pm0.10$	7		
54	$Ag^{107}-Cr^{54}-Ag^{109}$	2.38 ± 0.05	7	$Ag^{107}-Ag^{109}: \bar{f}=-4.95\pm0.5$	$Cr^{54}: f=-7.33\pm0.55$
54	$Ag^{107}-Fe^{54}-Ag^{109}$	2.42 ± 0.05	9	$Ag^{107}-Ag^{109}: \bar{f}=-4.95\pm0.5$	$Fe^{54}: f=-7.37\pm0.55$
55	$Ag^{107}-Ag^{109}-Mn^{55}$	2.03 ± 0.10	5	$Ag^{107}-Ag^{109}: \bar{f}=-4.95\pm0.5$	$Mn^{55}: f=-6.98\pm0.60$
65.67	$Zn^{64}-Au^{197}-Zn^{66}$	-11.36 ± 0.05	11	$Au^{197}: f=2.08\pm0.3$	$Zn^{64}-Zn^{66}: \bar{f}=-9.28\pm0.35$
65.67	$Zn^{64}-Au^{197}-Zn^{67}$	-11.50 ± 0.15	2		
65.67	$Zn^{64}-Au^{197}-Zn^{68}$	-11.34 ± 0.10	7		
64	$Cu^{63}-Zn^{64}-Cu^{65}$	1.01 ± 0.4	1	$Zn^{64}: f=-9.28\pm0.35$	$Cu^{63}-Cu^{65}: \bar{f}=-8.27\pm0.75$
65	$Zn^{64}-Cu^{65}-Zn^{66}$	$-\ 1.20\pm0.15$	4	$Zn^{64}-Zn^{66}: \bar{f}=-9.28\pm0.35$	$Cu^{65}: f=-8.07\pm0.50$
31.5	$Cu^{63}-Mo^{95}-Mo^{97}$	$-\ 2.14\pm0.4$	3	$Mo^{95}-Mo^{97}: \bar{f}=-5.96\pm0.45$	$Cu^{63}: f=-8.10\pm0.85$
31.5	$Cu^{63}-Mo^{95}-Mo^{98}$	$-\ 2.31\pm0.4$	4	$Mo^{95}-Mo^{98}: \bar{f}=-5.96\pm0.45$	$Cu^{63}: f=-8.27\pm0.85$
63.7	$Cu^{63}-Ir^{191}-Cu^{65}$	-10.55 ± 0.05	14		
64.3	$Cu^{63}-Ir^{193}-Cu^{65}$	-10.58 ± 0.05	14		
64.0	$Cu^{63}-Pt^{192}-Cu^{65}$	-10.53 ± 0.15	6		
64.7	$Cu^{63}-Pt^{194}-Cu^{65}$	-10.49 ± 0.15	12		
65	$Cu^{65}-Pt^{195}$	-10.74 ± 0.20	12		
65.3	$Cu^{63}-Cu^{65}-Pt^{196}$	-10.80 ± 0.15	8		
63	$Cu^{63}-Ir^{191}-Ir^{193}$	-10.49 ± 0.1	8		
65	$Ir^{191}-Ir^{193}-Cu^{65}$	10.75 ± 0.1	8		
65	$Pt^{194}-Cu^{65}-Pt^{196}$	10.62 ± 0.1	8		
65.67	$Cu^{63}-Cu^{65}-Au^{197}$	$-\ 9.82\pm0.4$	8	$Au^{197}: f=2.08\pm0.3$	$Cu^{63}-Cu^{65}: \bar{f}=-7.74\pm0.7$
65.67	$Pt^{195}-Pt^{196}-Au^{197}$	0.23 ± 0.10	3	$Au^{197}: f=2.08\pm0.3$	$Pt^{195}-Pt^{196}: \bar{f}=2.31\pm0.40$

rule advanced above may be due to errors. It is believed that the copper-platinum-iridium results are reliable in the main, but the copper-gold plates were not as satisfactory. If the rule is to hold, we must assume that Δf of the copper-gold comparison is too small and that copper possesses an algebraically lower packing fraction than the one which has been assigned (-7.92), possibly about -8.5. On the other hand, an error in Dempster's titanium-gold comparison may have resulted in a packing fraction for gold which is too small. Dempster's plates for this comparison were very good but it should be pointed out that a larger f for Au^{197} would improve the agreement between mass-spectrographic and chemical values for the atomic weight of both gold and zinc (as calculated earlier). The adoption of f of gold $=2.4$, which is the gold packing fraction necessary to bring the zinc values into agreement, would still leave a huge gap of 0.1 mass unit between the gold values.

SUMMARY

In Table II, a summary is given of the comparisons described in this paper. As in the rest of the paper, the factor 10^{-4} must be written with the listed packing fraction values and differences in order to obtain the true packing fractions and differences. The errors are expressed in the form of probable errors.

ACKNOWLEDGMENTS

The author is deeply sensible of the privilege of carrying out this research under the direction of Professor A. J. Dempster. He is grateful not only for permission to use Professor Dempster's mass spectrograph, and for his advice and encouragement so readily given, but also for the opportunity of associating with a man who is at once so excellent a scientist and so considerate a mentor. Appreciation is expressed to Dr. Hans Beutler and Dr. A. E. Shaw for several helpful suggestions, and to the other members of the University of Chicago Physics Department for their interest and help.