# THE SYNTHESIS OF ATOMS, THE WHOLE NUMBER RULE, AND THE PERIODIC SYSTEM OF THE ATOMIC SPECIES<sup>1</sup>

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#### I. THE DISINTEGRATIVE SYNTHESIS OF ATOMS

#### 1. Introduction

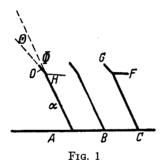
In the decade between 1860 and 1870 a study of the relations between the chemical and physical properties of the elements and their atomic weights led to the formulation of what is commonly designated as the periodic system of the elements. The development of this system was due to de Chancourtois, Newlands, Mendeléeff, Meyer, and a number of others. Recent investigation concerning what is commonly called the structure of the atom seem to indicate that the part of the atom to which this system is particularly related is the non-nuclear portion. It must be admitted that while in recent years much has been learned of the properties of materials and of the behavior of very small amounts of substances, particularly dilute gases, almost nothing is known of the structure of any atom. The idea concerning this structure which seems best supported by the experimental evidence is that the portion of the atom which contains nearly all of its mass (the nucleus) is extremely small in comparison with the entire atom, and is charged with positive electricity. Nothing is known as to the structure of either this nucleus or the more diffuse system (presumably charged with negative electricity) which seems to surround it, except that there seems to be a cer-

<sup>&</sup>lt;sup>1</sup> Since the preceding paper by Dr. S. C. Lind gives a summary of the work on the nucleus with the exception of that carried out in Kent Chemical Laboratory, the present paper is restricted to the work of this laboratory and that upon which it is based.

Harkins and Shadduck (6), but the success of the work in both cases was due to the use of the method of Harkins and Ryan without any essential or important change.

The work reported here is a continuation by Harkins and Shadduck of the work of Harkins and Ryan by the use of the same, but modified, apparatus, which consisted in a modified Shimizu-Wilson cloud track apparatus. The number of photographs taken by us is 34,000 with an average of slightly more than  $12 \alpha$ -tracks each, so that 270,000 tracks of 8.6 cm., and 145,000 tracks of 4.9 cm. range were obtained.

Figure 1A shows the arrangement in a plane of the tracks in one of the cases in which a fast  $\alpha$ -particle attaches itself to the



A, a disintegrative-synthesis of oxygen (O) from nitrogen and helium. B, the same disintegrative-synthesis with the track of the proton (H) omitted. C, an ordinary collision between the alpha-particle and the nucleus of an atom of nitrogen.

nucleus of a nitrogen atom. This, as has been stated, forms momentarily the nucleus of an atom of fluorine which disintegrates almost at once to give a fast H-particle or proton, and the nucleus of a heavier atom, presumably that of oxygen of mass 17. All three tracks lie in a plane. The angle between the direction of the track of the  $\alpha$ -particle and that of the H-particle is 118°. The photograph indicates that the track of the proton has only about one-sixth to one-tenth the intensity of that of the  $\alpha$ -particle or of the oxygen atom. Figure 1B illustrates what would be formed provided the track of the H-particle should be so faint as to be invisible. Figure 1C illustrates the marked differ-

ence in the appearance when the  $\alpha$ -particle rebounds in the case of an ordinary (but extremely rare) collision. Here the thin track is not present and the angles are noticeably different. Figure 2 is a reproduction of a photograph of the disintegrative-synthesis or atomic metathesis represented by figure 1A.

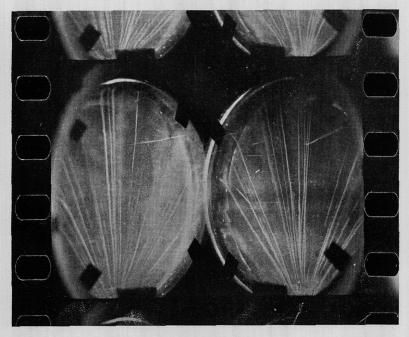


Fig. 2. Two Views at Right Angles of the Non-elastic Collision Represented in Figure 1, A

At the left of each view an  $\alpha$ -particle impinges upon the nucleus of a nitrogen atom and unites with it. An extremely faint straight track extends horizontally to the right of the fork and is due to a porton or H-particle emitted at high speed. This has a range of 19.6 cm. in air at 15° and 760 mm. pressure. The track of the oxygen atom which is formed slants upward to the left from the fork. The left-hand view is overexposed as a result of an irregularity in the lighting, but the track of the H-particle is easily visible on the original negative.

Table 1 gives the characteristics of this fork.

The only evidence that the inelastic collision represented by fork 2 is a disintegrative synthesis, is the appearance of the fork. The thin track has exactly the appearance given by hydrogen.

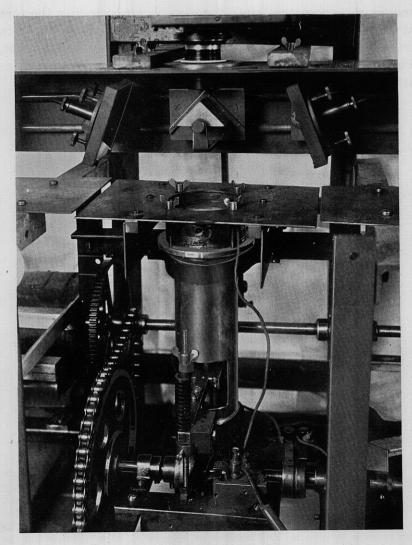


Fig. 3. Modified Wilson (Shimizu) Apparatus for the Photography of Alpha-Ray Tracks

However, measurements on the fork cannot be exact on account of curvature near the point of impact.

Figure 3 represents the ionization chamber used in the work,

and figure 4 the natural disintegration of thoron to give thorium A, and the subsequent disintegration of thorium A to give thorium B (Lead). As in all other cases, the two views are taken at right angles to each other.

### 4. The mesh plate method for the measurement of alpha- and beta-ray tracks

The measurements of the tracks were made by a new method which may be called the "Mesh Plate Method." It consists in

TABLE 1
Characteristics of forks, which represent the disintegrative-synthesis of atoms

	φ	0	$R_{\alpha}$	$V_{\alpha} \times 10^{-3}$	$V_{\rho} \times 10^{-9}$	$R_{ ho}$
Fork 1				1.86 1.57	2.7	19.6
				$V_{\rm n} \times 10^{-9}$	Rn	$\mathbf{E}_2/\mathbf{E}_1$
Fork 1				0.534	0.43	0.89

- R = remaining range of α-particle; the distance the α-particle would have traveled beyond the point of the fork if the collision were not incurred.
- $V = \text{velocity of the } \alpha\text{-particle in centimeters per second immediately before the collision.}$
- $V_{\rho}$  = velocity of the proton immediately after its escape from the heavy
- $R_o = \text{range of the proton, or H-particle.}$
- $V_n$  = velocity of the oxygen nucleus immediately after the escape of the
- $R_n$  = range of the nucleus of the oxygen atom.
- $E_2/E_1$  = kinetic energy of the H-particle plus that of the oxygen atom, divided by the kinetic energy of the  $\alpha$ -particle immediately before impact.  $1 E_2/E_1$  represents the loss of kinetic energy due to the non-elastic impact, and is presumably stored up in the oxygen nucleus of mass 17.

the establishment in the ionization chamber, which consists of a cylinder of optical glass, of a system of coördinates in space. A circular plate of yellow brass was made of such a size as to fit the cylinder with a clearance of about 0.5 mm. This was ruled on the milling machine into squares of 1 sq. mm. area by lines 0.1 mm. in diameter. These lines were then filled with tin solder and the brass surface blackened by the use of ammoniacal am-



Fig. 4. Two Views of the Natural Disintegration of Thoron to give Thorium A, and the Subsequent Disintegration of Thorium A to Give thorium B

(Views at right angles)

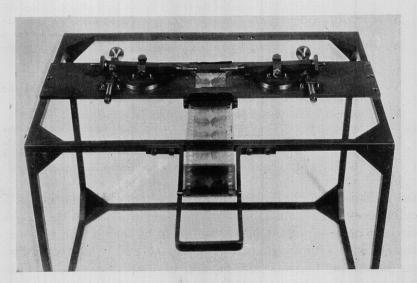


Fig. 5. Apparatus for the Measurement of Atom Ray-Tracks

monium carbonate. This gave very uniform, silver-white lines against black on an entirely plane surface, since the surface was planed prior to the blackening.

The mesh plate is then set on the brass piston, which closes the bottom of the ionization chamber and fits inside a cylinder of brass below the glass. The mesh plate is set in an exact position by the use of a stand with three legs fitted with screws. The bottom of each leg fits a fine screw hole in the top of the piston. By the use of the fixed mirrors, used in obtaining two photographs at right angles of each set of  $\alpha$ -tracks, two photographs at right angles, are taken of the mesh plate. The piston is then raised a distance of 0.02 cm. and another pair of photographs taken. This operation is repeated until the top of the chamber is reached.

In order to find the position in space represented by any point in the photographs, it is only necessary to find the point in both photographs. The film which shows the point is then placed in a special apparatus (fig. 5) which fits the film with considerable This is provided with two movable and very sharp One of these sharp pointers is put on each projection of the point on the film, and fastened in place, except that a vertical movement is possible. The film is then taken out, and the film with the mesh plate images is inserted. This is moved until the reading given for the position of each pointer is the same on one projection as on the other. The reading gives the x and y coördinates, and the number of the film, the z-coördinate, of This method gives the position of a point in space to within 0.1 mm. The use of this method of measurement eliminates errors due to the lens, which was, however, a Taylor-Cooke-Hobson lens of the best grade. Also errors due to the glass at the top of the cylinder, and to the glass of the mirrors are eliminated. Nevertheless all of these were made optically plane in Ryerson Physical Laboratory. If the lines are too short or too highly curved, other methods are used. The mesh-plate method has the great advantage that it allows not only for all errors due to the use of the lens but also for differences in magnification due to the use of two views at right angles.

#### 5. Minimum velocity of the $\alpha$ -particle in a disintegrative-synthesis

The minimum velocity for the  $\alpha$ -particle which has thus far been shown definitely to give rise to an atomic disintegrativesynthesis corresponds to a remaining range of 6.2 cm. This corresponds to the energy given by  $7 \times 10^6$  electron-volts or to the velocity given to the  $\alpha$ -particle by 3.5  $\times$  106 volts. However, Fork 2 seems to represent a synthesis in which a much lower velocity  $(1.61 \times 10^9 \text{ cm. per second})$ , which corresponds to  $5.4 \times 10^6$  electron-volts, was effective. This is only slightly higher than the initial velocity of the  $\alpha$ -particles from polonium  $(1.587 \times 10^9 \text{ cm. per second, or } 5.224 \times 10^6 \text{ electron-volts})$ , so it is probable that it is possible to obtain such a disintegrativesynthesis even when polonium is used as the source of  $\alpha$ -rays, but polonium is altogether unsuitable in the photographic method as applied in this laboratory, that is if a photograph of all of the rays of the fork are obtained. The method of Holubek (7) is of an entirely different order, since it gives no indication that a synthesis has occurred, and is more nearly analogous to the ordinary scintillation method. Up to the present, the photographic method has not demonstrated the occurrence of disintegrations which give short H-rays.

#### II. THE WHOLE NUMBER RULE OF HARKINS AND WILSON (1)

In 1815 Prout, on the basis of extremely inexact data, announced the relation that if the atomic weight of hydrogen is taken as unity, the atomic weights of the other elements are exactly whole numbers.

Table 2 gives the atomic weights according to Prout, and also as given in the latest tables. It is apparent that deviations from the true values as high as 5.9 units for carbon, 15.8 units for sulphur, 19.7 units for calcium, 27.4 units for iron, and 66.3 units for barium, together with the general smaller deviations, indicate that there was no real basis at the time for the whole number rule with hydrogen taken as unity.

While Prout's Whole Number Rule is thus shown to be altogether without justification, the idea which he based upon these

faulty data, that hydrogen is the fundamental element, or protyle, is in accord with the views of the present time. This is a good illustration of the fact that a good theory may be based upon altogether unsound data.

The modern whole number rule is more properly called the law of the approximate constancy of the packing effect.

According to this rule the atomic weight of any atomic species (other than hydrogen) is very close to a whole number on the basis of 16 as the atomic weight of oxygen.

TABLE 2

Prout's table of the more accurately determined (1815) atomic weights

Atomic weight (H = 1)

ELEMENT	PROUT (1815)	VALUE 1928	DIFFERENCE
н	1	1	0 (Standard)
C	6	11.907	5.907
N	14	13.900	-0.1
P	1 <b>4</b>	30.787	16.787
0	16	15.876	-0.124
S	16	31.819	15.819
Ca	20	39.70	19.70
Na	24	22.819	-1.181
Fe	28	55.41	27.41
Zn	32	64.87	32.87
Cl	36	35.183	-0.817
K	40	38.793	-1.207
Ba	70	136.31	66.31
I	124	125.950	1.950

Since the atomic weight of hydrogen is, as nearly as is known, equal to 1.0078, this indicates that the packing effect in a synthesis of any other element from hydrogen is very nearly equal to a loss of mass of 0.77 per cent. When this law was first published it was considered that the packing effect would be constant to within about 0.1 per cent.<sup>2</sup>

<sup>&</sup>lt;sup>2</sup> It was shown later (fig. 22) that the atoms of odd atomic number are less stable than those of even atomic number, so it is not surprising that for the lightest atoms of odd atomic number the packing is 0.57 and 0.60 per cent (lithium

A few quotations from the first paper concerning the whole number rule, are given below.

Not only is the variation of the atomic weight from a whole number a negative number, but in addition its numerical value is nearly constant, the average value for the 21 elements being 0.77 per cent, while the six elements from boron to sodium show values of 0.77, 0.77, 0.70, 0.77, 0.77, and 0.77 per cent. The deviation is therefore not a periodic, but a constant one.

It has formerly seemed difficult to explain why the atomic weights referred to that of oxygen as 16 are so much closer to whole numbers than those referred to that of hydrogen as one, but, the explanation is a very simple one when the facts of the case are considered. The closeness of the atomic weights on the oxygen basis to whole numbers, is indeed extremely remarkable. If an oxygen atom is a structure built up from 16 hydrogen atoms, then the weight according to the law of summation should be 16 times 1.0078 or 16.125. The difference between 16.125 and 16.00 is the value of the packing effect, and if this effect were the same for all of the elements, except hydrogen, then the choice of a whole number as the atomic weight of any one of them, would, of necessity, cause all of the other atomic weights to be whole numbers. Though this is not quite true, it is seen that the packing effect for oxygen is 0.77 per cent, which is the average of the packing effects for the other 21 elements considered. Therefore, those elements which have packing effects equal to that of oxygen will have whole numbers for their atomic weights, and since the other elements show nearly the same percentage effect, their atomic weights must also lie close to whole numbers.

It has usually been assumed, and without any really logical basis for the assumption, that if a complex atom is made up by the union of simple atoms, the mass of the complex atom must be exactly equal to the masses of the simple atoms entering into its structure. Rutherford, from data on the scattering of  $\alpha$ -rays in passing through gold leaf, has calculated an *upper* limit for the radius of the nucleus of a gold atom as

of atomic weights 6 and 7) as found by Costa. In these two special cases the variation from the whole number relation is abnormally large: 0.2, and 0.17 per cent.

Also it would not be surprising if the packing is found to be somewhat smaller for uranium and thorium and adjacent elements, than for elements of half their atomic weight.

 $3.4 \times 10^{-12}$  cm. The mass of this relatively heavy atom is, according to this calculation, practically all concentrated in this extremely small space, which is so small that it could no longer be expected that the mass of such a nucleus, if complex, would be equal to the sum of the masses of its component parts. In fact, since the electromagnetic fields of the electrons would be so extremely closely intermingled in the nucleus, it would seem more reasonable to suppose that the mass of the whole would not be equal to the sum of the masses of its parts. The deviation from the law of summation cannot be calculated on a theoretical basis, but it can easily be determined from the atomic weights, if the assumption is made that the heavier atoms are condensation products of the lightest of the ordinary elements, that is of hydrogen. This deviation expressed in terms of the percentage change, is what has already been determined, and designated as the packing effect.

The closeness to which a positive and a negative electron would have to approach to give a decrease of mass equal to 0.77 per cent, or the average value of the packing effect, is found by calculation to be to a distance of 400 times the radius of the positive electron. This case does not correspond to any element actually known, for the simplest of the atoms considered, helium, may be supposed to have a nucleus built up from four hydrogen nuclei and two negative electrons. However, the magnitude of the effect seems to be of the order which would be expected.

- 1. The atomic weights of the first 27 elements, beginning with helium, are not multiples of the atomic weight of hydrogen by a whole number, as they would be if Prout's original hypothesis in its numerical form were true. This may be expressed by the statement that the atomic weights on the hydrogen basis are not whole numbers. However, when these atomic weights are examined critically it is found that they differ from the corresponding whole numbers by a nearly constant percentage difference, and that the deviation is negative in sign, with an average value of -0.77 per cent.
- 2. This percentage difference has been called the packing effect, and it represents the decrease of weight, and presumably the decrease of mass, which must take place if the other atoms are complexes built up from hydrogen atoms. The regularity in this effect is very striking, the values for a number of the lighter atoms being as follows: He, -0.77; B, -0.77; C, -0.77; N, -0.70; O, -0.77; F, -0.77; and Na, -0.77 per cent, while the average value for the first 27 elements is -0.77 per cent.

As has been seen, there are 27 atomic weights distributed over 59 units of atomic weight. The greatest common divisor of the whole numbers corresponding to the atomic weights is one. The atomic weights are therefore such that numerically they seem to be built up from a unit of a mass of one, and the probability results seem to show that this unit of mass must be very close to 1.000, expressed to three decimal places.<sup>3</sup>

Now that certain elements have been found to exist in isotopic forms, it becomes apparent that still other elements may do the same in cases which have not been recognized, so that in dealing with any single species of element it is uncertain whether this is an individual with respect to its atomic weight. The great regularity with which the elements follow the relationships given in these papers, up to an atomic weight of 59, suggests that with the exception of the cases of neon, silicon, magnesium, and chlorine, isotopes probably do not exist to any large extent for any of these elements, if they exist at all.

### III. THEORETICAL: THE BUILDING OF OTHER ATOMS FROM HYDROGEN

#### 1. Introduction

The fact that the atomic weights of the atomic species are whole numbers (to within 0.1 per cent in general) suggests that all of the more complex atoms are built up from hydrogen, but that in the formation of the complex structure the atomic weight of the hydrogen is changed from 1.0078 to 1.000 ( $\pm$ 0.001). Since, however, hydrogen is supposed to consist of a positive electron (proton) and a negative electron, this is equivalent to the statement that complex atoms are built up from protons and electrons.

In such complex atoms the masses of the protons are not independent, but the average mass of the proton-electron pair in any atom may be considered as  $1.000 \pm 0.001$  ( $1.649 \times 10^{-24}$ g.), if the atomic weight of oxygen is taken as 16.

Since only about 1/1840 of the mass of the hydrogen atom is attributed to the electron, the proton is supposed to contain nearly all the mass; that is, the mass is found to be associated almost wholly with the positively charged particles.

<sup>&</sup>lt;sup>3</sup> Not italicized in the original.

As nearly as is known the absolute magnitude of the positive charge of the proton is equal to that of the negative electron. Any complete atom is apparently electrically neutral, and an atom which is not electrically neutral is designated as an ion.

#### 2. General stability relations of atom nuclei

The existence of atoms and of atomic species is highly dependent upon the numerical values of certain experimental

TABLE 3

Experimental and theoretical significance of fundamental variables related to atomic stability

	The state of the s	THEORETICAL SIGNIFICANCE
VARIABLE	EXPERIMENTAL SIGNIFICANCE	THEORETICAL SIGNIFICANCE
P	Atomic weight of atomic spe- cies or protonic number	Number of protons in the nucleus (respectively in the atom)
${f z}$	Atomic number	Net positive charge on the nucleus
N	P - Z Electronic number	Number of negative electrons in the nucleus
n	P - 2Z 2N - P N - M Isotopic number	·
$\frac{N}{P}$	$\frac{P-Z}{P}$ Ratio of electronic to protonic number	Ratio of negative to positive electrons in the nucleus

quantities as listed in table 3. Since existence depends upon stability, it seems apparent that the stability of atoms is dependent upon the magnitudes of the same quantities.

It is important to emphasize that N is an entirely experimental number, since it is the difference between the atomic weight and the atomic number, both of which are determined by experiment. The same is true of the isotopic number (n). It may be noted

that from the experimental standpoint all of the four variables P, Z, N, and n, are very close to whole numbers, while from the theoretical standpoint they are exactly whole numbers.

The protonic number may now be defined as the whole number which is nearest to the mass number (or atomic weight) of the atomic species. The electronic number is the whole number which is nearest to the difference between the protonic number and the atomic number (N = P - Z).

#### 3. The nucleus as a proton-electron aggregate

If the nucleus is considered as built up from protons and electrons, without any reference to secondary groups or structure, some interesting relations emerge.

All of the protons of the atom, and usually half the electrons, go into the nucleus, while the other half of the electrons make up a more diffuse system, which is responsible for the emission and absorption spectra in the visible, ultra-violet, and x-ray regions of the spectrum. In the case of only about 18 per cent of known atoms, more than half of the eletrons enter the nucleus, so for these atoms the ratio N/P is greater than 1/2. The numerical relations involved are discussed in Section IV.

#### IV. GENERAL NON-PERIODIC RELATIONS CONCERNING ATOM NUCLEI

1. Stability and the ratio of negative to positive nuclear electrons (3)

One of the most fundamental of all of the relations or laws which concern the stability of atom nuclei is as follows:4

No species of atoms (other than hydrogen) which has been found to exist either on earth or in the meteorites has a value for the ratio of negative to positive electrons  $\left(\frac{N}{P}\right)$  of less than 1/2 or 0.5.

It is remarkable, too, that in most of the atoms which exist the

<sup>&</sup>lt;sup>4</sup> This relation was used by the writer as a basis for the development of the "Hydrogen-Helium Theory," J. Am. Chem. Soc., 37, 1383 (1915), 39, 856 (1917), and is discussed in considerable detail in ibid., 42, 1964-75 (1920), Phil. Mag., 42, 308 (1921).

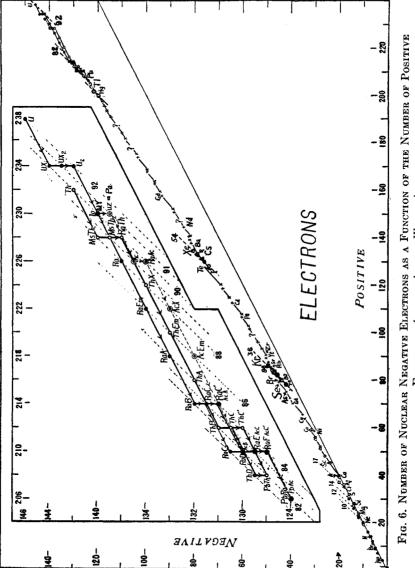
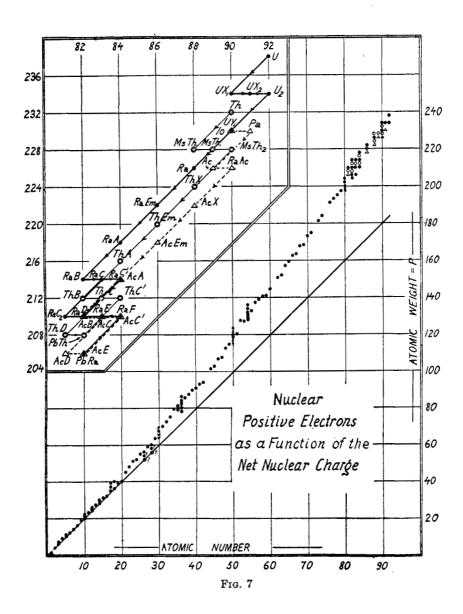


Fig. 6. Number of Nuclear Negative Electrons as a Function of the Number of Positive Electrons (= the Atomic Weight)



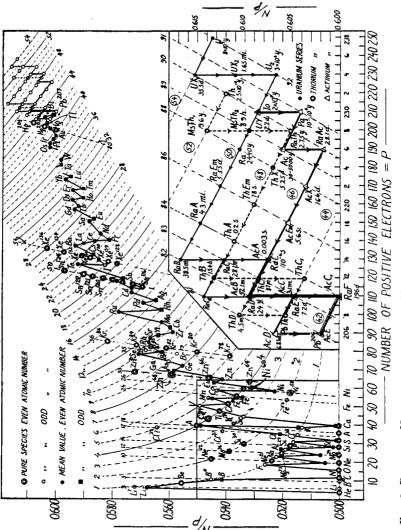
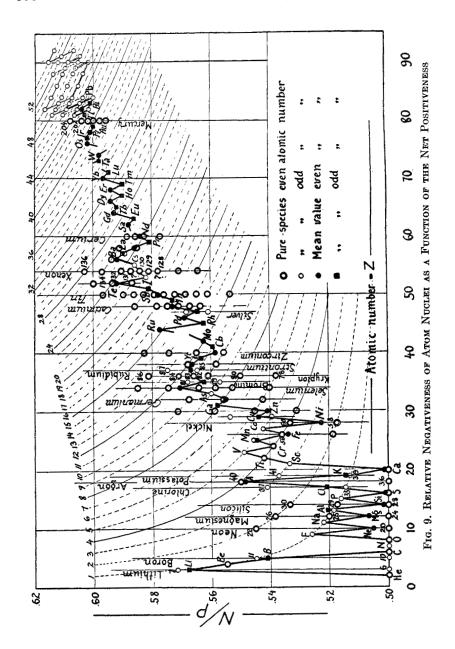


Fig. 8. Relative Negativeness of Atom Nuclei as a Function of Their Total Positiveness



ratio  $\frac{N}{P}$  is exactly 0.5, since such atoms constitute 80 per cent of the meteorites and 85 per cent of the earth's crust.

In figure 6 the electronic number is given on the Y-axis, and the protonic number on the X-axis. It is apparent that most of the light atom species up to atomic number 20 (Ca) lie either exactly on, or else only slightly above, the straight line of slope 1/2. As the atomic number increases beyond 20 the curve, which represents the mean value for each element, rises farther from this line, not very far above it. The plot of the radioactive series indicates that all radioactive changes are represented by a shift along either a vertical line, or along a line whose slope is 1/2.

The vertical extent of the band (fig. 6) in which atoms are stable, is very small. Thus for any definite value of the number (P) of protons in the nucleus, the range in the number of electrons (N) is very small for any atomic species (which is sufficiently abundant to have been discovered). Figure 7 gives the protonic number as a function of the atomic number.

The relation is shown in a more useful form in figure 8 which gives the ratio N/P on the Y-axis and, P, the number of protons on the X-axis. However, the plot shown in figure 9 is even more suggestive. It gives the atomic number or net nuclear positive charge (Z = P - N) on the X-axis, and the ratio N/P on the Y-axis. Thus the net positiveness of the nucleus is plotted against its relative negativeness.

#### 2. General theory of nuclear stability

The most abundant light atom nuclei have atomic numbers and weights which may be represented by the formula  $a_J$ , in which a designates an  $\alpha$ -particle, and J is any whole number (except 2) from 1 to 10 (J = Z/2). This shows either (1) that such nuclei are built up by the aggregation of  $\alpha$ -particles, or (2) that, though not built up entirely by the aggregation of  $\alpha$ -particles, the stability of nuclei whose composition is represented by that of a whole number of  $\alpha$ -particles, is very much higher than that of other nuclei.

That a fast  $\alpha$ -particle in some cases attaches itself to the nucleus of an atom has already been shown (4, 5, 6).

The heaviest known atomic species whose nucleus may be represented as  $\alpha_J$  is calcium of atomic weight 40, for which J has the value 10, and for which N/P has the value 1/2.

The preliminary assumption of the theory is that in atomic nuclei positively charged particles repel each other. However the binding action of negative electrons is so great that 3, 4, 5, 6, 7, 8, 9, or  $10 \alpha$ -particles will unite with each other in an atomnucleus, even although each  $\alpha$ -particle has a net positive charge of two. If somewhat more than 10 alpha particles are to unite, two additional (cementing) electrons must also enter the nucleus (1, 2, 7). Thus the charge of one  $\alpha$ -particle does not appear in the net charge on the nucleus. If the number of  $\alpha$ -particles rises above 16 (approximately) an additional pair of cementing electrons must be utilized, while in the heaviest atoms 13 such pairs of cementing electrons are present.

The theory may be stated in a much more general form as follows: The net positive charge on the nucleus of an atom may rise as high as 20 while the relative negativeness (ratio of negative to positive electrons) remains at its most general value of 0.5, but in order that the nucleus may be stable and attain a higher net positive charge, the relative negativeness must increase to an extent which increases with increase in the net positive charge. Thus the atom nucleus, in order to be stable, must have a relative negativeness (N/P) which matches its net positiveness (Z).

The band of stability is shown in figure 8. It is of great interest that the small number of atomic species contained in the rectangle in the lower left-hand corner of this figure constitute 99.9 per cent of all known material.

The usefulness of the relationship between stability and the relative negativeness (N/P) of the nucleus is illustrated by the fact that before the discovery of the isotopes of lithium four predictions were made concerning their existence. Three of these, made by others without a consideration of this relationship, were that the mass numbers are (1) 6 and 8, (2) 5 and 7, and (3) 6, 7, 8. Prediction (3), made by Rutherford, was the best

of the three, since its single error consisted in the assumption of the existence of a lithium isotope of isotopic number 2, with a value of N/P equal to 0.625. This is 0.125 in excess of the minimum value 0.5, while the highest value for any atom, even in the radioactive region, is only 0.117 in excess. Moreover, in the region of the plot in which lithium is found, the excess (0.072) for lithium, of mass number 7, is the maximum found for any atomic species. The prediction of an isotope of mass 5 is in discord with the fundamental relation, since the ratio N/P for such a species would be 0.4, or less than the minimum value.

The writer's prediction of lithium isotopes of masses 6 and 7 was made, on the basis of the relation between N/P and Z, in a paper presented to the American Chemical Society on April 12, 1920. The prediction was verified a year later by G. P. Thomson. A similar prediction, that boron would be found to have mass numbers 10 and 11, was also verified.

A study of the composition of the meteorites and of the crust of the earth, indicates that no atomic species of positive nuclear charge higher than 29, and also no species in which the ratio N/P is greater than 0.54 occurs in an amount greater than 1 atom in  $10^4$  (0.01 per cent).

While the ratio N/P is not the only factor upon which the stability of an atom depends, it is evidently of primary importance. Thus, beginning with species of low atomic number, it is found that lithium, beryllium, and boron, have ratios for the principal isotope of 0.572, 0.556, and 0.545. Now, all of these are abnormally high for such low atomic numbers (3, 4, and 5), and the species are found to be rare both on earth and in the meteorites. Carbon, the first species in the system (excluding the fundamental hydrogen and helium) in which the ratio falls to the normal value 0.5 is far more abundant, and is the lightest complex atom found in any considerable quantity in the meteorites. It is also about 26 times more abundant in the earth's crust than any one of the three other atomic species just mentioned. Nitrogen (Z = 5), though it has a value of N/P of 0.5, is not at all abundant. This may be explained as due to the fact

that it does not meet another extremely important stability condition discussed later.

The next three atomic species for which N/P is 0.5, are the principal isotopes of oxygen, magnesium, and silicon, with percentages of 61.94, 1.40, and 19.06, in the earth's crust, and 53.16, 9.86, and 13.82, in the meteorites. It is striking that these three atomic species constitute about 80 per cent of the atoms in known material. Neon is omitted from consideration since it is so light a gas that it escapes from the meteorites.

Until argon (Z=18) is reached the most abundant isotope of each element of even atomic number, exhibits a value of 0.5 for N/P. With argon the ratio for the most abundant isotope rises to 0.55. This falls to 0.5 again for calcium (Z=20), and rises again to 0.542 for titanium (Z=22), 0.539 for chromium (Z=24), and 0.536 for iron (Z=26, n = 4). In general with increasing atomic number, the ratio N/P rises suddenly, then falls very gradually, and then repeats the procedure. The abundance of iron is remarkable if its relatively high atomic number (26) is taken into account, but this may be considered as due in part to a proper value of N/P (0.536) to give stability.

### 3. The relative negativeness (N/P) of the nucleus and the stability of radioactive atoms

The greater the ratio N/P, that is the relative negativeness of the nucleus, the more energetic is the emission of a  $\beta$ -particle which is negative, and the less energetic is the emission of an  $\alpha$ -particle which is positive, provided the net positive charge on all of the nuclei compared is kept constant, that is when only isotopes are considered.

The above statement applies only to changes which actually occur. It puts into a new form an empirical relation discovered by Fajans.

The existence of this relation indicates that the term "stability of the nucleus" is not definite in its meaning since an atom nucleus which is extremely stable in the sense of an  $\alpha$ -disintegration may be extremely unstable with respect to a  $\beta$ -disintegration.

### V. GENERAL REPRESENTATION OF THE PERIODIC SYSTEM OF THE ATOMIC SPECIES

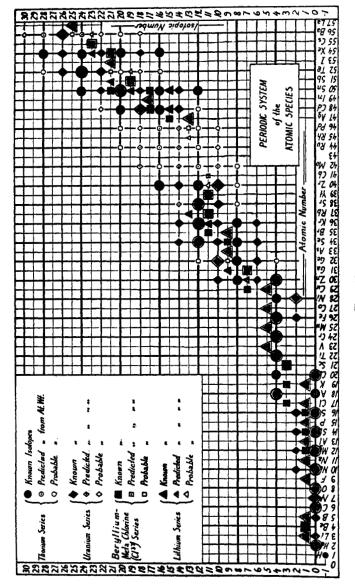
The periodic relations which concern atom nuclei are even more striking than those for the outer atom as exhibited in the periodic system of the elements as outlined by de Chancourtois, Newlands, Mendeléeff, and Mayer. The most general relation may be stated as follows: Atomic stability, the abundance of atoms, and the number of atomic species of any type are all much higher for even than for odd numbers of (1) the electronic number (N) (number of negative electrons in the nucleus), (2) the protonic number (P) (atomic weight or number of protons in the nucleus), (3) the atomic number (Z) (number of net positive charges on the nucleus of the atom, and (4) the isotopic number (n). In addition there is a superimposed periodicity of 4 in the protonic number, and in the isotopic number.

The new periodic system requires for its representation a figure in at least three dimensions. On account of the complexity of a 3 (or more) dimensional representation on the plane surface of a page, the relations of the system are exhibited in this paper by a series of projections.

The simplest two dimensional representation may be described as a double network of squares (fig. 10) with the atomic number on the X-axis, and the isotopic number on the Y-axis. The isotopic number for any atomic species other than hydrogen gives the number of the isotope according to a system in which the lowest possible isotope of any element is given the number zero. For example the isotopic numbers for the known species of a few elements are: He, 0; Li, 0 and 1; Be, 1; B, 0 and 1; C, 0; Mg, 0, 1, and 2; Al, 1; Si, 0, 1, and 2; Fe, 2 and 4. If the empirical formula of any nucleus is written (p<sub>2</sub>e)<sub>Z</sub> (pe)<sub>n</sub>, then the subscript n gives the isotopic number, just as the subscript Z gives the atomic number.

The isotopic number is as important to distinguish the species as the atomic number to distinguish the element.

In figure 10 the heavy coördinates represent even numbers and the light lines odd numbers. Now it is remarkable that where heavy



èra. 10

lines meet heavy lines the greatest number of species occur (67 known species in the figure), and these are also by far the most abundant. Where light lines intersect light lines the number of known species is less (32), and the atoms of these species are in general far less abundant. Where heavy vertical lines intersect light horizontal lines the number of known species is again reduced by about 1/2 to 14 species. By far the fewest species occur where light vertical meet heavy horizontal lines. It is indeed remarkable that only 3 species (Li<sub>0</sub>, B<sub>0</sub>, and N<sub>0</sub>) of this type are known in the whole system of 92 elements, and it is even more remarkable that all of the three lie on the lowest heavy line of the diagram, that of isotopic number 0.

The further details of these periodicities are discussed in the sections which follow.

VI. FIRST PERIODIC RELATION: HIGH STABILITY AND ABUNDANCE FOR ATOMS OF EVEN ATOMIC NUMBER (EVEN NUCLEAR CHARGE)

#### 1. Introduction

The first periodic relation was discovered in the winter of 1914–1915 in an attempt to obtain evidence that the nuclei of complex atoms, that is all atoms other than hydrogen, are built up largely from  $\alpha$ -particles, an idea suggested by the fact that the atomic weights of the light elements, with the single exception of glucinium, are divisible by 4 provided their atomic numbers are divisible by 2, that is the atomic weight and atomic number of helium. The development of the idea is shown best by a quotation from the second paper on the stability and abundance of the elements (2). It seems evident that the importance of the relation does not depend at all on the correctness of the point of view which led to its discovery.

It is the purpose of this paper to present a periodic system of the elements which relates their abundance to the structure of the nuclei of their atoms. This system is called *new* to distinguish it from the ordinary periodic system of Mendeleeff, to which it bears no relation. It is important, too, that this is the first periodic relationship of the elements to be discovered which is independent of the ordinary periodic system.

While the periodic variation of the abundance of the elements as here presented might have been discovered empirically, the fact is that the relations found were first predicted from the standpoint of theory, and were afterward found to be true. It therefore seems important to trace, at least in part, the reasoning by which this new law was discovered, even although its validity in no way depends upon the truth of the hypotheses which were used. However, the bearing of these new facts upon the important problem of the evolution of the elements, can best be understood if the subject is approached from a theoretical standpoint.

In the fifth paper of this series evidence has been presented for the theory that the variation in the chemical, and in such physical properties of the elements as cohesion, atomic volume, compressibility, coefficient of expansion, melting point, etc., depends first of all upon the arrangement in space of the negative electrons in the atom external to the nucleus, and also upon the number of such electrons. The number of these electrons presumably depends upon the nuclear charge. The structure of the nucleus determines what is called the stability of the atom, since it is not considered that the atom breaks up unless the nucleus disintegrates. For example, when negative electrons are given off under the influence of light, or from other similar causes, the atom is said not to decompose, but only to become electrically charged (ionized).

Now, the earlier papers have shown that the 91 elements other than hydrogen, of our ordinary system, fall into two series. At least among the elements of lower atomic weight, the atoms which have even atomic numbers are in general built up from helium atoms, and therefore may be said to have the general formula nHe', where the prime is added to indicate that these elements are intra-atomic, not chemical, compounds. The odd-numbered elements, beginning with lithium, seem in general to have the formula nHe' +  $H_3$ '. Thus these elements fall into two series which may be distinguished as even or odd.

Now, if the variations in the chemical and the ordinary physical properties of the cohesional type (properties of aggregation) are dependent upon the arrangement and number of the electrons external to the nucleus, which seems probable if the nucleus is extremely minute, as the results obtained by Rutherford and his students<sup>5</sup> seem to indicate, then the differences between the odd- and even-numbered atoms with

<sup>5</sup> Their results were obtained by the scintillation method. The photographic method, as applied by Harkins, Ryan, Shadduck, and Shah shows the apparent small size of the nucleus even more simply.

respect to nuclear structure would not be expected to be apparent in these properties. That this expectation is confirmed is easily seen, since neither with respect to chemical nor physical properties is there any difference apparent which shows a variation in periods of two elements from even to odd, or odd to even.

#### 2. The hydrogen-helium system and the abundance of the elements

It might be expected, however, that the composition of the nucleus should affect its own stability, which from radioactive evidence means the stability of the atom. From this standpoint it might be reasonable to suppose that the atoms of one of the series, the even or the odd, should be more stable than those of the other. Now, unfortunately, there is no known method of testing the stability of the lighter atoms, but it might seem. at least at first thought, that the more stable atoms should be the more abundantly formed, and to a certain extent this is undoubtedly true. If then, at the stage of evolution represented by the solar system, or by the earth, it is found that the even-numbered elements are more abundant than the odd, as seems to be the case, then it might be assumed that the even-numbered elements are on the whole the more stable. However, there is at least one other factor than stability which must be considered in this connection. The formula of the evennumbered (light) elements has been shown to be nHe'. Now, since the formula for the odd-numbered elements is  $nHe' + H_3'$ , it is evident that, if the supply of H<sub>3</sub>' was relatively small at the time of their formation, not so much material would go into this system. This would be true whether the H3' represents three atoms of hydrogen or one atom of some other element.6

# 3. The composition of meteorites as related to the structure of complex atoms

There is, however, material available of which accurate quantitative analyses can be made, and which falls upon the earth's surface from space. The bodies which fall are called meteorites, and no matter what theory of their origin is adopted, it is evident that this material comes from much more varied sources than the rocks on the surface of the earth.

<sup>6</sup> If it were one element it might have an atomic number of 0, 1, or 2, and thus be a neutron, an atom of hydrogen, or an atom of helium, of mass 3. The stability relations are not favorable to the existence of such a unit as a stable atom.

In any event, it seems probable that the meteorites represent more accurately the average composition of material at the stage of evolution corresponding to the earth than does the very limited part of the earth's material to which we have access. At least it might seem proper to assume that the meteorites would not exhibit any special fondness for the even-numbered elements in comparison with the odd, or *vice versa*, any more than the earth or the sun as a whole, at least not unless there is an important difference between these two systems of elements, which is just what it is desired to prove.

A preliminary study of the most recent analyses of meteorites of different classes showed that, either for any one class or for the meteorites as a whole, the even-numbered or helium system elements are very much more abundant than those of the old-numbered or helium-hydrogen system. For a more detailed study use was made of the older but much more complete and more valuable data collected by Merrill and by Farrington (10), who suggest that the average composition of meteorites may represent the composition of the earth as a whole.

The results obtained by averaging the analysis of 318 iron and 125 stone meteorites, 443 in all, show that the first seven elements in order of abundance are iron, oxygen, nickel, silicon, magnesium, sulfur, and calcium; and not only do all of these elements have even atomic numbers, but in addition they make up 98.8 per cent of the material of the meteorites. Of the remaining elements present to a great enough extent to have an appreciable effect upon the percentage values, 7 are odd and 5 are even, but in all only 1.22 per cent are odd numbered, while 98.78 per cent are even. Of the iron meteorites, 99.22 per cent of the material is made up of even-numbered elements, and of the stone meteorites, 97.59 per cent.

The percentages by weight given above are proportional to the number of hydrogen atoms utilized in building up the respective atoms. The general relations are not at all affected if the atomic percentages (number of atoms per 100 present) are used. The six most abundant elements on this basis all have even atomic numbers, and in order of abundance are iron, oxygen, silicon, magnesium, nickel, and sulfur.

In studying the relative abundance of the elements the ideal method would be to sample one or more solar systems at the desired stage of evolution, and to make a quantitative analysis for all of the 92 elements of the ordinary system. Since this is impossible, even in case of the

earth, it might be considered that sufficiently good data could be obtained from the earth's crust, or the lithosphere.

However, there are several important factors which cause our knowledge of the quantitative composition of the earth's crust to be of much less value for the solution of our problem than it might seem to possess on first thought. In the first place the quantitative analyses which have been made represent the composition of only the mere skin of the earth, the depth of which does not exceed the ten to twenty miles caused by geologic displacements. The surface of the earth has been markedly influenced both by igneous processes which have resulted in magmatic differentiation, and by weathering, solution, and redeposition. For example, the common idea that sodium is a very abundant element undoubtedly has its origin in the fact that the solubility of its salts has caused their very considerable concentration in the oceans. Again, the fact that the salts of sodium are much more fusible than similar salts of the alkaline earths and most other metals in the rocks, has probably caused it to be segregated by magmatic solution and redeposition. Thus, while in the average igneous rock found on the surface of the earth there seems to be about 2.23 per cent of sodium, it is not improbable that this is a larger percentage than would be found if the whole material of the earth could be taken as a sample for analysis.

If the sun is next considered it is found that although a large amount of its surface is exposed to us for spectroscopic investigation, the spectroscope gives no accurate measure of the quantitative composition, and that its findings are largely influenced by the height in the gaseous envelope of the sun at which the observation is taken.

Table 4 in columns 10 to 13, gives the average composition of iron and stone meteorites, arranged according to the atomic numbers. It will be noted that the even-numbered elements are in every case more abundant than the adjacent odd-numbered elements. The helium group elements form no chemical compounds, and are all gases, so they could probably not remain in large quantities in meteorites. For this reason, and also because the data are not available, the helium or zero group is omitted from the table. The only criticism which could be made of the system of averaging used in these columns, which is that of including all accurate analyses, is that it places undue emphasis upon the iron as compared with the stone meteorites. However, since the two relations shown are true for each class of meteorites separately, it is evident that they will be true whatever system of averaging may be chosen.

The results given in the last two columns of table 4 are expressed

Average composition of meteorites, showing the predominance of the even-numbered elements, and of the elements of low atomic number

		PERC	ENTAGI	Percentage by Weight Stone Meteorites	EIGHT	WE	WEIGHT PERCENTAGE	ATOMIC	MIC	WEIGHT PERCENTAGE	TAGE	ATOMIC PERCENTAGE	TAGE	ATOMIC PERCENTAGE	MIC	ATOMIC PERCENTAGE 350 STONE TO	IIC TAGE TE TO
NUM- BER	ELEMENT	15 snal	126 analyses	ana	99 analyses	METEC	53 Stone Meteorites*	125 STONE METEORITES	ONE	A ve. 125	rage of 3 stone n	A verage of 318 iron and 125 stone meteorites	nd s	318 IRON METEORITES	RITES	10 IRON METEORITES	ON ITES
- <del></del>		Even	Odd	Even	Odd	Even	Odd	Even	Odd	Even	Odd	Even	Odd	Even	Odd	Even	Odd
1	Hydrogen		:		:	:	0.084	:	:	:	:	:	:	:	:	:	:
7		:	:	<u>:</u>	:		Trace	:	:	:	:	:	:	:	-:	:	:
က	Lithium	:	:	<u>:</u>	:	:	:		:	:	:	:	:	:	-:	:	:
4		:	:	:	:		:	:	:	:::::::::::::::::::::::::::::::::::::::	:	:	:	:	:	:	:
5		:	:	:	:		:	:	:	:	:		:		:	:	:
9	Carbon	0.06	:	:	:	0.150	:	0.12	:	0.04	:	0.14	:	0.03	:	0.12	:
7	Nitrogen	:	:	<u>:</u>	:		:	:	:	:	:	:	:	:	:	:	:
<b>∞</b>	Oxygen	36.02	:	35.82	:	36.290	:	54.70	:	10.10	:	25.87	:	:	:	53.16	:
6	Fluorine	:	:	:	:	:	:	:	:	:	:	:	:	:	:	:	:
10		:	:	<u>:</u>	:	:	:	:	:	:	:	:	:	:	:	:	:
11	Sodium	:	0.59	:	0.70	:	0.645	<u>:</u>	0.62	:	0.17		0.30	:	:	:	0.62
12	Magnesium	13.54	:	13.80	:	13.673	:	13.52	:	3.80	:	6.41	:	:	:	13.15	:
13	Aluminium	:	1.39	:	1.45	:	1.527	:	1.25	:	0.39	$\overline{}$	0.59	:	:	:	1.21
14	Silicon	18.41	: 	18.18	:	18.154		15.79	:	5.20	:	7.53	:	:	:	15.35	:
15	Phosphorus	:	0.0	<u>:</u>	0.11	:	0.113	: :	0.02	:	0.14		0.18		0.17		90.0
91	Sulfur	1.98	:	1.84	:	1.80	:	1.51	:	0.49	:	0.63	:	0.04	:	1.46	:
17	Chlorine	:	:	:	:	:	0.080	:	:	:	:	:	::	:	:		:
18		<u>:</u>	:	:	:	:	:	:	:	:	:	:::::::::::::::::::::::::::::::::::::::		:	:	:	:
19	Potassium	:	0.17	:	0.27	:	0.174	:	0.11	:	0.04	:::::::::::::::::::::::::::::::::::::::	0.04	:	:		0.11
8	Calcium	1.65	:	1.26	:	1.730	:	1.01	:	0.046	:	0.47	:	:	:	0.97	:
21	Scandium	<u>:</u>	:	<u>:</u>	:	:	:	<u>:</u>	:	:	:	:	:	:	:		:
		-		-	-											,	

22	Titanium	0.01	:	:	:	0.108	:	0.002	:	0.01	:	0.108 0.005 0.01 0.005		 :	:	0.005	:
23		:	:	:	:	:	Trace	:	:	:	:	:	:	-	<u>:</u>		:
24	:	0.28	:	0.58	:	0.321	:	0.13	:	0.09	:	20.0	:	0.01	-:	0.13	:
25	Manganese	:	0.14	:	0.36	:	0.224	:	90.0	:	0.03	:	:	:	:		90.0
56	:	24.32	:	24.32	:	23.313	:	10.57	:	72.09	:	52.93	:	90.64	<del>-</del>	2.79	:
27	Cobalt	:	0.05		0.05	:	0.017		0.02	:	0.44	<u></u> ::	3.31	<del>-</del> : :	. 59	:	0.04
28		1.31	:	1.26	:	1.527	:	0.54	:	6.50	:	4.52	:	8.50	-:	0.76	:
83	Copper	:	10.0	:	10.0	:	0.014	: :	0.005	:	0.01	$\dots  0.01  \dots  0.01  \dots  0.014  \dots  0.005  \dots  0.001  \dots  0.005  \dots  0.005  \dots  0.005  \dots  0.005  \dots  \dots  0.005  \dots  \dots  \dots  \dots  \dots  \dots  \dots  \dots  \dots  $	0.005	:	0.02	0.005	:
Tota		97.59	2.41	97.06	2.94	97.022	2.978	97.89	2.11	98.78	1.22	97.59 2.41 97.06 2.94 97.022 2.978 97.89 2.11 98.78 1.22 98.575 1.425 99.22 0.78 97.95 2.10	. 425	99.22	282	7.95	2.10
		<u> </u>	%001	100%	%	) []	100%	) 01 1	%	100% 100%	%	100%	20	100,	100%	100%	%
					-						-		-		١		

\*40, zirconium, none; 50, tin, none; 56, barium, none; elements 44, ruthenium; 46, palladium; 77, iridium; and 78, platinum, were found in stone meteorites, while 30, zinc; 33, arsenic; 51, antimony; 50, tin; 79, gold; 74, tungsten; and 92, uranium, were not found in the investigations reported by Merrill.

† It has been found by Farrington that the known falls of meteorites are in the ratio of 350 stone meteorites to 10 of iron.

graphically in figure 11, where the atomic number are given as abscissae, and the percentage abundance of the elements is plotted on the Y axis. This figure shows that there is a very marked periodicity in the abundance of the elements in periods of two elements each. The peaks which represent the even-numbered elements are shown to be extremely high in comparison with the troughs in which the odd-numbered elements lie

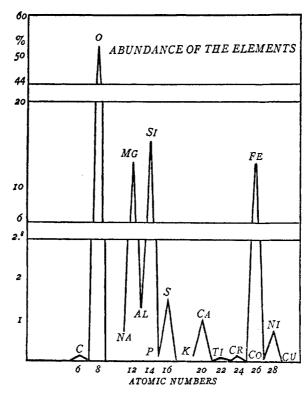


Fig. 11. Abundance of the Chemical Elements in the Meteorites (Atomic Percentage)

It is interesting to note that where two high peaks for even-numbered elements lie adjacent, between them the abundance of the odd-numbered element also becomes high in comparison with that of the other odd-numbered elements. Thus, by far the most abundant of the odd-numbered elements, aluminium and cobalt, lie in each case between two extremely abundant even-numbered elements.

This would seem to indicate that there is some relationship between the even and the odd series, and that the abundance not only follows the periods of two elements but also larger periods or waves, which cannot be said to have regularity in length and do not correspond with any of the divisions found in the ordinary periodic system.

# 4. A comparison of stone and iron meteorites with respect to their composition

Table 4 gives the average composition, in terms of both atomic and weight percentage, of both the iron and stone meteorites. This table shows in a very marked way that the extreme variation

TABLE 5

Abundance (percentage by weight) of the elements in the lithosphere, illustrating the predominance of elements of even atomic number

ATOMIC NUMBER	ELEMENT	EVEN	020
8	Oxygen	47.33	
14	Silicon	27.74	
13	Aluminium		7.85
26	Iron	4.50	
20	Calcium	3.47	
11	Sodium		2.46
19	Potassium		2.46
12	Magnesium	2.24	
22	Titanium	0.46	
		85.74	12.77

in composition between the iron and the stone meteorites does not affect markedly the predominance of the even-numbered elements. When calculated from the standpoint of the atomic percentages the iron meteorites contain less than 1 per cent of oddnumbered elements, and the stone meteorites less than 3 per cent.

It is very remarkable that in none of the average analyses presented does the percentage of any odd-numbered element, whether calculated by weight or by atomic proportions, rise higher than 1.53 per cent, which is the highest value given by Merrill for aluminium in any of his averages, while among the even-numbered elements large percentages are common, and range as high as 90.64 per cent.

5. The predominance of elements of even atomic number as illustrated by the composition of the earth's crust

Since it has been shown that the elements of even atomic number are 70 times as abundant in the meteorites as those of odd atomic number, it should be of interest to see if the relations on the surface of the earth are at all similar. It should be remembered, however, that the crust of the earth is much more local in its character, and cannot therefore be expected to give such good evidence as that presented by the meteorites. It is found, however, that the elements of even number again show their predominance, since of the five most abundant elements, four have even atomic numbers, while of the first nine, six are even numbered, according to Clark's average for the composition of the lithosphere, as presented in table 5.

The six even-numbered elements make up 85.74 per cent of the lithosphere, while the three odd-numbered elements amount to only 12.77 per cent.

Table 6 gives another set of data for all of the elements as recalculated from a recent estimate made by Clarke and Washington.

The latter table shows that in the twenty top miles of the earth's crust the atoms of elements of even atomic number are about nine times as numerous as those of odd atomic number.

Since the data presented for the composition of the lithosphere suffer from the disadvantage for the present purpose, that the surface of the earth has been subjected to extensive processes of differentiation, it would be of extreme value if evidence could be obtained in regard to elements which have not been thus affected. While this cannot be done, it would seem that elements which are very much alike both chemically and physically should be affected more nearly to the same extent than those which differ widely. Now the rare earths are so similar that they are separated in the laboratory only with great difficulty, so the writer first made an estimate of the relative abundance of these elements, and then secured estimates made independently by two noted authorities, Profs. C. James and C. W. Balke. It is of interest

TABLE 6
Abundance of the elements in the crust of the earth\*
(Atomic percentage for a twenty-mile crust)

ORDER	ELEMENT	ATOMIC	HYGRO-	LIGHT EL	EMENTS	HEA7	Y ELEMENTS
		NUMBER	GEN	Even	Odd	Even	Odd
1	Oxygen	8		57.44			
2	Silicon	14		18.23			
3	Hydrogen	1	9.48				
4	Aluminium	13			5.55		
5	Sodium	11			2.27		
6	Iron	26		1.68	No.	Ì	
7	Calcium	20		1.68	A.		
8	Magnesium	14	l	1.59	381	1	
9	Potassium	19			1.223		
10	Titanium	22		0.273		{	
11	Carbon	6		0.152			
12	Phosphorus	15			0.093		
13	Chlorine	17			0.088		
14	Sulfur	16		0.052			
15	Manganese	25			0.041		
16	Fluorine	9			0.030	į	
17	Chromium	24		0.024		Í	
18	Nitrogen	7			0.022		
19	Vanadium	23			0.015		
20	Lithium	3			0.014		
21	Zirconium	40				0.012	i
22	Barium	56				0.011	
23	Nickel	28		0.010			
24	Strontium	38				0.007	
25	Ce-Yt		·		i '	(0	0.0037)†
26	Copper	29	1				0.0033
27	Glucinium	4	ł	0.0019			Ī
28	Boron	5			0.0017		
29	Zinc	30				0.0012	
30	Cobalt	27			0.0010		
31	Lead	82			[	0.0002	
Tota	al		9.48	81.1329	9.3487	0.0314	0.0033 7)† = 100.00

<sup>\*</sup> Recalculated from a recent estimate by Clarke and Washington.

<sup>†</sup> In the analyses for these elements those of odd and those of even atomic number are reported together.

TABLE 6-Continued

0.000,0×	0.000,00×	0.000,000×	0.000,000,0×	0.000,000,000×
Rubidium Arsenic Molybdenum Tin Bromine Scandium	Cadmium Mercury Iodine Caesium Antimony	Bismuth Tungsten Thorium Cb-Ta Uranium Silver Selenium	Platinum Tellurium Gold	Thallium     0.000,000,000 × Indium Gallium     0.000,000,000,00 Germanium     0.000,000,000,000 × Radium

that these three estimates were all in agreement. They are given in table 7, in which c indicates common in comparison with the adjacent elements and r represents rare; ccc represents a relatively very common element, etc. The comparison is only a rough one, but it indicates that the even-numbered rare earths are much more abundant than the adjacent ones which are odd.

TABLE 7

The predominance of even-numbered elements among the rare earths

ATOMIC NUMBER	ABUNDANCE	ELEMENT	ATOMIC NUMBER	ABUNDANCE	ELEMENT
55	С	Caesium	63	rr	Europium
56	ccc	Barium	64	r	Gadolinium
57	c	Lanthanum	65	rrr	Terbium
58	cc	Cerium	66	r	Dysprosium
59	r	Praseodymium	67	rrr	Holmium
60	√·· e {	Neodymium	68	r	Erbium
61 ·	rrr	Illinium	69	rr	Thulium
62	c	Samarium	[]		

A striking confirmation of the idea that each element of even atomic number is more abundant than the two adjacent elements of odd number was obtained in an extremely extensive investigation of the composition of rare earth minerals by Goldschmidt and Thomassen (11). Their quantitative results confirm the relation in an extremely remarkable way, as is shown in figure 12.

The above results may be summarized in the statement that

in the formation of the elements much more material has gone into the elements of even atomic number than into those which are odd.

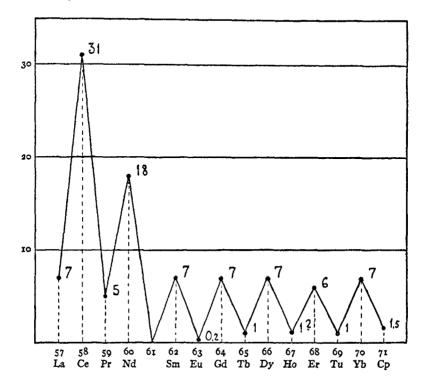


Fig. 12. Periodicity of Two in the Abundance of the Rare Earths as a Function of the Number, as Determined by Goldschmidt and Thomassen. The Height of each Point Represents the Abundance of the Rare Earth Element in Minerals on the Surface of the Earth

Obviously each element of even number (number given below) is very much more abundant than either adjacent element of odd number. This accords exactly with the predictions of the hydrogen-helium theory of atom building developed eight years before the work represented above was done by Goldschmidt and Thomassen.

### 6. The unknown elements

Of the ninety-two elements within the limits of our present system, all but two have been discovered. It is of interest to note that both of the unknown elements have odd atomic numbers, and that they lie in the radioactive region.

7. The greater stability of radioactive elements of even number

Among the radioactive elements it will be seen too that if the most stable isotope of each element is considered, then in each case the odd-numbered element has either a shorter period than the adjacent even-numbered elements, or else is entirely unknown. This is shown by table 8.

8. Classification into light elements (atomic numbers 1 to 27) and heavy elements (atomic numbers 28 to 92)

It has been seen that an important natural classification of the elements with respect to their nuclear characteristics is into those

TABLE 8
Half life of the most stable isotope of the radioactive elements

ATOMIC	ELEMENT	ONE-HALF PERIOD OF MOST STABLE ISOTOPE				
NUMBER		Even	Odd			
92	Uranium	5 billion years				
91	Uranium X2	_	1.15 minutes			
90	Thorium	18 billion years				
89	Actinium	_	Period unknown, but almost certainly less than radium			
88	Radium	1730 years				
87			Element undiscovered			
86	Niton	3.85 days				
85		•	Element undiscovered			
84	Polonium	136 days				

of even and those of odd atomic number. A second important natural classification, introduced in 1915 (1, 12), is into light elements up to atomic number 27, and heavy elements between atomic numbers 28 and 92. Of the latter elements 81 to 92 may be classed as specifically radioactive.

9. Primary and secondary factors which determine the abundance of the elements

It is evident from the preceding discussion that there is a direct relation between the abundance of the elements and the atomic number, and that there is no very marked relation to the periodic system of Mendeléeff. This indicates that the abundance of the elements is determined in a primary sense by the stability relations of the complex atoms themselves, and also by the stability of any electron-proton groups, such as the alpha particle, from which they are built. However, it is obvious, even if in the formation of the elements there is no relationship to the ordinary periodic system, that in all of the differentiative processes which occur after the original formation, the chemical and physical properties would play their part, so that if any special material is taken for consideration, relationships to that system would appear more and more as the process of differentiation takes place.

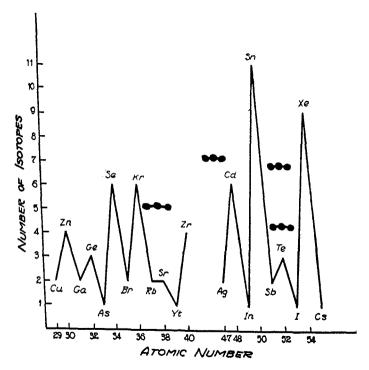
The fact that in the meteorites the elements of even atomic number are in all cases more abundant than the adjacent elements of odd number, seems to indicate that in such bodies the process of differentiation has not proceeded to such an extent as to cause the chemical and physical properties to have an influence sufficient to obscure in any marked degree the most general of the abundance relations due to nuclear stability, but it does not show that there has been an entire absence of differentiation. position of the earth's surface, as compared with that of the meteorites, shows a less rigid, but very marked adherence to the odd and even system, but certain groups of elements related in chemical and physical properties, are relatively more abundant. Thus the alkali metals, sodium and potassium, the halogens, and aluminium are present in much larger quantities. In the meteorites calcium is much less abundant than magnesium, while on earth both elements are of the same order of plentifulness.

## 10. The number of isotopes per element

It is evident that if the elements of even atomic number have in general more highly stable atoms than those of odd number, they should also have more kinds of atoms, that is a greater number of isotopes. The fact that this is true for the radioactive elements was used by the writer as one basis for the development of the relation, and was noted independently by N. F. Hall (13), to whom the idea was suggested by the relation as published.

The average number of isotopes of the known radioactive series averages 4.5 for elements of even, and 1.8 for those of odd atomic number.

That the periodicity is extremely striking in the case of ordinary elements is shown by figure 13 which gives the number of known isotopes for elements 29 to 40 and 47 to 55. This



• Fig. 13. Number of Isotopes per Element

periodicity is much less pronounced in the region defined by the writer as that of the "light elements," that is in the region of atomic numbers from 2 to 28.

VII. SECOND PERIODIC RELATION: HIGH STABILITY AND ABUN-DANCE FOR ATOMIC SPECIES OF EVEN ELECTRONIC NUMBER

The second periodic relation is even more general and important than the first. The number N which is defined as the difference between the protonic number P, and the atomic number Z, may be designated as the *electronic number*.

$$N = P - Z$$

Since P and Z are experimental quantities, N is also an experimental quantity. From the theoretical point of view N gives

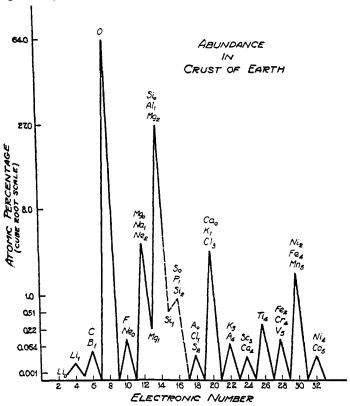


Fig. 14. Abundance of Atomic Species for Electronic Numbers 3 to 33

the number of negative electrons in the nucleus of the atom, and is a whole number.

In general atoms of even electronic number are extremely more abundant and stable than those of odd number, and the number of stable species of atoms is also very much larger. That high abundance is associated with even electronic number is exhibited in a remarkable way by figures 14 and 15 which represent the abundance in the earth's crust and in the meteorites respectively. The extraordinary nature of the contrast in abundance will not be realized unless it is noted that the *cube root* of the atomic per-

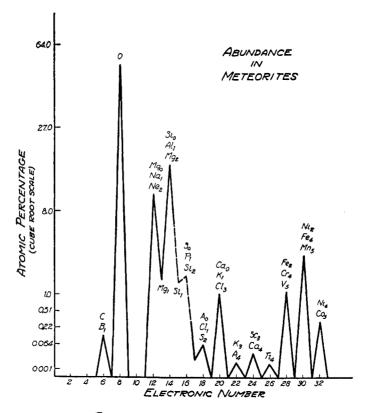


FIG. 15. ABUNDANCE OF ATOMIC SPECIES FOR EACH ELECTRONIC NUMBER

centage is plotted on the Y-axis, and the effect of the cube root plot is to obscure the great contrast in abundance. Thus the peaks are much higher in comparison with the troughs than they appear to be.

In general the abundance of the atomic species for any even electronic number is much greater than for the two adjacent odd electronic numbers. If the percentages themselves were to be plotted it would be necessary to make figure 14 more than 100 meters high in order to give the points for electronic numbers 3 and 5 the height given to them in the figure.

That the periodicity of 2 in the number of atomic species found for the atomic number appears also in the electronic number is exhibited by figure 16 also, since it shows that between electronic numbers 25 and 51 the number of known species for each even electronic number is much higher than for the two adjacent odd

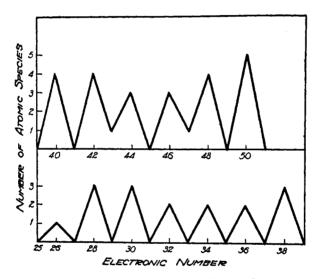


Fig. 16. Periodicity of Two in the Number of Atomic Species as a Function of the Electronic Number

numbers. Of the 112 atomic species thus far found between atomic numbers 2 and 60, only 16 have an odd, while 96 have an even electronic number.

Among the radioactive elements all of the most stable species are those of even electronic number: for example, the half-life of U is 5 billion years; of U<sub>2</sub>, 2 million years; of Io, 200 thousand years, and of radium 1730 years. The longest half-life for a species of odd electronic number is only 5 days (RaE).

Also, among the ordinary elements the most abundant isotope

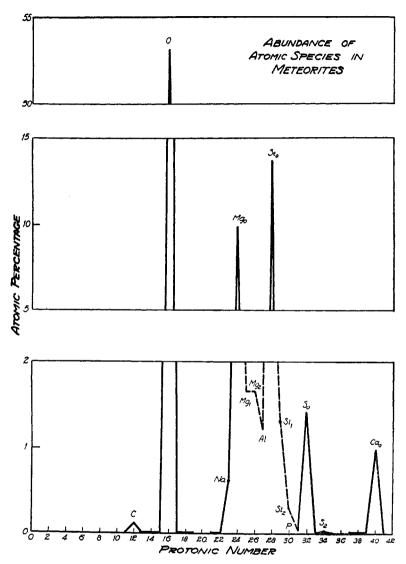


Fig. 17. Abundance of Atomic Species for Each Protonic Number (Atomic Weight)

of every element thus far investigated (atomic numbers 2 to 40, and 47 to 60) has an even electronic number, with the two exceptions of nitrogen and xenon.

VIII. THIRD PERIODIC RELATION: HIGH STABILITY AND ABUNDANCE FOR ATOMS WHOSE PROTONIC NUMBER (ATOMIC WEIGHT) IS DIVISIBLE BY 4, AND A SECONDARY PERIODICITY OF 2

The abundance of the light atomic species exhibits an extremely marked periodicity of 4 with respect to the protonic number.

TABLE 9

Atomic percentage of atomic species in meteorites, showing the importance of the weight 4

ELEMENT	ATOMIC WEIGHT						
SHIMBIN L	4q	3q	4q not 3q	3q not 4q	4q - 3	4q - 2	4q - 1
Carbon	0.12	0.12					
Oxygen	53.16		53.16				
Sodium	0.62		0.62		0.62		
Magnesium	9.86	9.86				1.65	1.65
Aluminium		1.21		1.21	1.21		
Silicon <sup>28</sup>	13.82		13.82				1.38
Phosphorus					0.06		
Sulphur	1.46		1.46				
Potassium		0.11		0.11	0.11		
Calcium	0.97		0.97				
Titanium	0.005	0.005					
Chromium	0.13		0.13				
Manganese	:		-		0.06		
Iron <sup>56</sup>	12.30		12.30				
Cobalt					0.04		
Totals	92.45		82.46	1.32	2.10	1.65	3.03

q = a whole number.

In figure 17, which refers to the meteorites every high peak in abundance occurs for species whose atomic weight is divisible by 4. Thus such peaks occur for oxygen, silicon (n = 0), magnesium (n = 0), sulfur (n = 0), calcium (n = 0), and carbon. Atoms in which the protonic number is even are much more abundant than those in which the number is odd; about 18

times as abundant in the meteorites and  $6\frac{1}{2}$  times as abundant in the crust of the earth, but this contrast is due mainly to the periodicity of 4, and only to a slight extent to that of 2.

The number of known species of even protonic number is found to be 71, and of odd protonic number 49, if the summation is made between protonic numbers 4 and 89, and between 109 and 146, that is in the regions most carefully investigated. Thus the emphasis upon even numbers is not at all as striking as if electronic numbers are involved.

The most abundant isotope of each element of even number from 4 to 28 has an atomic weight divisible by 4. Between atomic numbers 28 and 60, however, the atomic weight is divisible

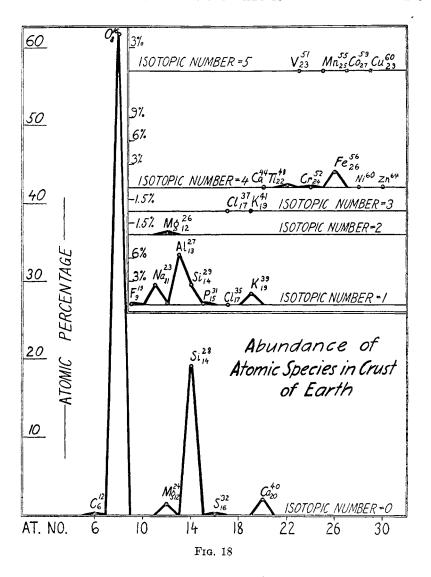
TABLE 10
Classification of atomic species according to even and odd number of electrons

		ABUNDANCE IN ATOMIC PERCENTAGE	
	Earth's crust	Meteorites	
Class I, N = even, P = even	87.4	95.4	
Class II, N = even, P = odd	10.8	2.1	
Class III, N = odd, P = odd	1.8	2.5	
Class IV, N = odd, P = even	0.0007	0.0	

by 4 in 8 known cases, by 2 but not by 4, in 5 cases. That is the periodicity of 4 becomes less general for even atomic numbers greater than 26, and is changed largely to a periodicity of 2.

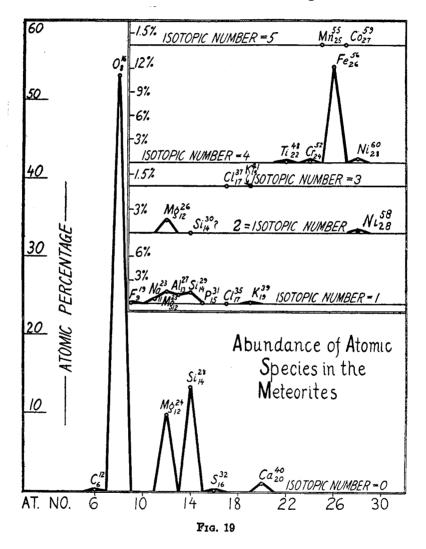
The extreme emphasis on the weight 4 (weight of the alphaparticle) is exhibited in table 9, in which q is a whole number.

The totals of the columns in a similar table for the earth's crust are almost the same, except that the column 4q - 3 adds up about 8 atomic per cent, and therefore is much more prominent. In both tables the abundance of the atomic species whose atomic weights are divisible by 4 is much greater than that of those divisible by 3, and the latter seems to gain its relative importance only through the inclusion of those weights which are common to the column 4q.



IX. CLASSIFICATION OF ATOMIC SPECIES (ISOTOPES) ACCORDING TO EVEN AND ODD NUMBER OF ELECTRONS AND PROTONS (15)

The atomic species represented in figure 10 may be classified in a perfectly natural way according to the type of line intercepts on which they lie in the figure, but more fundamentally according to the evenness or oddness of the number of electrons and protons present in the nucleus. The classification is given in table 10.



It is apparent that N is usually even whether P is even or odd, but that if N is odd, then P is also almost always odd. Thus

there is a certain matching of P with N. The class which is most rare, Class IV, includes only 3 atomic species, the lower isotopes of lithium and boron, and the only known isotope of nitrogen. Moreover, these species are all extremely rare. If governed by pure chance this class should contain 25 per cent of all of the atoms in the earth's crust, while it actually contains only about 1/40,000 of this amount. There are almost no atoms in which the number of nuclear electrons is odd and the number of protons even.

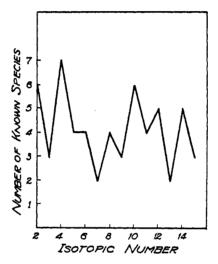


Fig. 20. The Number of Known Species of Atoms as a Function of the Isotopic Number

X. FOURTH PERIODIC RELATION: HIGH STABILITY AND ABUNDANCE FOR ATOMS OF ISOTOPIC NUMBER DIVISIBLE BY 4,

AND A SECONDARY PERIODICITY OF 2

Among the light elements (atomic numbers 6 to 28) there is a periodicity of 4 in the abundance of the atomic species as related to the isotopic number. This is apparent in figures 18 and 19, and tables 11 and 12.

Furthermore, as is shown plainly by figure 20, there is a periodicity of 2 in the number of species now known with respect

TABLE 11

Abundance of the atomic species in the meteorites as classified according to their isotopic and class numbers

(Atomic percentage)

		(Atomi	c percentag	e)	
ATOMIC NUMBER	SYMBOL	PER CENT	ATOMIC NUMBER	SYMBOL	PER CENT
Isot	opic number	= 0	I	sotopic numl	per = 1
	ss number	= I		Class number	= II
6	C	0.12	11	Na	0.62
8	0	53.36	13	Al	1.21  P = odd
12	${ m Mg^{24}}$	9.86	15	P	0.06 N = even
1 <b>4</b>	Si <sup>28</sup>	14.45	19	$\mathbf{K}$	0.11
16	$S^{32}$	1.42			
20	Ca40	0.97	Total	• • • • • • • • • • • • • • • • • • • •	2.00
Total		80.18			
Cla	ass number =	- IV		Class number	· = III
7	N	0.00	12	$\mathrm{Mg}^{25}$	1.65
			14	Si <sup>29</sup>	0.80 P = odd
			16	S <sup>33</sup>	0.01 N = even
			ļ———		2.46
	opic number			sotopic numb	
Clas	ss number	= I		lass number	= II
12	Mg <sup>26</sup>	1.65	17	Cl	0.00
14	Si <sup>30</sup>	0.20	19	$\mathbf{K}^{41}$	0.005
26	Fe54	0.60	ļ		
16	S84	0.03			
<b>2</b> 8	Ni*8	0.31	Total		0.005
Total		2.79			
Isot	opic number	= 4	I	sotopic numb	per = 5
	ss number	= I		class number	= II
20	Ca44	0.002	23	v	0.00
22	Ti	0.005	25	Mn	0.06
24	Cr	0.13	27	Co	0.04
26	Fe <sup>56</sup>	12.10	J		0.10
<b>2</b> 8	Ni <sup>60</sup>	0.19	Total	• • • • • • • • • •	0.10
			1		

It is worthy of note that the above percentages make up 99.9 per cent of all of the material of the meteorites, and the similar atomic species, atomic numbers up to 28 and isotopic numbers—1 to 5, make up 99.9 per cent of the earth's surface, which indicates that no increase in the ratio of electrons to protons (N/P) is able to sufficiently stabilize atoms with a positive nuclear charge greater than 28 as to make such atomic species any considerable factor in the abundance relations. The important feature of this relation is that the abundance does not fall off gradually but suddenly, and remarkably enough the fall in abundance comes just beyond nickel, while nickel is the first of the elements in the region of abundant isotopes. All abundant atomic species have isotopic numbers 4 or less.

to the isotopic number. The appearance of the plot (14, 23) represented by figure 10 seems to indicate that this periodicity will remain, at least between isotopic numbers 2 and 10, even after much more thorough investigations have been carried out.

The periodic relations are supposized in table 12.

The periodic relations are summarized in table 13.

TABLE 12

Abundance of the atomic species in the earth's crust as classified according to their isotopic and class numbers

ATOMIC NUMBER	SYMBOL	PER CENT	ATOMIC NUMBER	SYMBOL	PER CENT
	topic number $(N/P = 0.5)$ ss number	= 0 = I		opic number = s number =	= 1 = II
6 8 12 14 16 20	C O Mg <sup>24</sup> Si <sup>28</sup> S Ca	0.047 61.94 1.40 19.40 0.034 1.88	3 5 9 11 13 15 17	Li <sup>7</sup> B <sup>11</sup> F Na Al P Cl <sup>35</sup> K	0.009 0.086 2.64 6.43 0.089 0.025 1.42
	uss number =	84.70 IV		ss number =	10.70
3 5 7	Li <sup>6</sup> B <sup>10</sup> N	0.0007	12 14 16	Mg <sup>25</sup> Si <sup>29</sup> S <sup>23</sup>	0.22 1.50 0.0003
Total		0.0007	Total		1.76
Total isoto	pic number =	1 = 84.70	Total isoto	pic number =	2 = 12.46
	sopic number s	= 2 = I		opic number = s number =	= 3 = II
12 14 16 26 28	Mg <sup>26</sup> Si <sup>30</sup> S <sup>34</sup> (Fe <sup>54</sup> ) (Ni <sup>58</sup> )	0.21 0.30 0.0006 0.09 0.006	17 19 21 Total	Cl <sup>37</sup> K <sup>41</sup> Sc	0.007 0.07 0.00000 0.007
Total		0.60		ļ	

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ATOMIC NUMBER	SYMBOL	PER CENT	ATOMIC NUMBER	BYMBOL	PER CENT	ATOMIC NUMBER	SYMBOL	PER CENT
-	oic numb number	er = 4* = I		pic numb number			pic num s number	ber = 6 r = II
10 22	Ca <sup>44</sup> Ti	0.05	17 23	Cl <sup>29</sup> V	0.009	30	(Zn66)	(0.0000×?)
24 26	Cr Fe <sup>56</sup>	0.017	25 27	Mn Co	0.037	32	(Ge <sup>70</sup> )	
28 30	Ni <sup>60</sup> Zn <sup>64</sup>	0.004	29	Cu	0.0006(?)	36	Kr <sup>78</sup>	
Total	•••••	2.15	Total		0.047			<u>'</u>

<sup>\*</sup> It should be noted that in isotopic number 4 the relations of isotopic number 0 are repeated, but with a much smaller abundance. Thus both have a high abundance relative to the adjacent atomic numbers; in both Class I is prominent, and while Class IV has not yet been found in isotopic number 4, it may be present, but only in a very minute quantity. Isotopic number 5 repeats in a lesser degree the relations of isotopic number 1, again with the omission of the secondary class, Class III. While the numbers given in the table may be changed slightly in a few cases by the discovery of new isotopes, the atomic weight and abundance relations indicate that such changes will not affect the general relations presented.

TABLE 13
Periodicity of 4 in the isotopic number

ISOTOPIC NUMBER	ATOMIC P	ERCENTAGE
LEGIOFIC NUMBER	Meteorites	Earth's crust
0	80.1	84.7
1	4.5	12.5
2	2.8	0.6
3	0.005	0.08
4	12.4	2.2
5	0.1	0.05
6	0.000	0.000

# XI. THE NEW PERIODIC SYSTEM IN THREE DIMENSIONS (14, 15)

The new periodic system needs at least 3 dimensions in order to exhibit its characteristics. Consider figure 10 to be in a horizontal plane. Then the peaks of figure 18 (or figure 19) may be considered to rise above the respective lines of constant isotopic number (isoneutronal lines). That is if the X- and Y-axes

represent the atomic and isotopic numbers, the Z-axis (vertical may be taken to represent either the abundance or the stability of the atoms. Since there are, however, several types of stability, several such 3 dimensional figures are needed.

XII. FIFTH PERIODIC RELATION: ATOMS OF ODD PROTONIC AND ISOTOPIC NUMBERS (16)

If in figure 10 lines of constant *even* isotopic number are considered, the difference in the atomic number is almost universally 2, and the change of atomic weight 4, between species which are sufficiently abundant to be already discovered.

The number of horizontal steps or layers lying one above the other is from 4 to 7, for atomic numbers higher than 28. On changing to a consideration of the lines of odd isotopic number and atomic weight, a very different pattern is revealed. Here it seems common for the change of atomic number along a line of constant isotopic number to be 1, although a change by 2 remains prominent, and instead of from 4 to 7 horizontal steps lying one above another in the region of the heavier elements, there is often only a single step. Also, quite commonly, two steps lie one over the other, but this occurs most generally where there is a transition from a lower to a higher level in going from left to right. In only one case now known do 3 such steps lie above one another, and this is in tin, which is an element of even atomic number. Thus far, more than 2 isotopes have in no case been discovered by Aston or others for any non-radioactive element of odd atomic number.

While the periodicity of the pattern under discussion is 2 in the isotopic number, it has been mentioned that it is often 1 in atomic number; however, this means a periodicity of 2 in atomic weight. Thus, whenever on a line of constant isotopic number (isoneutronal line) the atomic number changes by 2, which is the more general change for the whole system, the atomic weight changes by 4 and the nucleus adds one  $\alpha$ -particle in going from left to right. However, when the change in the atomic number is only 1, as is common when the isotopic number and the atomic weight are odd, the addition is equivalent to a half  $\alpha$ -particle, or  $p_2e$ . Now, it is of great interest that the formula of any

atomic species of zero isotopic number is  $(p_2e)_z$ , since zero isotopic number is the fundamental base line of the system, and no nucleus which is complex is represented by a point below this line. Thus the hypothetical group  $(p_2e)^+$  seems to play a part which is secondary only to that played by its double  $(p_4e_2)^{++}$  which is the  $\alpha$ -particle. The writer has suggested the hypothesis that the first step in the formation of an  $\alpha$ -particle may be the union of 2 electrons with 1 proton to form the group, which is very difficult to break up, but readily unites with a like group to form an  $\alpha$ -particle. Sometimes it may add itself to a complex nucleus. The above relation may mean that it adds on more readily when the atomic weight is odd. However, there are so many other ways in which these nuclei might be formed that little weight should be given to this last hypothesis.

The system followed by the atomic species of odd atomic weight needs expression in a different way from that given above in order that it may be more fully understood. It was supposed by Harkins and Wilson (1) that in general isotopes of elements of even atomic number should have even atomic weights, and elements of odd atomic number should have odd atomic weights. Their supposition has, indeed, been justified. It may be stated, then, that normally elements of odd atomic weight and isotopic number have odd atomic numbers. However, in some exceptional cases the atomic number is even. The question arises, "When does this occur?" This question may be answered empirically by consulting figure 10. This figure shows that in all known instances up to and including atomic number 36, each atomic species of odd atomic weight and even atomic number lies on the same isoneutronal line (line of constant isotopic number) as, and directly between two adjacent atomic species of next lower and higher odd atomic number. However the condition for the existence of very small amounts of such isotopes may be that given in the following paragraph for heavier elements. Thus Be 1, lies adjacent to and between Li<sub>1</sub><sup>7</sup> and B<sub>1</sub><sup>11</sup>. Here the superscript gives the atomic weight, and the subscript the isotopic number. Se $_{9}^{77}$  lies in the same way between As $_{9}^{75}$  and Br $_{11}^{79}$ : Kr $_{11}^{83}$  lies between Br $_{11}^{81}$  and Rb $_{11}^{85}$ ; Mg $_{1}^{25}$  between Na $_{1}^{28}$  and Al $_{1}^{27}$ ; and Si $_{1}^{29}$  between Al $_{1}^{27}$  and P $_{1}^{81}$ .

As the number of particles in the nucleus increases, which occurs as the atomic and isotopic numbers rise, it is natural to suppose that the occurrence of abnormal types of atomic species would become more frequent, and the relations of periodicity less stringently followed.7 It is, therefore, not surprising that the condition for the existence of species of odd atomic weight, but even isotopic number, should be less exacting when the atomic number rises as high as 50 to 54. Here, in the elements tin and xenon, the condition seems to be that there shall be one adjacent atomic species of the same isotopic number and of odd atomic number. Thus Xe<sub>21</sub> lies just to the right of I<sub>21</sub>, but there is no isotope of caesium, of next higher atomic number than xenon, to the right. Also to the right of Xe<sub>23</sub> is Cs<sub>23</sub>, but to the left there is no isotope of iodine. While the element to the left of tin (indium) has not been tested for isotopes, the mean atomic weight of indium (which may not be accurate), and the known isotopes of antimony, seem to indicate similar relations for the type of isotopes under discussion, to those found in the case of xenon.

XIII. SIXTH PERIODIC RELATION: PERIODICITY OF TWO IN THE RELATIVE NEGATIVENESS OF NUCLEI WITH RESPECT TO THE ATOMIC NUMBER: PERIODICITY OF TWO IN THE MEAN ISOTOPIC NUMBER

The mean value for the elements of the ratio N/P varies with a general periodicity of 2 as is shown in figures 8 and 9. Here a peculiar relation is found. For the light elements up to atomic number 32 the mean value of N/P (or the isotopic number) is higher in general for elements of odd atomic number, while for heavier elements the higher values occur in general for elements of even atomic number. The extent to which these relations hold is shown by the figures 8 or 9.

<sup>&</sup>lt;sup>7</sup> The writer has shown that when the number of particles becomes abnormally large, as in the radioactive species, the regularity again increases, due to the fact that the less stable isotopes here become so unstable as to cease to exist in detectable quantities.

## XIV. HYPOTHESIS THAT THE NEGATIVE ELECTRONS IN ATOM NUCLEI ARE ASSOCIATED IN PAIRS

# 1. The negative electrons in atom nuclei seem to be usually associated in pairs

This idea is suggested by the fact that there are two electrons in the  $\alpha$ -particle, and that the number is even for almost all nuclei. Furthermore whenever electrons are emitted in the disintegration of the radioactive atoms, there are usually two successive emissions of an electron, and in the remaining cases

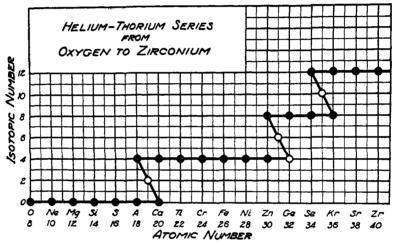


Fig. 21

one electron is emitted just before, and a second just after, the ejection of an  $\alpha$ -particle.

About 39 out of 40 known atoms have an even, and 1 in 40 an odd, number of negative electrons. It should be kept in mind, too, that if a nucleus contains 50 pairs of electrons, the presence of one odd electron would give oddness to the total number.

#### XV. ATOM BUILDING

## 1. Are atom nuclei built up from alpha particles?

a. The data and the periodic relations already presented in this paper give extremely strong support to the idea which led to the discovery of the relations: that atom nuclei are built up largely from or of alpha particles. Other evidence is outlined below.

- b. The radioactive atoms emit alpha particles in most of their disintegrations.
- c. A plot of the light atomic species (fig. 21) exhibits exactly the relations of a radioactive series (14), with respect to changes which correspond to the composition of an alpha-particle. Thus in the figure the series of 26 species contains every species but one which could be formed by a change in composition by an alpha-particle. It is remarkable that in this whole series there

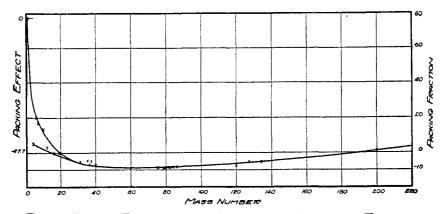


Fig. 22. Packing Effects in the Formation of Atoms from Hydrogen

is not a single species which does not belong to the heliumthorium series. That is, every one of the 19 species of an isotopic number 0, or which is divisible by 4, but with an odd atomic number, is missing, while every one of the 23 species of even atomic number, except one, has already been discovered. The exceedingly remarkable nature of this relation has not been appreciated.

Three pairs of beta-differences are represented in the figure. It may be noted that each of the intermediate beta-derivatives (19  $K_2$ , 31  $Ga_6$ , and 35  $Br_{10}$ ) is missing, but this is just what is to be expected from the stability relations of the radioactive series, since atoms which give the second of two beta-disintegrations are in general extremely unstable.

- d. A fast-moving alpha particle has been found to attach itself directly to the nucleus of a nitrogen atom.
- e. The work of Rutherford (17) and of Rutherford and Chadwick (18) shows that the light atoms of even atomic number are more stable with respect to a disintegration to give hydrogen than the adjacent atoms of odd number. This is additional evidence that periodic relation (I) developed by the writer is of fundamental importance.
- f. This difference between odd and even elements is shown also by the packing effects. It is found that the packing effect for very light atoms figure 22 is larger for even than for odd (Costa (20) and Aston (21)) atomic numbers. This indicates a greater general stability for atoms of even number. It is to be expected that this will be obscured in the heavier atoms, as is the case.
- 2. Energy of formation of helium from hydrogen; formation of alpha particles from protons and electrons (helium and hydrogen)

That the energy which would be liberated if hydrogen were to be converted into helium is very great was shown by Harkins and Wilson in 1915 (19). The amount of energy given off per mol. of helium formed was found to be  $2.8 \times 10^{19}$  ergs or  $6.7 \times 10^{11}$  calories, which is equivalent to  $4.57 \times 10^{-5}$  ergs per  $\alpha$ -particle formed. If the reaction were to occur in one step,

$$4 p + 2 e \rightarrow p_4 e_2$$

as was assumed in the early papers, then the radiation emitted should have a wave length of  $4.30 \times 10^{-4}$ Å, which is almost the value estimated as the wave length of the penetrating or cosmic rays.

However, it seems *possible* that the reaction may occur in two or more steps, since the probability of a sufficiently close approach of 4 protons and two electrons at one instant would be small at any ordinary concentration of protons and electrons.

The intermediate products which seem most probable are: (a) the neutron (pe); (b) a nucleus of isohydrogen ( $p_2e$ ). Thus an alpha-particle could be formed by the meeting of four neutrons and the loss of two electrons; or by the union of two neutrons to

form  $p_2e_2$ , and the loss of one electron to give isohydrogen  $(p_2e)$ . The alpha particle could then be formed either by the union of two neutrons with an isohydrogen nucleus and the loss of an electron, or by the union of two isohydrogen  $(p_2e)$  groups. If neutrons do not form, then it is possible that the primary process is the union of two protons with one electron to form isohydrogen.

It is obvious that if the formation of the helium nucleus occurs in several steps, the wave length of the radiation emitted would be greater than if it were to occur in only one step; and that the wave length emitted may depend upon the kinetic energy of the particles concerned. The union of a proton and an electron at low velocity to form radiation alone would give a wave length equal to  $3.3 \times 10^{-6}$ Å, while their union to form a neutron may be expected to give a wave length of the order of  $2 \times 10^{-3}$ Å.

Thus it seems probable that if the wave lengths of the spectrum of the cosmic radiation could be determined, some basis for a judgment as to the steps which occur would be obtained.

The beautiful work of Paneth and Peters (22) reported recently serves to demonstrate that if helium is formed at all from hydrogen activated by adsorption on a metal catalyst, the rate of reaction is extremely slow, since their extremely delicate tests revealed no helium.

# 3. Are neutrons used in the building of atom nuclei? (3)

There are two arguments for the idea that neutrons, as well as alpha particles, have been used in the building of atom nuclei.

 $^8$  Since this was written, Millikan and Cameron (Phys. Rev., 31, 921 (1928)) have found three bands ( $\mu=0.04$ , 0.08, and 0.04) in the cosmic radiation. The smallest amount of energy, which according to the Compton-Dirac equation corresponds with one of these coefficients, agrees well with the wave length given above for the formation of helium, but there are also two higher frequencies or lower wave lengths, and the processes which they represent are entirely unknown, although Millikan (Science, 48, 281 (1928)) seems to imply that such atom nuclei as those of silicon are built up in one direct step from protons and electrons. He also assumes that the nuclei of atoms of iron are built up in the same way. It should be realized that this would mean that 56 protons and 30 electrons, or 86 particles in all meet in space in the minute volume occupied by an atom nucleus or possibly a slightly larger volume, and then unite so suddenly that all of the energy is given off as a single quantum of energy. Other experimental evidence seems to indicate that an atom nucleus has a volume of only about a trillionth that of the atom.

The first of these is that, if neutrons exist, they could approach the positive nucleus of an atom without suffering the great repulsion experienced by an  $\alpha$ -particle, or a proton, or any other positively charged particle. This becomes an extremely important factor in the case of the atoms with high nuclear charge. Also, unless it is assumed that the radioactive elements have always existed, it is difficult to see from what source  $\alpha$ -particles with sufficient speeds could have come. That neutrons play a part in atom-building was suggested independently by Rutherford (22) and by the writer (3).

The second argument is that the relative abundance of the isotopes of the non-radioactive elements varies only slightly, while the abundance of the elements varies by a factor of the order of a billion. It must be admitted that this argument is partly illusory, since if isotopes vary greatly in abundance the least abundant ones are not discovered, as the sensitivity of the positive ray method is at present probably not better than 1 per cent. Nevertheless, the fact that the known isotopes vary as little in abundance as is the case, is remarkable, especially among the elements of atomic number higher than 29.

If a single neutron is added to an atom nucleus the atomic number remains constant, but the isotopic number increases by unity. Thus magnesium has abundant isotopes of numbers 1 and 2, which may have been formed from the fundamental isotope of number 0, and just the same isotopes are also found in silicon and sulfur. That neutrons have not been discovered, is not a valid argument that they are non-existent, since they would have no known ordinary property except mass. Therefore they could have been discovered only by finding that the chemical analysis of a body does not indicate 100 per cent of known elements. They would not adhere to ordinary substances, and could pass through atoms without noticeable effects except when they approach atom nuclei or electrons with extreme closeness (say to a distance of the order of  $10^{-12}$  cm.). Their concentration in ordinary materials should therefore be very small.

The only evidence which hints at their non-existence is the fact that all known complex nuclei exhibit ratios of N/P between

0.5 and 0.614 whereas this ratio in the neutron would be 1.0, or very much greater than in ordinary stable nuclei. Also this ratio does not exceed 0.572 in the case of any known light atom. This suggests that a single neutron (pe) may unite readily with a proton to form  $(p_2e)$  in which the ratio of N/P is 0.5 or that exhibited by the majority of complex nuclei in existence. It may

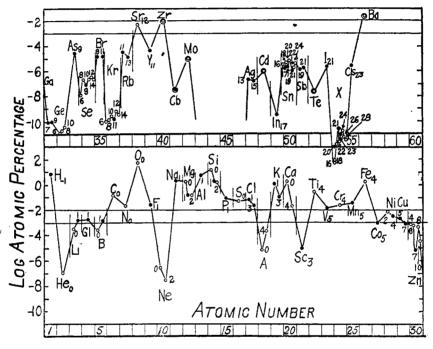


FIG. 23. ABUNDANCE OF THE ATOMIC SPECIES ON EARTH Where isotopes are given they are designated by their isotopic numbers

also be assumed that neutrons may also unite with complex positive nuclei, but that a large number of neutrons will not exist as such without additional protons in any complex nucleus, but will unite with protons to form more stable groups, such as  $p_4e_2$ ,  $p_2e$ , etc. It is of interest that the other groups suggested by the composition of complex nuclei are the double neutron  $(pe)_2$ , and the group  $p_3e_2$  which is equivalent to  $pe + p_2e$ .

## 4. Abundance of atomic species on earth

Figure 23 shows that there is an enormous difference in the abundance of the various elements in the crust of the earth. The abundance of any elment in the figure is the sum of the values given for its isotopes. Obviously the abundance of known isotopes of non-radioactive elements cannot vary greatly, since the existence of the isotopes is determined by analyzing a specimen of the element, and the sensitiveness of the methods used thus far is not sufficient to detect small percentages.

#### XV. FIRST PERIODIC RELATION AND THE BUILDING OF THE EARTH

In closing it may be well to consider the bearing of the periodic system of the atomic species upon other seemingly unrelated theories or ideas connected with the origin of the earth upon which we live.

The familiar saying: "The exception proves the rule," might better be stated: "The exception to one rule may give evidence for another." This seems to be true of the relation that even numbered elements are much more abundant than the adjacent elements of odd number.

The exceptions to this rule give evidence in favor of the theory that the surface of the earth has been built up from small bodies and thus supports the planitesimal hypothesis of Chamberlin rather than the theory of gaseous condensation. Thus it is found that though neon and argon should be extremely abundant if the earth's composition were to represent the general composition of material, they are actually extremely rare. Evidently, they have escaped, which indicates that the earth must have been built from bodies too small to hold these elements by virtue of their gravitational attraction. It is extremely striking that no gaseous element incapable of forming solid or liquid material at ordinary temperatures, is at all abundant on earth. The ordinary gaseous substances, oxygen and nitrogen, seem to have been held mostly as solids or liquids.

Another set of facts agrees with this idea. Both chlorine and

A similar figure has been used by Aston.

nickel are mixtures of two different kinds of atoms. Now it has been found by Baxter for nickel, and by Stone and the writer for chlorine, that the composition of each of these mixtures is exactly the same on earth as it is in the meteorites.

Although chlorine may be separated in the laboratory by very simple physical means, the chlorine on earth does not seem to have changed in its composition in the whole period of existence of the earth's crust.

#### REFERENCES

- HARKINS AND WILSON, J. Am. Chem. Soc., 37, 1367 (1915); Proc. Nat. Acad. Sci., 1, 276 (1915); Z. Anorg. allgem. Chem., 97, 175 (1916).
- (2) HARKINS, J. Am. Chem. Soc., 39, 856 (1917).
- (3) HARKINS, J. Am. Chem. Soc., 42, 1965 (1920).
- (4) HARKINS AND RYAN, J. Am. Chem. Soc., 45, 2095 (1923).
- (5) BLACKETT, Proc. Roy. Soc. (London), A, 107, 349 (1925).
- (6) HARKINS AND SHADDUCK, Proc. Nat. Acad. Sci., 12,707 (1926); Z. Physik, 50, 97-122 (1928).
- (7) HOLUBEK, Z. Physik., 42, 704 (1927).
- (8) DURRANT, J. Am. Chem. Soc., 39, 621 (1917). KOSSEL, Physik. Z., 12, 265-9 (1919).
- (9) GEIGER AND MARSDEN, Proc. Roy. Soc. (London), 82A, 495 (1909).
   GEIGER, Ibid., 83, 492 (1910); Phil. Mag., 5, 604 (1913).
   RUTHERFORD, Ibid., 21, 669 (1911).
- (10) Farrington, Publications 120 and 151, Field Columbian Museum, Chicago, "Meteorites," 205 (1915); I, Geol., 9, 623 (1901).

Bvisse, Mem. Soc. Lits. Sci. Arts l'Aveyron, 7, 168.

MEUNER, "Cours. de Geol. Comparee."

Suess, "The Face of the Earth," (English translation), Vol. 4, p. 543.

- (11) Goldschmidt and Thomassen, Videnskaps. Skrift. I. Mat.-naturw. Kl., 1924, No. 5.
- (12) HARKINS, Phys. Rev., 15, 79-94 (1920).
- (13) Hall, N. F., J. Am. Chem. Soc., 39, 1616 (1917).
- (14) HARKINS, Phil. Mag., 42, 306 (1921).
- (15) HARKINS, J. Am. Chem. Soc., 43, 1038 (1921).
- (16) HARKINS, Ibid., 45, 1426 (1923).
- (17) RUTHERFORD, Phil. Mag., 37, 538 (1919).
- (18) RUTHERFORD AND CHADWICK, Phil. Mag., 42, 809 (1921), 44, 417 (1922).
- (19) HARKINS AND WILSON, Phil. Mag., 30, 723 (1915).
- (20) Costa, Ann. Phys. (10), 4, 425 (1925).
- (21) ASTON, Proc. Roy. Soc. London, A, 115, 487 (1927).
- (22) PANETH AND PETERS, Ber., 59, 2039 (1926).
- (23) RUTHERFORD, Proc. Roy. Soc. London, 42, 1964 (1920).