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THE NATURAL SYSTEM OF ATOMIC NUCLEI

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Recently Barton[1] has shown several interesting regularities in the known atomic nuclei when these are plotted with the numbers of nuclear protons and electrons as coördinates. The known nuclei lie on this diagram between two lines whose slopes are 2 and 1.61,[2] the points for nuclei of low mass lie on or near the first line while, with increasing mass, the points approach the second line so that the occupied region of the diagram is a band, narrow near the origin and wider for nuclei of higher masses. Barton pointed out that several well-defined groups of nuclei, which he called "clusters," appear in this diagram such that each cluster possesses an approximate center of symmetry. These centers of symmetry he places at $P = 24, E = 12$; $P = 80, E = 45$; and $P = 124, E = 72$, while Urey and Miss Johnston[3] showed that there are indications of another cluster of radioactive nuclei with a center at $P = 222$, $E = 136$. In this paper it will be shown that there are indications of another cluster with center at $P = 38$, $E = 20$, that there is probably no cluster in the region of the diagram below A^{36}, and a correlation of these clusters with the stability of the nuclei as indicated by radioactivity and relative abundances of nuclei may exist. Further, a suggestion is made in regard to nuclei, as yet unknown, to be expected in the region from A to Cu.

Figure 1 is the proton–electron plot of the known nuclei from hydrogen to neodymium. The numbers of protons and electrons are plotted as abscissa and ordinate, respectively, with the scale drawn near the nuclei to facilitate comparison. Figure 2 is a similar plot of the radioactive nuclei in which it has been assumed that the nuclei of the actinium series have odd mass numbers.[4]

It is evident that the number of kinds of nuclei per unit length of the occupied band varies greatly with especially large numbers between $P =$ 65 and $P = 100$ and between $P = 110$ and $P = 150$. There is another dense region near the radioactive elements shown in Fig. 2. The pattern of the nuclei from Li^6 to A^{36} is very regular and leads one to postulate that all possible nuclei of this region are known or that any unknown ones are very rare. Between A^{36} and Cu^{63} is a region having a very low density of points scattered in what appears to be a very irregular way on the diagram.

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[1] Barton. *Phys. Rev.*. 35, 408 (1930).

[2] Harkins. *Chem. Rev.*. 5, 371–435 (1928). See this article for the many preceding references.

[3] Urey and Johnston. *Phys. Rev.*. 35, 869 (1930).

[4] Aston reports that Pb^{207} is present in sufficient amounts in Norwegian broeggerite to indicate that this lead is the end-product of the actinium series.

It is almost a certainty that all possible types of nuclei in this region have not yet been detected. The removal of Be^{8}, C^{13}, N^{15}, O^{17}, O^{18} and Ne^{21}[5] from the diagram would cause the plot in the region of these atoms to

Fig. 2.—The proton–electron plot of the radioactive nuclei and of Hg. Pb and Bi. Io and UY both occupy the point of $P = 230$, $E = 140$ and UX$_2$ and UZ probably the point $P = 234$, $E = 143$.

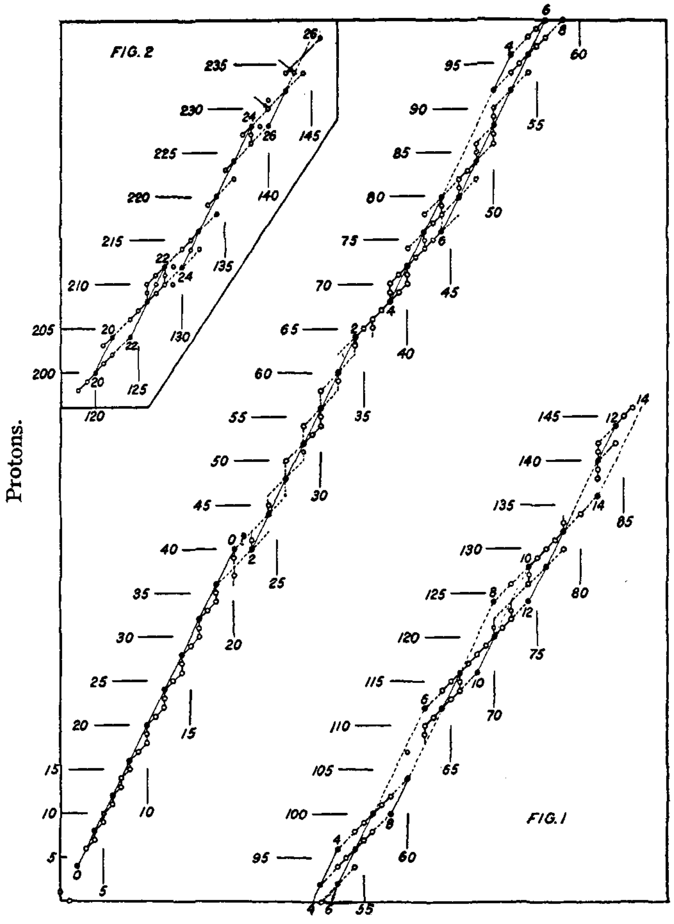

Fig. 1.—The proton–electron plot of non-radioactive nuclei. There may be a Ca^{41} since K^{41} is radioactive.

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[5] Be^{8}. Watson and Parker. *Phys. Rev.*, **37**, 167 (1931). C^{13}. Birge and King. *Nature*, **124**, 127 and 182 (1929); *Astrophys. J.*. **72**, 19 (1930). N^{15}. Naude. *Phys. Rev.*, **36**, 333 (1930); Herzberg. *Z. physik. Chem.*, [B] 9, 43 (1930). O^{17}, O^{18}, Giauque and Johnston, THIS JOURNAL, **51**, 1436, 3528 (1929). Ne^{21}, Hogness and Kvalnes, *Nature*. **122**, 441 (1928).

assume an irregular appearance similar to that of the region from A^{36} to Cu^{63} and we are led to expect that more refined methods will add many more points to the latter region.[6]

In searching for a possible regularity of nuclei in the region above A^{36}, we have well-known rules in regard to the relative abundance of nuclei of different types, due mainly to Harkins.[2] The most abundant nuclei of the earth's crust are those with proton numbers divisible by 4. The solid circles of Figs. 1 and 2 are such nuclei. From helium to Ca^{40} there are ten nuclei which are multiples of the helium nucleus in so far as the numbers of protons and electrons are concerned. From A^{40} to Zn^{64} there are seven nuclei which contain two electrons in addition to an integral number of helium nuclei. This region appears fairly complete in so far as this type of nucleus is concerned. Above Zn^{64} more of these "binding" electrons are required in these nuclei with mass numbers divisible by four. These nuclei are labeled as Series 0, 2, 4, ... in the diagram; the figures represent the numbers of binding electrons present in addition to those present in the α-particles or helium nuclei present.

From O^{16} to A^{36}, three nuclei in a vertical or diagonal row is a prominent feature of the diagram. If we draw lines through all such rows of three nuclei, we secure parallelogram figures as shown by the solid lines of Fig. 1. This feature is so prominent that the completion of the figures as indicated by broken lines is the next obvious step. In completing them, the rule that no element of odd atomic number has more than two isotopes has not been violated for no exceptions to this rule are known outside the radioactive elements.[7] The diagram as so drawn predicts the existence of many more nuclei than are now known, but methods used up to the present would probably not detect isotopes whose abundance is one-thousandth of the abundance of that of the most abundant one. The recent discovery of such rare isotopes by the molecular spectra method shows that the existence of more isotopes in this region is not only possible but very probable.

This pattern for existing nuclei in the proton–electron plot was derived as indicated above, but it is really only an expression of well-known rules due to Harkins[2] in regard to the abundance of nuclei in point of numbers in the earth's crust and in meteorites and the varieties of nuclei classified according to atomic number and nuclear electrons. These can be stated as follows.

(1) The most abundant nuclei, in point of numbers, have mass numbers divisible by 4 and are at the acute angles of the parallelograms. (2)

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[6] The points Cr^{50}. Cr^{53} and Cr^{54} have recently been added by Aston, *Nature*, **126**, 200 (1930).

[7] An isotope of chlorine of mass 39 has been reported by Becker [*Z. Physik*, **59**, 601 (1930)]. but not confirmed by Ashley and Jenkins (Washington meeting of the American Physical Society, April 30 to May 2, 1931).

Nuclei of even atomic number and even proton number are very abundant. In addition to the nuclei given under (1), this class includes the nuclei at the obtuse corners of the parallelograms. (3) Nuclei of even atomic number and odd numbers of nuclear electrons are less abundant. These occupy the midpoints on the diagonal sides. (4) Nuclei of odd atomic number and even number of nuclear electrons are less abundant and only two for each element are known. These occupy the midpoints of the vertical sides. (5) Finally, nuclei of odd atomic number and odd number of nuclear electrons are very rare, Li^6, B^{10} and N^{14} being the only known examples among non-radioactive nuclei. Such nuclei would occupy the centers of the parallelograms. In drawing in these parallelogram figures, then, we have only indicated the regions of the proton–electron diagram where it is most probable on the basis of the relative abundance of known nuclear types that other nuclei exist, with the additional assumption that these additional nuclear types will fall near the known nuclei having mass numbers divisible by 4.

The Structure of Nuclei.—This proton–electron plot with its parallelogram patterns suggests certain points in regard to nuclear structure. A nucleus may be considered as composed of (1) helium nuclei, (2) internal binding electrons, (3) additional protons and electrons. A chemical formula for a nucleus might be written $\alpha_n E_m p_k e_l$, where n gives the number of α-particles, m (always even) the number of binding electrons, k and l the numbers of additional nuclear protons and electrons, respectively. There is considerable evidence that the maximum possible number of helium nuclei is not present in all nuclei. Assuming that the protons alone contribute to the nuclear spin, it would be impossible to have nuclear spins greater than $^3/_2$, if the maximum possible number of protons and electrons were combined into helium nuclei having no spins. On the other hand, in the case of nuclei of low atomic number, either the maximum possible numbers of helium nuclei or other combinations of four protons and two electrons having about the same stability as helium nuclei are present. This is shown by the packing fraction curve, which shows a marked periodicity with atoms with mass numbers divisible by four having minimum packing fractions.[8]

Nuclei with mass numbers divisible by four may have additional binding electrons, 0, 2, 4, . . . , as indicated by the numbers of the series defined above. Latimer[9] has constructed a model of this type of nucleus in which extra pairs of electrons are bound in the interior of the nucleus in a regular way. Finally, the additional protons and electrons give the approximate parallelogram pattern of Fig. 1. Having a nucleus containing an integral number of helium nuclei, additional probable nuclei can be secured by

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[8] H. Olson, *Phys. Rev.*, 35, 213 (1930).

[9] W. M. Latimer, THIS JOURNAL, 53, 981 (1931).

adding a proton + a proton + a neutron + a neutron or a neutron + a neutron + a proton + a proton. (A "neutron" is used here as a convenient way of referring to a proton + an electron.) In the region where the series overlap, it is possible to have nuclei occupying the common obtuse corners of the overlapping series. There is the possibility that these two nuclei are not identical, for their formulas as written above differ in the number of internal binding electrons, E, and the number of external electrons, e. The internal electrons occupy some deep-lying position of the nucleus such as suggested by Latimer, while the outer electrons with the addition of two protons may form another α-particle within the nucleus. These formulas would be $\alpha_nE_mp_ke_l$ and $\alpha_nE_{m+2}p_ke_{l-2}$. Such a case is known, for Io^{230} and UY occupy the same position in the plot, which is one of these obtuse corners as can be seen in Fig. 2. UX_2 and UZ also occupy the same position but not such an obtuse corner position.

The nuclei of elements with odd atomic numbers lie toward the middle of the occupied band of this plot. In most cases no attempt is made to predict additional nuclei of odd atomic number and what predictions are attempted are based on this regularity only.

Barton's clusters seem to be due to the overlapping of these series of approximate parallelograms in certain regions. Wherever this occurs it will be possible to select some point in such a region that the rotation of half the figure through 180° will result in the many coincidences of points just as observed by Barton. The best illustration of this is given by his cluster whose center was located by him at $P = 80$, $E = 45$. Because of the discovery of additional germanium isotopes, this is no longer a very good center but the point $P = 78$, $E = 44$ is. This is due to the overlapping of Series 4 and Series 6 and the diagram as sketched in (without any thought of the cluster by the way) is completely symmetrical from $P = 68$, $E = 38$ to $P = 88$, $E = 50$. The isotopes of Mo and Ru recently reported by Aston[10] do not destroy this symmetry within these limits. It may be that nuclei will be found which will fill in the diagram and so destroy the symmetry. Another cluster may be postulated with center at $P = 38$, $E = 20$ (A^{38}) due to the overlapping of Series 0 and Series 2. The cluster with center at $P = 124$, $E = 72$, may be regarded as due to the overlapping of Series 8, 10 and 12.[11]

The cluster with center at $P = 38$, $E = 20$ is imperfect partly because there are fewer known nuclei above this point than postulated by the

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[10] Aston, *Nature*, **126**, 348 (1930); *Proc. Roy. Soc.*, (London). **A130**, 302 (1931).

[11] It may be said that the cluster at $P = 124$. $E = 72$ is only a symmetry of the isotopes of xenon and tin. In point of numbers this is true but from Cd^{110} to Ce^{140} it is possible to assume isotopes of elements of odd atomic number so that the symmetry in these nuclei is perfect and yet so that there are only two isotopes of odd atomic number differing in mass numbers by two. It would be necessary to add I^{125} to secure complete symmetry in the odd-numbered elements in this region.

parallelogram pattern of Series 2, and partly because the pattern below this point obviously consists of only half parallelograms. If these parallelograms were completed, the additional nuclei required would violate the rule that the number of nuclear electrons can never be less than half the number of protons except in the case of the proton itself. The pattern as drawn for Series 2 does not violate this rule.

The Relation of the Clusters to Nuclear Stability.—It is well known that the elements which occur with greatest abundance in nature have comparatively few isotopes. Thus the most abundant elements, oxygen, silicon and iron, have 3, 3 and 2 known isotopes, respectively, while the rarer elements may have many more. Moreover, the minimum of Aston's packing fraction curve comes near iron and, as mentioned above, the region from Ca^{40} to Cu^{63} evidently has comparatively few nuclear species. Moreover, radioactivity outside the characteristically radioactive group of elements appears to be confined to K, Rb and Cs and though these elements have only 2, 2 and 1 known isotopes, respectively, the elements of even atomic number near the latter two have many isotopes. Thus krypton and xenon have 6, and 9 known isotopes, respectively. Even if the positions outlined in Fig. 1 were completely filled in, there would still be a relatively large number of nuclear varieties in the neighborhood of K, Rb and Cs.

It seems possible to relate the clusters of nuclei to the regions of nuclear instability. This relationship can be illustrated most readily by means of the imperfect cluster at argon. Series 0 breaks off at Ca^{40} and if we assume that higher members of the series are too unstable to exist in detectable amounts, it may be expected that the last members of the series are relatively unstable as compared to the preceding members. On the other hand, Series 2 begins only with Cl^{37}, Cl^{39}(?) and A^{40}, which may mean that the series below this point is unstable and that the first members of the series are relatively so compared to members of the series with larger numbers of protons and electrons. Thus the overlapping parts of the two series will be relatively unstable, but nuclei of both series may occur. Many varieties of nuclei will be found, though due to the instability of these they will be comparatively few in actual numbers as compared to nuclei of elements outside these regions. In the region where the series do not overlap, the one series appears to be very much more stable than the two neighboring series, so that nuclei of only one series are present (referring now to the region of Series 0 and 2) and due to high stability these are present in large numbers. Thus the clusters in agreement with general and well-known facts should lie in regions of comparatively low nuclear stability.

By making the assumption that the relative abundance of nuclei is a measure of their relative stability, we can secure a partial confirmation of the suggestion made in this paper that the clusters should occur in regions of comparatively low stability. The relative abundance of different atomic nuclei should be a test for relative stability if a sample of matter could be studied which conformed to the following conditions: (1) the nuclei of the sample were synthesized from protons and electrons under such conditions that a close approach to thermodynamic equilibrium was attained; (2) since that synthesis, no changes in relative abundance occurred due to the synthesis or disintegration of some nuclei and not of others so as to cause deviations from the equilibrium mixture; (3) no fractionation of the elements has occurred. All available samples do not conform to the third condition certainly regardless of the first two conditions. Thus meteorites consist of three "phases," the stone, iron and troilite phases, of which the first approximates to the crust of the earth.[12] We may be very cautious in accepting the reality of any regularities in abundances which follow the periodic system of the elements, for this may be due only to the well-known chemical similarities. An excellent example is the extreme rarity of the inert gases in the earth's crust, due possibly to their inability to form high-boiling compounds with the other elements.

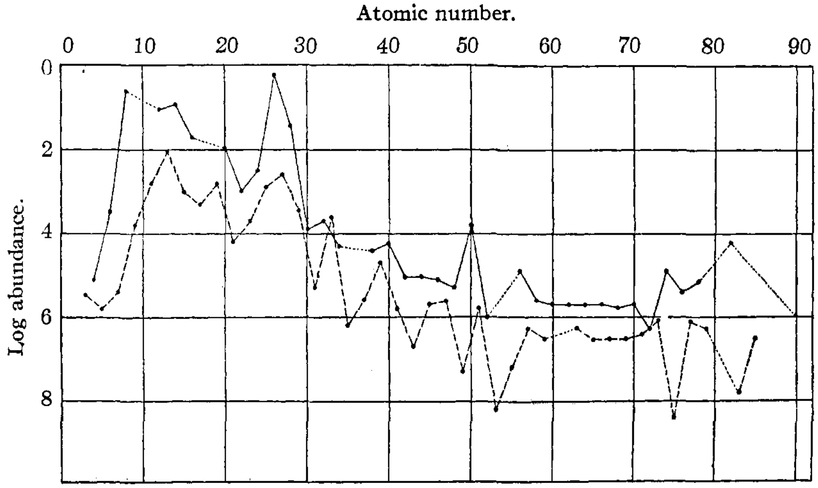

Fig. 3.—The logarithm of the atomic abundances in meteorites plotted against the atomic number. The abundances are expressed as fractions by mass taken from the data by Noddack and Noddack.

Figure 3 is a plot of the logarithm of the fraction of the meteorites by mass contributed by an element against its atomic number as given by the Noddacks. The solid line connects the points of the even-numbered elements and a broken line (long dashes) those of the odd-numbered ones. The short dark lines indicate that an intermediate element has not been

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[12] See Noddack and Noddack. *Naturwissenschaften*. **18**, 757 (1930).

detected in meteorites. The method of estimating the amounts of elements not directly determined is described in the legend of the figure.

The well-known Harkins rule that elements of even atomic number are more abundant than those of odd number is evident from the figure and there are at least two well-defined maxima in the even-numbered curve at oxygen and at iron and two in the odd-numbered curve at aluminum and cobalt. The minima between these come at titanium and at scandium. These minima do not coincide exactly with the suggested region of instability at chlorine, argon, potassium and calcium. The abundances of elements in the sun as given by Russell[13] follow a very similar curve. Titanium is relatively more abundant in the sun than in the meteorites, but potassium and calcium have rather high abundances. It seems impossible to decide definitely whether this disagreement in detail is real, especially since we have no assurance that the abundance curve is exactly related to stability anyway. Roughly, however, the abundance curve indicates that the elements of both Series 0 and Series 2 first increase in abundance, reach a maximum and then decrease again, and that there is a minimum near the region where the two series overlap.

Beyond copper the abundance curve is so irregular and the overlapping of the series so continuous that all correlation of the two is uncertain. The present data would indicate a complete contradiction to the assumption of a maximum of stability in each series, for the addition of Mo^{92} and Ru^{96} would indicate that Series 4 does not follow a regular course.

Whether a minimum occurs at neon is an interesting point. The curves as drawn from the data on meteorites certainly do not suggest the presence of such a minimum. Similar curves for the crust of the earth do, but the crust of the earth approximates the stone phase of meteorites as mentioned so that meteorites probably are the better "sample." An interpolation from the even-numbered curve would indicate that neon should be a very abundant element (between oxygen and magnesium), and the same conclusion is reached on the basis of Russell's curve for the elements in the sun. Yet the spectrum of neon has not been detected in the sun or stars.[14] Since Na^{23} has a nuclear spin greater than $^5/_2$, it may be that Ne^{20} does not consist of five helium nuclei and the rarity of neon may be due to a less stable arrangement of protons and electrons, an instability, however, which must be compensated in Na^{23}.

The system of nuclei suggested by Johnston[15] is very nearly identical

[13] H. N. Russell. "Contributions from the Mt. Wilson Observatory." No. 383, 1929.

[14] Latimer [THIS JOURNAL, 53, 981 (1931)] has suggested a very interesting model for nuclei which would indicate that Ne^{20} should be less abundant than O^{16} and Si^{28}. However. the model is not able to satisfy the writer in regard to the striking rarity of Ne^{20}, Ne^{21} and Ne^{22} with respect to F^{19} and Na^{23}, both of which are known to occur in the sun's atmosphere.

[15] Johnston. *ibid.*. **53**, 2866 (1931).

with that suggested here and all conclusions in regard to the relative abundances of the nuclei apply equally well to his system. He does not postulate the existence of so many additional nuclear species, but all those postulated here can be fitted into his scheme. In fact, his classification of nuclei and that of this paper appear to be only different presentations of the same scheme. Beck's[16] classification of nuclei is formally more like that presented by Johnston.

Harkins[2] has given a number of classifications of nuclei emphasizing particularly the use of his isotopic number. The writer believes that the simple proton–electron plot is simpler and more direct than these methods of classification, since it emphasizes the number of ultimate particles in nuclei of different types.[17] In view of the many systems of classification proposed, it would be difficult to originate another system which did not overlap previous ones in many points. This paper is written to emphasize the prominent parallelogram feature of these diagrams and the indication that additional stable nuclei are secured from those having most numbers divisible by four by adding protons and neutrons as indicated by these figures. In this way, it fits in very well with the recent interesting suggestions of Latimer.[9]

Summary

1. By using the proton–electron plot for atomic nuclei, predictions are made in regard to unknown nuclei between A^{36} and Cu^{63}.

2. The formula of a nucleus as consisting of helium nuclei, internal binding electrons, external protons and external electrons is proposed. It is shown that the isotopic isobars Io^{230} and UY may be expected.

3. It is suggested that Barton's clusters may be related to nuclear stability.

4. The system of nuclei proposed is compared to those proposed by other authors.

NEW YORK, N. Y.

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[16] Guido Beck, *Z. Physik*, **47**, 407 (1928).

[17] The writer is aware that Harkins has used the proton–electron plot, but the essential regularities brought out in Barton's paper and in this one are not at all evident from his papers [see *Chem. Rev.*, **5**, 387, 388 (1928) in particular].