Complex ions are usually considered in the same way as more simple ions, *i. e.*, to be more or less spherical. Pauling[11] has discussed the ionic radii of a few complex ions and their radius ratios. There is no literature which is opposed to assigning a definite skeleton structure to a complex ion of the type AX^+, and indeed such a structure appears to exist. Quantitative verification of the exact effect of the substituted anion in these oxalato complexes will be possible only after an x-ray determination of their structural types and cell dimensions. If the structural type is not altered by the substitution of the anion, then, as stated in the introduction, the axial ratios of a series of compounds which contain a complex ion will be adequate to measure the effect produced by the substituted anion.

Summary

1. The following new compounds have been prepared and analyzed: the chromate, dichromate and chlorate of the oxalato tetrammine series of cobalt, also the chromate and dichromate of the carbonato series.

2. Crystals of all the above compounds, except the carbonato dichromate have been prepared. These crystals, in addition to the oxalato perchlorate, were measured and the crystalline forms determined.

3. The difficulty encountered in preparing the oxalato chromate suggests an interesting example of the manner in which affinity of ions for each other may limit their deforming influence.

4. It is suggested that the complex oxalato cobalti tetrammine ion, and other acido complexes, may have a definite skeleton structure in crystals containing this complex ion, and that the substitution of various anions merely alters the shape and dimensions of this structure.

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[CONTRIBUTION FROM THE CHEMICAL LABORATORY OF THE OHIO STATE UNIVERSITY]

A PERIODIC ARRANGEMENT OF THE ATOMIC NUCLEI. THE PREDICTION OF ISOTOPES

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Interesting and suggestive regularities in the existence and abundance of mass numbers have been called to attention by various authors in recent years.[1] The most thorough tabulations of such empirical regularities are those of Harkins. The discovery of such relationships is useful not only in

[11] L. Pauling, THIS JOURNAL, **49**, 765 (1927).

[1] (a) Harkins, *ibid.*, **39**, 859 (1917); **42**, 1976 (1920); **43**, 1050 (1921); **45**, 1426 (1923); *Phil. Mag.*, **42**, 331 (1921); *Phys. Rev.*, **15**, 85 (1920); *J. Franklin Institute*, **195**, 554 (1923); *Chem. Reviews*, **5**, 371 (1928); (b) Beck, *Z. Physik*, **47**, 407 (1928); (c) Barton, *Phys. Rev.*, **35**, 408 (1930); (d) Latimer, THIS JOURNAL, **53**, 981 (1931).

offering a basis for the prediction of new isotopes but also in providing a key to the as yet obscure nature of nuclear structure. In these two respects such empirical tabulations play a role entirely analogous to that of the Periodic Table in the search for new elements and in the explanation of the structure lying outside the nucleus.

A new arrangement of the atoms in order of isotopic mass numbers is presented in Table I, which may be referred to as a Periodic Table of the nucleus. We believe that this arrangement possesses the advantage of easy comprehension and is, at the same time, suggestive both of unambiguous predictions of many undiscovered isotopes and of significant relationships with respect to nuclear structure.

In this table the atoms are first classified into four main types, differing in respect to the numbers of "free" protons in the respective nuclei. These four types are: the $4\ N$ type, with atomic weight evenly divisible by four, which may be arbitrarily regarded as composed of exactly N alpha particles; the $4\ N + 1$ type, made up of N alpha particles plus one "free" proton; the $4\ N + 2$ type, consisting of N alpha particles with two additional protons; and the $4\ N + 3$ type, with three extra protons. This classification is made arbitrarily from consideration of mass number alone and is not regarded as precluding the possibility that in some nuclei four or more "free" protons may exist without combination into an alpha particle. N, of course, differs for various atoms and, in general, may assume any value from 0 to about 60, the latter corresponding to the mean value of N for isotopes of uranium. Successive rows in the table are characterized by successive values of N, given for convenience at the extreme left of the columns for each main type. A further classification is carried out, based on the numbers of "free" electrons in respective nuclei. The numbers which stand at the heads of columns represent the numbers of *nuclear* electrons in *excess* of $2\ N$. These may be arbitrarily taken as nuclear electrons outside of alpha particles.

Atoms experimentally observed and reported in the literature[2] are shown in bold-faced type. O^{17} and O^{18},[3] C^{13},[4] N^{15},[5] Be^{8},[6] and Cl^{39},[7] were observed

[2] It is perhaps worthy of comment that about twenty-five isotopes have been reported since this table was prepared in its present outline several months ago. That these have fitted into the table without exception, as the reader may observe, inspires confidence in the use of the table for further predictions.

[3] Giauque and Johnston, THIS JOURNAL, **51**, 1436, 3528 (1929). See also reference 5 and Ruchardt, *Naturwiss.*, **18**, 534 (1930).

[4] King and Birge, *Nature*, **124**, 127 (1929); *Astrophys. J.*, **72**, 19 (1930); Birge, *Nature*, **124**, 182 (1929).

[5] Naude, *Phys. Rev.*, **36**, 333 (1930); see also Herzberg, *Z. physik. Chem.*, [B] **9**, 43 (1930). Note: Herzberg's prediction of N^{16} on a "theoretical" ground, and which he failed to find experimentally, is ruled out by Table I.

[6] Watson and Parker, *Phys. Rev.*, **37**, 167 (1931).

[7] Becker, *Z. Physik*, **59**, 583 (1930). Ashley and Jenkins failed to confirm this iso-

4N														
N	0	1	2	3	4	5	6	7	8	9	10	11	12	13
0														
1	He 4													
2	Be 8													
3	C 12													
4	O 16													
5	Ne 20													
6	Mg 24													
7	Si 28													
8	S 32													
9	A 36													
10	Ca 40		A 40											
11			Ca 44											
12			Ti 48											
13			Cr 52											
14			Fe 56											
15			Ni 60		?									
16			Zn 64		?									
17					Zn 68									
18					Ge 72									
19					Se 76		Ge 76							
20					Kr 80		Se 80							
21					Sr 84		Kr 84							
22					Zr 88		Sr 88							
23					Mo 92		Zr 92							
24					Ru 96		Mo 96		(Zr) 96					
25							Ru 100		Mo 100					
26							Pd 104		Ru 104					
27							Cd 108		Pd 108					
28							Sn 112		Cd 112		?			
29									Sn 116		Cd 116			
30									Te 120		Sn 120			
31									Xe 124		Te 124		Sn 124	
32											Xe 128		Te 128	
33											?		Xe 132	
34													Ba 136	
35													Ce 140	

4N + 1														
N	0	1	2	3	4	5	6	7	8	9	10	11	12	13
0	H 1													
1	Li 5	He 5												
2		Be 9												
3		C 13												
4		O 17												
5		Ne 21												
6		Mg 25												
7		Si 29												
8		S 33												
9			Cl 37											
10			K 41											
11			Sc 45											
12			V 49	Ti 49										
13				Cr 53										
14				Fe 57										
15				Ni 61										
16				Zn 65	Cu 65									
17					Ga 69	Zn 69								
18						Ge 73	Ga 73							
19						Se 77	As 77	Ge 77						
20							Br 81	Se 81						
21							Rb 85	Kr 85						
22							Y 89	Sr 89						
23								Zr 93						
24								Mo 97						
25								Ru 101						
26								Pd 105	Rh 105					
27									Ag 109					
28										Cd 113				
29										Sn 117	In 117			
30											Sb 121	Sn 121		
31												Te 125		
32												Xe 129		
33													Cs 133	
Xe 136													La 137	
Ba 140													Pr 141	

A PERIODIC TABLE OF

spectroscopically. The remainder were found with the mass spectrograph.[8,9]

tope in the band spectrum of AgCl [Washington meeting of the American Physical Society, April 30 to May 2 (1931)]. On the other hand, Hettner and Bohme [*Naturwiss.*, 11, 252 (1931)] claim to have confirmed Cl^{39} by new measurements on the 1.7μ band of HCl.

[8] Hogness and Kvalnes, *Nature*, **122**, 441 (1928).

[9] Aston, *Phil. Mag.*, **49**, 1199 (1925); *Proc. Roy. Soc.* (London), [A] **115**, 487 (1927); [A] **130**, 302 (1931); *Nature*, **122**, 167, 345 (1928); **126**, 200, 348 (1930); **127**, 233 (1931).

4N + 2

N	0	1	2	3	4	5	6	7	8	9	10	11	12	13
0		?												
1		Li 6												
2		B 10												
3		N 14												
4			O 18											
5			Ne 22											
6			Mg 26											
7			Si 30											
8			S 34											
9			A 38											
10			Ca 42											
11			Ti 46											
12			Cr 50											
13			Fe 54		Cr 54									
14			Ni 58		Fe 58									
15					Ni 62									
16					Zn 66									
17					Ge 70		Zn 70							
18					Se 74		Ge 74							
19					Kr 78		Se 78							
20							Kr 82		Se 82					
21							Sr 86		Kr 86					
22							Zr 90		Sr 90					
23							Mo 94		Zr 94					
24							Ru 98		Mo 98					
25									Ru 102					
26									Pd 106					
27									Cd 10		?			
28									Sn 114		Cd 114			
29											Sn 118			
30											Te 122		Sn 122	
31											Xe 126		Te 126	
32													Xe 130	
33													Ba 134	
34													Ce 138	
35													Nd 42	

4N + 3

N	0	1	2	3	4	5	6	7	8	9	10	11	12	13
0			?											
1			Li 7											
2			B 11											
3			N 15											
4			F 19											
5			Na 23											
6			Al 2											
7			P 31											
8			Cl 35											
9			K 39		Cl 39									
10					K 43									
11					Sc 47									
12					V 51									
13					Mn 55									
14					Co 59									
15					Cu 63	?								
16						Zn 67								
17						Ge 71	Ga 71							
18							As 75	Ge 75						
19							Br 79	Se 79						
20								Kr 83						
21								Sr 87	Rb 87					
22								Zr 91	Y 91					
23								Mo 95	Cb 95					
24								Ru 99	Ma 99					
25									Rh 103					
26									Ag 107					
27										Cd 111				
28										Sn 115	In 115			
29											?	Sn 119		
30												?	Sb 12	
													I 127	
Te 130														Xe 131
Xe 134														
Ba 138														
Ce 142														

THE ATOMIC NUCLEI

Isotopes as yet undiscovered but whose existence is predicted unambiguously by the regularities in the table, are marked lightly in squares bordered with hachuring. In three instances which occur in the $4N + 1$ column it is safe to predict only that either one, or perhaps both, of a pair of atoms must exist. These qualified predictions are indicated by means of hachuring at diagonally opposite corners. We do not feel that there is, in general, sufficient basis to predict either upward or downward extrapolations of members of columns. In a few instances observed regularities increase the probability of such extrapolations, indicated by question marks. However, the question marks which represent extrapolation to mass numbers 2 and 3 in positions which correspond to isotopes of hydrogen are intended literally as question marks.

The table is carried only to mass numbers of about 140 since the group of elements with mass numbers ranging from about 140 to 200 has received very little investigation. Lead, mercury, tungsten and bismuth fall regularly in order in the positions surrounding mass number 200, indicating that the regularities observed in the table persist to the heavier atoms. La^{139}, Nd^{144}, Nd^{145} and Nd^{146} are also known, and fit regularly into the table, but were omitted through an oversight.

Aston[9] has recently called attention to the existence of an isobaric triplet of mass 96. This table predicts the existence, as well, of isobaric triplets of masses 77 and 124. It is probable that several others exist in the latter half of Table I, although the uncertainty as to the upper and lower limits of the members of the columns does not permit definite predictions in other cases. It is not improbable that isobaric quadruplets may make their appearance among the heavy atoms.

We wish to point out certain very obvious regularities characteristic of the respective four chief nuclear types. The 4 N group is made up of a staggered progression occupying all *even-numbered* columns. The 4 $N + 1$ group, on the other hand, occupies *all* columns. The 4 $N + 2$ group makes its first progression in the manner of the preceding group *and then continues in the fashion of the 4 N group.* The 4 $N + 3$ group makes its first progression like the 4 N group, *and then continues like the 4 $N + 1$ group.* These regularities must possess some significance with respect to structural relationships within the nuclei.

We were at first inclined to doubt the authenticity of Ge^{77} until we observed at approximately the same position in column 7 of the 4 $N + 3$ group a sequence of at least seven atoms. We do not know what significance can be attached to these rather long sequences in the midst of groups consisting ordinarily of much shorter sequences.

In the light of the table we believe that we can make some modification of Harkins' abundance rules[1a] in a way which improves the usefulness of the latter. Thus Harkins' statement that atomic species which contain even numbers of nuclear electrons are much more plentiful than those which contain odd numbers can be better expressed as follows: Atoms made up of even numbers of protons as a rule do not contain odd numbers of nuclear electrons, while atoms of odd mass are distributed between even and odd nuclear electron types without essential distinction as to abundance. Likewise the distinction between species abundance (not gross abundance) of even and odd mass numbers seems to be overemphasized. Eighty-six known isotopes of even mass are listed in the table and nineteen more predicted with certainty. Sixty-four known isotopes of odd mass number are listed and twenty-three more are predicted. Nor is there any essential difference between the numbers of atomic species in the $4N$ and the $4N + 2$ groups. In terms of gross abundance, of course, O^{16} and Si^{28} alone furnish approximately three-fourths of matter as has been pointed out by Harkins[1a] and, in general, the prevalence of even mass types is well established. Even here there are frequent instances, particularly in higher atomic weights, in which the percentage of a particular species of odd mass among isotopes of a given element is considerably higher than one would be led to predict from a statement of the more general abundance ratios.

Since preparing the present table we have observed that the tabulation of Beck[1c] is capable of leading to the same predictions of isotopes as is the present arrangement, since it contains the same sequences of atoms which appear in the columns of Table I although the general plan of arrangement is quite different and, we believe, more difficult to comprehend. For atoms of mass $4N$, Latimer's[1d] tabulation is closely analagous to the present one. Professor Urey[10] has recently arrived at a, formally, quite different graphical plot of mass numbers which he informs us leads to predictions of isotopes nearly identical with those in this table.

We are at present engaged in the program of searching for predicted isotopes by means of spectroscopic analysis.

We do not attempt to offer, at present, any theoretical explanation of the observed arrangement.

Summary

A tabular arrangement of known isotopic species of atoms is presented, being essentially a Periodic Table of the Atomic Nuclei.

On the basis of the table, many undiscovered isotopes are predicted. The probable existence of certain isobaric triplets is pointed out.

Attention is called to significant relationships between the numbers of "free" protons and "free" electrons in atomic nuclei.

Certain of Harkins' abundance rules are restated in an amended form.

No theoretical explanation is offered for the observed regularities.

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[10] Urey, THIS JOURNAL, 53, 2872 (1931).