

than any others. The potassium hydroxide has the advantage of the alumina because it is easily prepared (by fusing the commercial material) but the alumina is much more easily handled.

COLUMBIA, MO.

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### NOTES.

**The Separation of Isotopes:** *A comment on the paper by T. W. Richards and N. F. Hall.*—In their recent paper,<sup>1</sup> T. W. Richards and N. F. Hall do not state the existing position very fairly with regard to the evidence upon which the chemical non-separability of isotopes is based. In their opening remarks they say that the methods used have been fractional crystallization and precipitation but these processes have seldom been carried out more than ten times in a particular case, and frequently six or seven crystallizations have been through a sufficiently thorough test of inseparability. I have no intention of giving a complete bibliography of the subject here, for I do it periodically in my Report on Radioactivity.<sup>2</sup> But I may mention that in the original work in which the conception of isotopes was first put forward, my wife carried out for me a very long fractionation of the barium-radium-mesothorium-I chloride derived from thorianite. I looked up the record and find in the preliminary work 80 fractionations were done. After that it did not seem worth while counting as the progress of the removal of the barium was quite regular. The original concentration of the barium to the radium was increased 340 times, but the ratio of mesothorium to radium was, within the 1 or 2% possible error of radioactive measurements, the same as before. In the technical separation of mesothorium and radium from monazite such fractionations are carried on from year to year continuously and the barium is eliminated completely, without separating in the least the mesothorium from the radium.

My work was published in 1910,<sup>3</sup> and the abstract in the Proceedings concludes, "The cases of complete identity between groups of radioactive elements differing presumably in atomic weight, which now appear to include all the radioactive elements of period of average life longer than a year, are in direct conflict with the principle of the periodic law." The general question was also discussed in the Annual Report on Radioactivity for that year. The authors cited attribute the generalization to Fajans, Russell, Fleck and Soddy, but in 1910 Fajans had not commenced his contributions to radioactivity and did not contribute anything on this subject to 1913. The generalization, that when an  $\alpha$ -ray is expelled in radioactive change the product occupies a place in the Periodic Table

<sup>1</sup> THIS JOURNAL, 39, 531 (1917).

<sup>2</sup> "Annual Reports of the Progress of Chemistry," published by the Chemical Society.

<sup>3</sup> *Proc. Chem. Soc.*, 26, 336 (1910); *Trans. Chem. Soc.*, 99, 72 (1911).

two places from the parent in the direction of diminishing mass and in subsequent changes in which  $\alpha$ -rays are not expelled reverts in some cases to the place it started from, I pointed out in 1911.<sup>1</sup> I think I can fairly claim to have originated both the conception of isotopes and the Periodic Law generalization, which, with regard to the one, Fleck in 1911-13, and with regard to the other, A. S. Russell and Fajans, independently, in 1913, made complete.

Every chemist who in 1917 considers that the chemical non-separability of isotopes is still insufficiently proved ought most certainly to follow the example of Richards and Hall and put their scepticism to the test. But there are some high standards to be bettered. One recalls, for example, that Auer von Welsbach, whose investigations in the most difficult field of chemical separation are world-famous, separated from 30 tons of Joachimsthal pitchblende, a mineral containing the majority of the known elements in detectable amount, a preparation, not much more than a millionth by weight of the whole, which behaved chemically as pure thorium. Anyone could tell in the dark that it was not, for its  $\alpha$ -activity was 30,000 times greater. This preparation has been subjected by Hönigschmid to the particular set of purification processes adopted in his recent revision of the atomic weight of thorium and compared spectroscopically with the thorium preparation used in that work. The spectra were identical and there was a complete absence of impurities. From the known period of ionium, and the  $\alpha$ -activity of the preparation from pitchblende, from 20 to 30% of the material must have been ionium, an element of atomic weight 230, instead of thorium. The atomic weight found, 231.51 instead of 232.12, confirms this proportion. Surely this evidence is even better than the mere repetition of the same process of purification an indefinite number of times as in the case of radium and mesothorium. But what is equally impressive is the unanimity for all the numerous cases recognized, bearing in mind the great number of researches and the sensitiveness of the methods to detect slight changes in the concentration ratio. Whether one covers the whole field, as Fleck first did, content to prove that each radio-element has an isotope to which it is *more* allied than to any of the hundred other known types of matter, or repeats on a single pair the same operation a great number of times, or applies to a single pair practically all the available methods of chemical separation, it is not by any one of these that the conception is to be judged, but by the unanimity of the conclusions from all three modes of work.

FREDERICK SODDY.

MARISCHAL COLLEGE, UNIVERSITY OF ABERDEEN.

<sup>1</sup> In the original edition of "The Chemistry of the Radio-Elements," published by Longmans, Green and Co.

**A Simple Ultra-Microscope.**—The need of an ultra-microscope in this laboratory has given rise to the instrument herewith described which has yielded good results in practice. The condensing system consists of a collimator removed from a spectroscope, the slit of which has been replaced by a one-inch objective. A five-hundred watt nitrogen lamp furnishes the illumination.

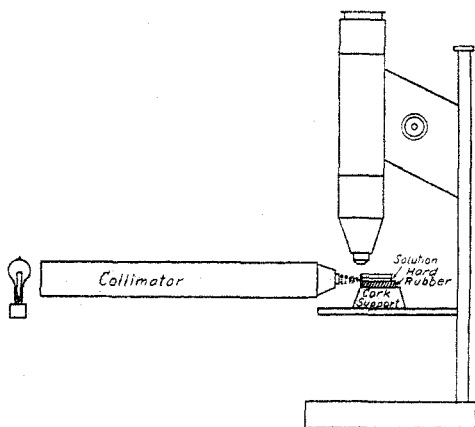


Fig. 1.

Fig. 2 shows the cell which comprises a flat piece of hard rubber and a square cover-glass, preferably of quartz. The polish is removed from the rubber with an abrasive so as to give a dead black surface. The edge of the cover against which the light is projected should be blackened. A depression is drilled at "D."

A drop of the solution to be tested is placed in "D" and a sufficient excess added to the slide to insure filling the depression and also the space between the slide and cover-glass, upon the addition of the latter. The edge of the cover-glass is brought flush with the end of the slide. The image of the lamp filament is focused on the edge of the solution film and slide. "D" should come just below the focal plane of the objective.

The apparatus described above is novel in its simplicity and ease of adjustment. The cell design is new and affords a thin film of solution under high illumination with a black background, the conditions most favorable for this type of work.

C. C. KIPLINGER.

UNIVERSITY OF MISSISSIPPI.

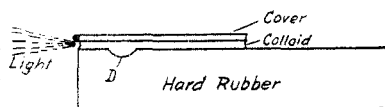


Fig. 2.

**On Periodicity among the Radioactive Elements.**—The recent papers<sup>1</sup> on atomic structure by W. D. Harkins and his co-workers have established the existence among the elements of a new periodicity of two which is distinct from the well-known (Newlands) periodicity of eight. This alternation, which appears to be the expression of nuclear structural variation, is seen in the abundance and the atomic weight of the lighter elements and in the life period of the heavier elements when the most stable isotope of a pleiad is considered. It is an interesting question

<sup>1</sup> THIS JOURNAL, 37, 1367 (1915); 38, 186 (1916); 39, 856 (1917); *Phil. Mag.*, 30, 723 (1915); *Proc. Nat. Acad. Sci.*, 1, 276 (1915); 2, 216 (1916).

whether any other characteristics of the elements show a similar variation in periods of two. As only properties which depend directly on the constitution of the nuclei of atoms may be expected to show such a variation, the radioactive elements are most promising subjects of study, since in them our information about the nucleus is most direct and definite.

Tabulation of the elements with atomic numbers 81 to 92 (those with the exception of neon, which are known to exhibit isotopy) reveals two regularities not mentioned by Harkins, nor clearly emphasized by others, which seem to be of the sort to be expected on the hydrogen-helium structure hypothesis. In the following table the first three columns are taken with slight amplifications from that given by Harkins.

TABLE I.

At. No.	Most stable isotope.	Abundance or life period of most stable isotope.	Radiation of most stable isotope.	Number of isotopes in pleiad.	Character of radiators in pleiad.	Predominant radiation of pleiad.
81	Tl	abun. <Pb	?	4	3 $\beta$ , 1 rayless	$\beta$
82	Pb(?)	abun. 0.002%	?	8	4 $\beta$ , 4 rayless	?
83	Bi	abun. <Pb	?	5	3 $\alpha$ , 4 $\beta$ , 1 rayless	$\beta$
84	Po	Per 136 days	$\alpha$	7	7 $\alpha$	$\alpha$
85	..	.....	...	0	.....	..
86	Nt	3.85 days	$\alpha$	3	3 $\alpha$	$\alpha$
87	..	.....	...	0	.....	..
88	Ra	1730 years	$\alpha$	4	3 $\alpha$ , 1 $\beta$ (?)	$\alpha$
89	Ac	<Ra, Th	$\beta$ (?)	2	1 $\beta$ , 1 $\beta$ (?)	$\beta$
90	Th	$1.8 \times 10^{10}$ years	$\alpha$	5	4 $\alpha$ , 1 $\beta$	$\alpha$
91	Bv	1.15 min.	$\beta$	1	1 $\beta$	$\beta$
92	U <sub>1</sub>	$5 \times 10^9$ years	$\alpha$	2	2 $\alpha$	$\alpha$

A graphical representation of the data in the 5th and 6th columns is given below.

The upper curve shows the variation, with the atomic number, of the number of known isotopes in a pleiad, and the lower shows similar variations in the character of the predominant type of disintegration. In drawing the lower curve, an alpha radiator is plotted as + 1, a beta radiator as -1, and a rayless isotope as 0. Only those isotopes are considered of which the position in the table is regarded as fairly well established: UY and RaAc' are not placed. No account is taken of possible differences between the isotopic end products of the two branches of each series. If these additional hypothetical isotopes are included in the lead pleiad, its predominant character becomes definitely rayless, and its membership then includes four beta radiators and seven rayless isotopes unless ordinary lead is identical with one of the end products, or is a mixture of two or more of them in constant proportions. In the bismuth pleiad the three C<sub>1</sub> members are only counted once each in the upper graph, but are reckoned as both alpha and beta radiators in the lower. Although no radiation from MsTh<sub>1</sub> or Ac has been detected, they are almost certainly shown to be weak beta radiators by their chem-

ical character and that of their products, and are listed as such with a query.

Evidently *even-numbered* isotopes are *more numerous* than those of odd number, and show a *well-marked tendency to undergo alpha rather than beta disintegration*; that this is to be expected from the hypothesis of Harkins

that even-numbered elements belong to the *helium* system is obvious. Harkins has already shown that they are longer-lived.

Of course these conclusions are rendered somewhat precarious by the facts that our knowledge of radioactive phenomena is probably far from complete, and that future refinements of technique may reveal a large number of new isotopes, or even show that some degree of radioactivity is a general property of matter. Still, as a first approximation, the data available seem to justify them. Even now, evidence of the relative instability of odd-numbered beta-ra-

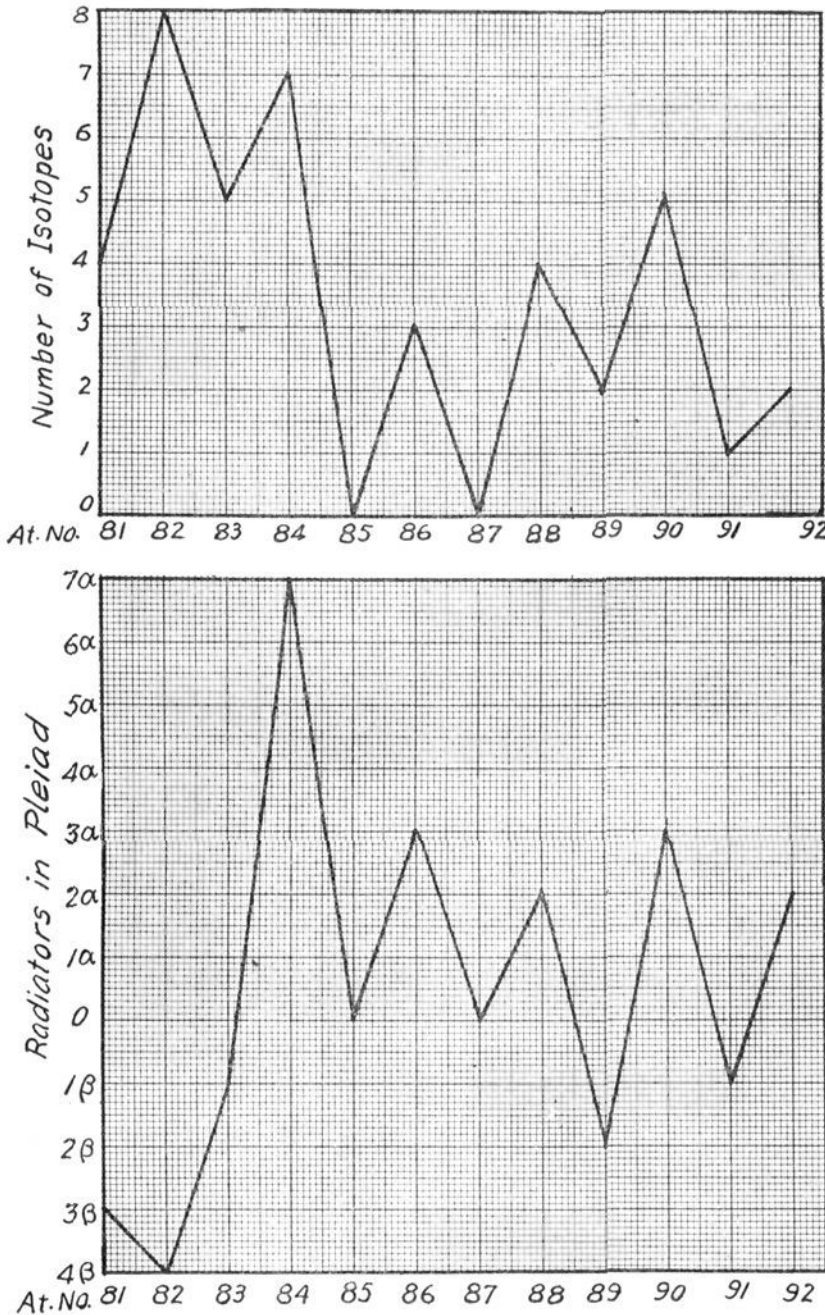


Fig. 1.

diating elements is not entirely lacking in the upper part of the table, since potassium and rubidium certainly give off beta particles. Such relationships as these can probably not be completely accounted for on a hypothesis of integration alone; we need a mechanism of disintegration

as well. Promising first steps in this direction have been taken by Lindemann<sup>1</sup> and Swinne,<sup>2</sup> following the lead of Rutherford and Bohr.

#### Summary.

The sharp alternation in atomic character with progressing atomic number, which Harkins has remarked in the abundance and the atomic weight of the lighter elements, and in the stability of the heavier, is here shown to extend to the *isotopic complexity* and the *predominant radiation* of the latter.

NORRIS FOLGER HALL.

CAMBRIDGE, MASS.

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**Elements neither Free nor Combined. A Third State.**—In the chemistry of our forefathers the elements were considered to be in either one of two chemical states, free or combined. If combined they may form a variety of combinations the bonds in which may be many or few, strong or weak; but these are differences in degree and not in kind; in all of them the element is said to be combined and not in its free state. In the free state several atoms may be combined to form a molecule, and this atomicity may be different for the same element when hot than when cold; but these again are differences in degree and not in kind; in all of them the element is said to be free and not in the combined state.

In modern chemistry, however, it would be very desirable to clearly recognize an intermediate or third state which is known to exist and which differs from either of the others in kind and not in degree. Confusion has arisen, some writers claiming it to be the free state, though with an apology, and others claiming it to be the combined state, also with an apology. The result is that the mind of the student is muddled, and confusion sometimes arises among chemists as to what is meant, because some definitions of terms have become indefinite. The purpose of this article is to urge that this third state be more generally recognized as such, and to show how this recognition would clarify and help to systematize our conceptions and to bring about a much-desired consistency.

When solid sodium chloride, for instance, is dissolved in water, it is known definitely, by the changes in the boiling and freezing points, that the number of molecules is thereby greatly increased, that is, that new molecules have been formed. The ionization theory states that these new molecules are those of sodium and chlorine which have in some mysterious way been separated so that they are no longer the original compound sodium chloride, but neither are they quite free sodium and chlorine, as they have not the chemical properties of these free elements. They are free yet not free, they are not combined yet the solution has the

<sup>1</sup> *Phil. Mag.*, 30, 560 (1915).

<sup>2</sup> *Physik. Z.*, 17, 481 (1916).

properties of the combined salt and not those of the free elements. To call them free is therefore as incorrect as to call them combined. The student who has been taught that there are only two states is therefore confused; and the physical chemists disagree as to whether these semi-free elements can be said to have been reduced or whether the real reduction occurs when they are subsequently freed by a current.

Changing a higher to a lower oxide has always been and is to-day recognized as a true process of reduction, yet it does not involve the freeing of the element; it is a step toward freeing, more of the same kind will ultimately free it. There may therefore be a true process of reduction without any freeing of the element. True reduction is a decrease of the valence, and it is only when the last one of the bonds is broken that the element has been set free.

Moreover when sulfur is set free from hydrogen sulfide, or oxygen from water, the freeing is accomplished by exactly the reverse process to reduction, namely oxidation, or adduction as it might better be called for reasons given below. Oxidation or adduction is the physical reverse of reduction, hence when the hydrogen in the above illustrations is reduced, its companion element is thereby subjected to the reverse operation, hence must be said to be oxidized during this reaction. Freeing an element therefore does not necessarily involve a reduction.

These semi-free, dissociated sodium and chlorine molecules, when in that state, are called ions and are said to be ionized because in that state (and only when in that state) they carry free + and — electric charges which the still combined molecules of sodium chloride do not carry. This term is well understood, is in common use and does not seem to be objectionable; then why not avoid the confusion which the older system of only two states has brought about, and simplify the teaching of this subject to students, by clearly recognizing that there are *three* distinct states, combined, ionized and free? The very fact that this ionized state has introduced such confusion because it is neither one or the other, and differs in kind and not merely in degree from both of the others, would seem to be a sufficiently good reason for recognizing it as a third state; the reasons are even better than those for the term "fourth state of matter" which is really one of degree rather than kind.

What the exact physical or chemical condition is, of these elements or radicals in this ionized state, may still be unknown, but a number of the properties of this state are known. It may be a sort of hydration in which neither the element nor the water has any bonds of the kind referred to by the term valence. But it is generally recognized and accepted that in electrolytes these dissociated or ionized elements or radicals have free electric charges while in that state; and that they do not have free charges while combined or after they have been set free by a current. They

obtain these charges by the process of dissociation or ionization, and they lose them again by the process of electrolysis.

Recognizing this as a third state would help to clarify and systematize some definitions and conceptions which are now controversial and inconsistent. Dissociation which implies decomposition and this in turn implies complete chemical separation, would then clearly be limited to changing a compound from its state of combination (solid salt) to this new state of ionization. The usual +, —, or other signs placed above the symbols of the elements would then clearly indicate that they are then in this ionized, semi-free state and are no longer completely combined, and the number of these signs would then indicate the number of unit charges or practical (enlarged) electrons called faradays<sup>1</sup> which each gram-ion of these semi-free elements then carries. It would show clearly that these elements are then not yet completely free but that relieving them of their charges will set them free; they are free only in the sense that the bonds between them have been broken, as they are now separate molecules, though they are still held together in some other way; their bonds have been replaced by the free charges which they have thereby acquired.

It would also establish clearly and definitely what the real physical process of reduction is, and would bring about a much-desired consistency. At present that term is vague; some apply it to the change from the combined to the ionized state, others to the change from the ionized to the free state, others to both together, leaving no terms for the two intermediate processes. It is universally accepted to apply correctly to changing a higher to a lower oxide either in solutions or by hot hydrogen when in the dry state; in the latter case there is no ionization yet there is a reduction of valence and a breaking of the bonds.

It is believed to be uncontested, and it brings about complete consistency, to define reduction as a decrease of valence, meaning by the latter term the number of existing bonds per atom which hold it in combination; hence when these bonds have been broken or destroyed the element is reduced. When iron is reduced from the ferric to the free state it has lost all three bonds; when reduced from the ferric to the ferrous state it has lost one; and when reduced from the ferrous to the free state it has lost the remaining two; all are correctly called reductions, whether produced by hot hydrogen or in solutions.

This seems to the writer to be the true and correct definition of reduction. To apply this to the ionized state, and thereby bring about consistency, the real physical process of reduction must be said to apply to the change from the combined to the ionized state, and not from the

<sup>1</sup> See "An Enlarged Electron of Practical Size: The Faraday," by Carl Hering, *Met. Chem. Eng.*, 16, 598 (1917).



latter to the free state, which latter process should be given a new name, as for instance some term implying the neutralization or discharging of the free charges of the ions; it is not always a freeing of an element, as for instance in the depolarizers in batteries, in the changing of ferric to ferrous solutions, etc.

The physical explanation of a chemical bond is thought by some to be the mutual attraction of positive and negative charges which when thus bound in a compound are neutral; when this bond is broken these charges are both freed. Hence free charges imply broken bonds; either the bond or the free charges exist, but not both simultaneously, as each one is converted into the other. Hence when the free charges exist (that is, after ionization) the bonds must have been broken, and as reduction is a breaking of the bonds it must have occurred during the ionization or dissociation.

This would in turn bring about consistency in other features also. In changing from the combined to the ionized state all the bonds have been destroyed and have been replaced by free charges; the cations, which are those which have been reduced by this ionizing process, have thereby gained positive charges or lost negative ones, and the anions, which have thereby been oxidized or adduced, have lost positive or gained negative ones. The signs over the symbols then indicate the kind of these charges and their number in faradays per gram-ion, of the elements which are then no longer combined. The three or the two + signs usually placed over the Fe in a ferric or a ferrous compound then mean that this ionized iron had been reduced by ionization from a ferric or a ferrous combination, respectively, its three or two bonds being changed into three or two charges, respectively; hence to say, as has been said in print, that because one has three and the other two charges the iron had lost one positive charge (gained a negative one) in being reduced from ferric to ferrous, is in the writer's opinion a wrong interpretation, it being only apparent but not real; they are both reduced, ionized iron but they contain a different number of charges, depending on their degree of oxidation (number of bonds) in the compound before the ionization took place, and in these un-ionized compounds the ferrous iron had lost one bond in being reduced from ferric; each of these bonds is subsequently convertible into a positive charge by ionization.

Reduction being, as the term implies, a loss or reduction of valence, and in a general sense a loss of bonds and companion elements, then means also and consistently a loss of negative electrons (faradays), while the reverse process, oxidation, which is a gain or addition of valence, and in a general sense a gain of bonds and companion elements, then means also a gain of negative electrons, for which reason (besides others) the

more appropriate and more consistent term adduction<sup>1</sup> is recommended in place of oxidation.

Modern developments in electricity compel us to recognize that a current of electricity consists of negative electrons moving in the opposite direction to the conventionally adopted arrow; our forefathers with whom the choice of direction was one between tweedledum and tweedledee, unfortunately guessed wrong. Hence it is the negative and not the positive charge that must be considered to be the unit of reference.

CARL HERING.

PHILADELPHIA, PA.

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## A SYSTEM OF ORGANIC NOMENCLATURE.

By AUSTIN M. PATTERSON AND CARLETON E. CURRAN.

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It has fallen to the lot of the authors, as part of their present work of indexing a very large number of organic compounds for the Decennial Index to *Chemical Abstracts*, to revise not only indexing methods but in many cases the names themselves in order to arrive at something like consistency. It is hoped that the following statement of the principles on which we have proceeded may be helpful to prospective users of the Index and also contribute, through discussion, to a clearer and more harmonious usage in naming organic compounds.

Ideally such a revision would be the work of an international, or at least a national, commission of organic chemists and free discussion would be of great value. Unfortunately the time allotment for preparing the Index has not permitted that exchange of opinion which the subject merits; this the authors fully realize and regret.<sup>2</sup>

We have not tried to invent a new system. Our aim has been to follow existing usage as far as it could be made fairly consistent, choosing what appeared to us good practices and rejecting bad, and introducing new features only when some very positive advantage was to be gained. In striving for names which would be suitable for general use, and not only for the Index, our tendency has been frankly toward compact naming and we have freely used short names for complex compounds as fresh starting points in naming derivatives of them. (This is the opposite to that of a work like Richter's "Lexikon," which reduces each compound to the derivative of as simple a parent substance as possible.) However, mere brief-

<sup>1</sup> See "Inadequacy and Inconsistency of Some Chemical Terms," *Met. Chem. Eng.*, 15, 649 (1916), and "Oxidation and Reduction in Physical Chemistry, Consistency of Terms and Conceptions," *Ibid.*, 16, 507 (1917), both by Carl Hering.

<sup>2</sup> Although the indexing work is now far advanced, the new plan of inverting the names so as to bring them under the parent compounds enables certain kinds of changes to be made readily. The authors will welcome criticism and adopt improvements or correct weak points as far as possible.