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THE PREDICTION OF THE RELATIVE STABILITIES OF ISOSTERIC ISOMERIC IONS AND MOLECULES

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When in the future we acquire a complete knowledge of atomic and molecular structure we shall be able to predict the stability of any suggested ionic or molecular configuration, provided that insuperable mathematical difficulties do not present themselves. In this paper it will be shown that in certain simple cases it is already possible, using the present conceptions regarding atomic structure and chemical combination and making plausible assumptions, to calculate qualitatively the relative stabilities of similar atomic aggregations.

Many cases are known of molecules or ions that contain the same numbers of atomic nuclei and the same numbers of electrons, but differ in the positive charges on the nuclei. Very often such compounds have pronouncedly similar physical properties. In the development of the modern theories of molecular structure these atomic groups have been supposed to have the same nuclear and electronic arrangement; for example, Lewis[2] assigns to the nitrogen molecule, the carbon monoxide molecule, the cyanide ion, and the acetylide ion the following structures, in which the letters signify the kernels of the various atoms, that is, the nuclei and accompanying K-electrons:

$$:\mathrm{N}:::\mathrm{N}: \qquad :\mathrm{C}:::\mathrm{O}: \qquad (:\mathrm{C}:::\mathrm{N}:)^- \qquad (:\mathrm{C}:::\mathrm{C}:)^=$$

Such molecules and ions are called *isosteric.*[3]

In some cases isosteric molecules or ions are also isomeric (composed of the same atoms), and differ only in the kernel arrangement; examples are the cyanate ion, $(\mathrm{NCO})^-$, and the fulminate ion, $(\mathrm{CNO})^-$. The relative stabilities of such isosteric isomers depend on their free energies, which like their energy contents are functions of their intramolecular energies. Since the two compounds contain the same atoms, their energy contents and free energies can be referred to the same reference substances. The identity in mass and similarity in physical properties of isosteric isomers would cause them to have the same or only very slightly different kinetic energies and entropies under the same conditions; hence, the free energy differences for these compounds would be nearly the same as the energy differences of their molecules.

[1] National Research Fellow in Chemistry.

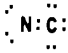
[2] Lewis, "Valence and the Structure of Atoms and Molecules," The Chemical Catalog Co., New York, **1923**, p. 127.

[3] Langmuir, THIS JOURNAL, **41**, 1543 (1919). Our attention has been called to the fact that our principle may be considered as a definite statement of a special case included in Postulate 3 given by Langmuir, *Science*, **54**, 59 (1921).

In order to determine qualitatively the relative stabilities of such atomic groups we shall use the following principle. The difference in the energies and free energies of isosteric isomeric molecules or ions has the same sign as the difference in the sums of the potential energies of their atom kernels with respect to one another, as calculated by Coulomb's law from the charges of the kernels and the distances between them. That is, the potential energy U of each molecule consisting of kernels with charges z_1e, z_2e, z_3e, ... at distances r_{21}, r_{31}, r_{32} ... from one another is given by the expression

$$U = e^2\left(\frac{z_2z_1}{r_{21}} + \frac{z_3z_1}{r_{31}} + \frac{z_3z_2}{r_{32}} + \ldots\right) + \text{electron energies} \qquad (1)$$

and the assumption is made that the energy terms due to the outer electrons (with respect to each other and to the nuclei) are, in considering the differences involved in isosteric isomeric molecules, not sufficiently large to cancel the effect due to the kernels. Furthermore, it is assumed that for the purpose of this calculation the interkernel distances can be taken as unchanged by a rearrangement of the kernels.

These assumptions seem to us to be not unreasonable. The fact that it is possible to interchange two atomic kernels without changing the arrangement of the external electrons indicates that the electronic arrangement about one of the two kernels is not greatly different from that about the other. For instance, it is possible that the cyanate ion has the structure  $:\mathrm{N}:\overset{..}{\underset{..}{\mathrm{C}}}:\mathrm{O}:$; and in this case interchanging the nitrogen and carbon atoms would involve removal of a kernel with a charge of five units from two unshared and one shared electron pair and its addition to two unshared and two shared electron pairs, and the reverse operation on a kernel with a charge of four units. The accompanying energy effect due to the electrons would accordingly be small.

On the other hand, it is not true that the interkernel distances would remain constant when the kernel arrangement is changed. Upon interchanging the nitrogen and carbon kernels in the cyanate ion, $(NCO)^-$, to form a fulminate ion, $(CNO)^-$, the end kernels will be more strongly repelled by the central kernel than they previously were. As a result of this greater repulsion the interkernel distances will be somewhat increased, and the difference in kernel potential energies of the two structures will be somewhat reduced; but inasmuch as the increase in interkernel distance is the result of the greater kernel potential energy of the fulminate ion, it is evident that this increase, while sufficient to decrease the kernel energy to some extent, will not bring it to a value smaller than that of the cyanate ion. Hence, the sign of the energy difference of the two isosteric isomeric compounds can be found by the use of Equation 1; but the absolute value of this difference will be considerably smaller than that given by this equation.

We propose, for the sake of simplicity, to limit ourselves mainly to a discussion of groups of atoms with small kernels, containing no electrons other than K-electrons. In calculating the kernel potential energy for a given configuration we have assumed that the K-electrons are sufficiently close to the nucleus so that the inverse square law can be used, with the effective kernel charge equal to the nuclear charge minus the charge of the K-electrons. We have taken the charge of the electron as unity, and used the Ångström as the unit of length. One energy unit in the table is then equal to 330,100 cal. per mole. It will be shown that the calculated differences in kernel potential energy for alternative configurations are of the order of one unit; it is evident that even though the effect of the electrons and of variations in the interkernel distance does neutralize the larger part of the calculated kernel effect, that remaining is still enough to account for great differences in stability. For example, it will be seen from the table that the calculated kernel energy difference of the fulminate ion and the cyanate ion has the extremely large value, 858,000 cal. per mole; without doubt the actual energy difference of the two ions is very much less than this, but still with the same sign.

The neglect of the effects due to external electrons allows us to apply the suggested principle without giving consideration to the actual distribution of these electrons, indeed, without even deciding whether to treat them as stationary, and distributed in pairs about the kernels, or as in motion in orbits about one or two nuclei. We shall, however, find it convenient to be more specific in discussing the chemical reactions considered; hence, we shall in these discussions assume definite electronic structures for the various compounds, and shall represent these structures by using double dots for electron pairs, as is done by Lewis. It is to be understood that we do not mean to imply that we consider the electrons to be stationary, for the symbol might be used to denote two electrons in orbits including either one or two nuclei; and it is to be strongly emphasized that the energy calculations and stability considerations made in this paper are independent of the assumption of any particular electronic configurations.

The Structure of Carbon Dioxide and its Isosteres.—There are a number of ions and molecules known containing three small kernels and sixteen external or valence electrons; namely, carbon dioxide, nitrous oxide, the trinitride ion, the acid fluoride ion, the cyanate ion, the fulminate ion, the metaborate ion and the beryllate ion. Of these the first five have been studied with X-rays, and in each case shown to have a structure in which the three atoms are collinear.[4] This result is verified by the infra-red oscillation-rotation absorption spectra of carbon dioxide.[5] It accordingly seems probable that all of these compounds are

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[4] For references and further discussion see Hendricks and Pauling, THIS JOURNAL, **47**, 2904 (1925).

[5] Barker, *Astrophys. J.*, **55**, 391 (1922).

isosteres, and possess the same stable arrangement of their external electrons; the acid fluoride ion may, because of the small charge on the proton as compared with the other kernels, possess a unique electronic arrangement.

It is generally believed that in the acid fluoride ion the proton is sharing only two electron pairs with other nuclei. The structure of this ion may then be represented thus: $:\overset{\cdot\cdot}{\underset{\cdot\cdot}{F}}:H:\overset{\cdot\cdot}{\underset{\cdot\cdot}{F}}:$. Possibly the same electronic arrangement is to be assigned to the other similar groups; however, it seems improbable that in a group such as the trinitride ion, NNN^-, the central kernel, with a charge of five units, should be surrounded by only four shared electrons. Of the alternative structures $\cdot\dot{\underset{\cdot}{N}}::N::\dot{\underset{\cdot}{N}}\cdot$ and $\cdot\dot{\underset{\cdot}{N}}:\overset{\cdot}{\underset{\cdot\cdot}{N}}:\dot{\underset{\cdot}{N}}\cdot$ we shall use the latter, having in mind the evidence recently adduced by Lewis[6] concerning the instability of the double bond, and the tendency of small kernels in some cases to surround themselves by six instead of eight electrons.[7] In this structure the four electron pairs are not arranged tetrahedrally about the central kernel, but are co-planar.

In every case the distance between adjacent atoms for these compounds as found by X-ray methods is 1.15 Å., within the rather wide limits of experimental error. In giving calculations in illustration of our arguments we shall adopt this distance throughout; similar results would be obtained with any other distance.

Carbon Dioxide and Nitrous Oxide.—Two atomic arrangements are possible for carbon dioxide, one in which the central atom is the carbon atom, with a kernel charge of four, and one in which it is the oxygen atom, with a kernel charge of six. It is evident that the potential energy of the first configuration is the smaller; numerical results are given in Table I. This structure, which is of course the actual one, would then be predicted on the basis of our principle.

The predicted stable structure for nitrous oxide is NNO, with the oxygen atom at one end. This structure, rather than NON, was advanced by Langmuir,[8] who has given arguments in its favor. The formation of sodium trinitride from sodium amide and nitrous oxide, according to the reaction $NaNH_2 + ONN \longrightarrow NaNNN + H_2O$, further indicates that NNO is the true structure. Indeed, the pronounced oxygen-like chemical character of the gas strongly suggests that the oxygen atom is not in the inaccessible position between two nitrogen atoms.

The molecules of nitrous oxide would then be expected to have a permanent electric moment. The available data on the dielectric constant of the substance are, however, not accurate enough to permit verification of this prediction.

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[6] Lewis, This Journal, **46**, 1027 (1924).

[7] As in the nitrate ion, for example.

[8] Langmuir, This Journal, **41**, 1543 (1919).

TABLE I

THE MUTUAL POTENTIAL ENERGY OF ATOMIC KERNELS FOR CERTAIN CONFIGURATIONS

Substance	Configuration	Kernel potential energy[a]	Substance	Configuration	Kernel potential energy[a]
Carbon dioxide	OCO	57.40	Cyanic acid	HCNO	61.30
	COO	62.61		HONC	62.47
Nitrous oxide	NON	63.05	Nitriles	RCN	40.00
	NNO	60.88	Isocyanides	RNC	41.74
Cyanate ion	NCO^-	51.32	Hydrocyanic acid	HCN	23.05
Fulminate ion	CNO^-	53.92		HNC	23.48
......	CON^-	55.67	Cyanogen	NCCN	73.36
Isocyanates	RNCO	82.61		NCNC	75.80
Cyanates	ROCN	84.96		CNNC	78.59
Fulminates (nitrile oxides)	RCNO	83.48	Cyanogen fluoride	NCF	56.97
......	RONC	88.12		CNF	60.00
......	RCON	85.81			
......	RNOC	88.12			

[a] It is to be borne in mind that only the relative, and not the absolute, values of the kernel potential energy are of significance.

The Cyanate and Fulminate Ions.—Three isosteric univalent negative ions of carbon, nitrogen and oxygen are possible, NCO^-, CNO^- and CON^-. Evidently the ion with the smallest charge on the central kernel will have the least potential energy and, hence, free energy, and that with the largest charge the greatest. The three ions are accordingly here arranged in the order of predicted greatest stability.

This prediction is in complete harmony with the available data. The structure NCO^- has been assigned by chemical reasoning to the stable cyanate ion, and the structure CNO^- to the fulminate ion, the compounds of which are explosive. Compounds containing the third ion CON^- have not yet been prepared, due no doubt to their great instability.

It is, moreover, possible to predict roughly the energy differences to be expected. From thermochemical experiments the following equations may be written.

$$CO\ (g) + \tfrac{1}{2}O_2\ (g) = CO_2\ (g) + 67{,}500 \text{ cal.} \qquad (2)$$

$$N_2\ (g) + \tfrac{1}{2}O_2\ (g) = N_2O\ (g) - 17{,}740 \text{ cal.} \qquad (3)$$

Subtracting (3) from (2) we obtain

$$CO - N_2 = CO_2 - N_2O + 85{,}240 \text{ cal.} \qquad (4)$$

Since carbon monoxide and nitrogen are isosteres, we may calculate their kernel potential energies in the usual way, obtaining the values 20.86 and 21.73 energy units, respectively. Using these and the values for carbon dioxide and nitrous oxide given in the table we may write

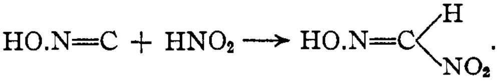
$$CO - N_2 = CO_2 - N_2O + 862{,}000 \text{ cal.} \qquad (5)$$

It is evident that the kernel energy calculation leads to a result approximately ten times too large. If we accept this factor of ten as applicable in other cases, then the energy and free-energy differences of the cyanate and fulminate ions should be about 86,000 cal. per mole.

Cyanates, Isocyanates, Fulminates, and Nitrile Oxides.—There are six possible isosteric compounds of an organic radical and a carbon-nitrogen-oxygen group, namely, RNCO, ROCN, RCNO, RONC, RCON and RNOC. The more stable compound of each pair differing only in the end attached to the radical, such as RNCO and ROCN, is, of course, the one in which the radical is near the kernel with the smaller charge. The relative stabilities of the six arrangements are roughly indicated by the calculations in the table; in these calculations the radical R is given the kernel charge of a carbon atom, and arbitrarily placed 1.15 Å. from the adjacent kernel.

Chemical data indicate that the isocyanates have the structure RNCO, and the normal cyanates ROCN; the relative stabilities are then the predicted ones, for the isocyanates are stable, while normal esters of cyanic acid have never been isolated, apparently because of the ease with which they condense to cyanurates.[9] It is worthy of mention that both the closely similar normal thiocyanates, RSCN, and isothiocyanates, RNCS, are known, and that the normal thiocyanates are the less stable, and are converted into the isothiocyanates on being heated.

Fulminic acid, through the work of Nef,[10] has been assigned the formula HONC, and its salts MONC, and these formulas have usually been accepted, although it is recognized that the evidence in their favor is not conclusive.[11] Our calculations show that compounds with these structures would have greater free energy than those with the structures HCNO and MCNO. Accordingly, we believe that the latter structures are the correct ones.

A consideration of the reactions on which the previous decision as to the structures of fulminates was based shows that they can be satisfactorily explained with the structures suggested by our potential calculations. One such reaction is the formation by fulminic acid of addition compounds with other acids; for example, with nitrous acid it forms methylnitrolic acid, the reaction being usually written  $HO.N{=}C + HNO_2 \longrightarrow HO.N{=}C\begin{matrix} / H \\ \backslash NO_2 \end{matrix}$.

We should, however, write this as a reaction first with the hydrogen ion, and then with the nitrite ion, eliminating the necessity of attaching the two ions to the same atom.[11a]

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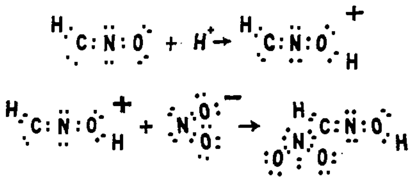
[9] Sidgwick, "Organic Chemistry of Nitrogen," Oxford, **1910**, p. 217.

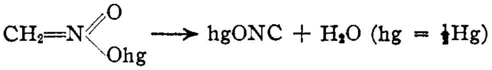
[10] Nef, *Ann.*, **280**, 303 (1894).

[11] Meyer and Jacobson, "Lehrbuch der Organischen Chemie," Veit, Leipzig, **1913**, vol. 1, p. 1303.

[11a] The argument does not depend on any particular choice of electronic structure for the nitrite ion. In the equations we have represented a structure which may not be the correct one.

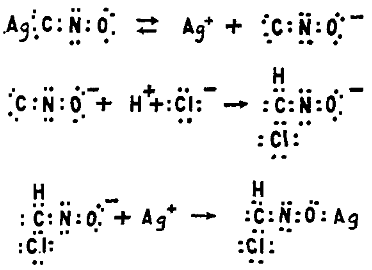


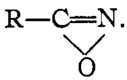
Other arguments are based on the assumption that rearrangements through ionization and recombination cannot occur with salts of fulminic acid. But such rearrangements might occur; for the salts of fulminic acid are ionized to a certain extent, the conductivity of an aqueous solution even of mercuric fulminate being slightly greater than that of water. In the formation of this salt from sodium nitromethane and mercuric chloride it has been assumed that mercury isonitromethane is first produced, which then by intramolecular oxidation forms the fulminate, according to the reaction



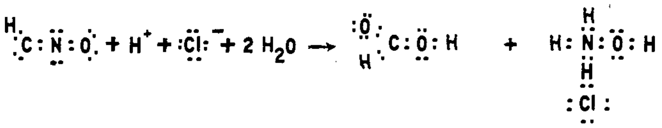
$$CH_2{=}N\genfrac{}{}{0pt}{}{{\nearrow}O}{{\searrow}Ohg} \longrightarrow hgONC + H_2O \quad (hg = \tfrac{1}{2}Hg)$$

Even if we accept this as the course of the reaction up to this point, it is still possible that the structure hgONC is not that of mercuric fulminate; for we believe that if the compound hgONC, which would be more highly ionized than the stable form, were produced, it would by ionization and recombination be converted into the stable form hgCNO. We similarly explain the formation of the silver salt of formyl chloride oxime, $AgONC\genfrac{}{}{0pt}{}{H}{Cl}$, from silver fulminate and hydrochloric acid by assuming that the silver fulminate molecule, AgCNO, is ionized to a certain extent, and that the hydrochloric acid adds to the fulminate ion to form the oxime ion, which then combines with the silver ion.

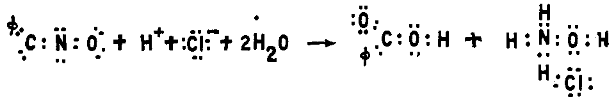


The structure usually assigned to the nitrile oxides is  It seems highly improbable to us that these three atoms should in this case form a ring, since in all other cases known three kernels with sixteen external electrons form a chain. Accordingly, we have been led to believe that the nitrile oxides are the esters of fulminic acid, and have the structure RCNO. This structure is, of course, the alternative structure previously proposed;[12] but since fulminic acid has been written HONC the nitrile-oxides have not before been designated fulminates.

The properties of the nitrile oxides indicate that they are indeed esters of fulminic acid. Oxalic ester nitrile oxide is formed from nitro-acetic ester by loss of water: $C_2H_5O.CO.CH_2NO_2 \longrightarrow C_2H_5O.CO.CNO + H_2O$; and other nitrile oxides are formed by similar reactions, all of which are compatible with the suggested structure. Most of the reactions of the nitrile oxides are just those which would be expected of the esters of fulminic acid, and these have up to this time required the explanation[13] that they are accompanied by a preliminary isomeric change into these esters. For example, fulminic acid[14] with hydrochloric acid hydrolyzes to hydroxylamine hydrochloride and formic acid, which partially decomposes to carbon monoxide and water.

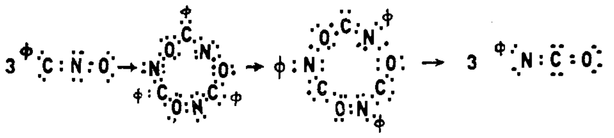


Completely analogous is the action of benzonitrile oxide and hydrochloric acid in forming benzoic acid and hydroxylamine hydrochloride.[15]



Evidently such reactions provide strong support for our view that the nitrile oxides are the esters of fulminic acid.

The remaining reactions of the nitrile oxides involve a change into the isocyanates; thus the triple polymer of benzonitrile oxide is converted into phenylisocyanate by heating with toluene. The migration postulated by us in explanation of this,



is at most no more violent than the migration and rearrangement previously accepted.

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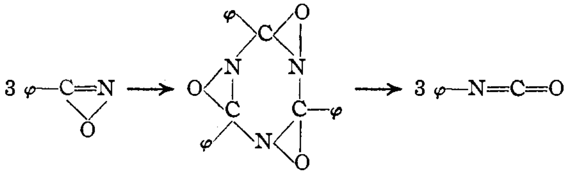
[12] Ley and Kissel, *Ber.*, **32**, 1365 (1899).

[13] Ref. 9, p. 230.

[14] Ref. 10, p. 1299.

[15] Wieland, *Ber.*, **40**, 1667 (1907).



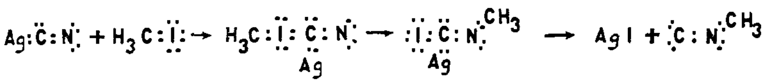
All of the chemical properties of the nitrile oxides are, then, in agreement with our suggestion that these substances are fulminates.

Compounds representing the three structures RONC, RCON and RNOC, which we have calculated to be the least stable, are unknown.

Cyanides, Nitriles and Isocyanides.—The structures of hydrocyanic acid and its salts and esters have been the subject of much discussion. Nef[16] has upheld the formula HNC for the acid, and MNC for its salts, while Wade[17] has argued for the formula HCN, retaining that of MNC. Of the esters the nitriles are generally conceded to have the structure RCN, and the isocyanides RNC.

The application of the principle suggested in this paper leads to the conclusion that the nitriles should be more stable than the isocyanides. This is in accordance with experiment; indeed, isocyanides reorganize to nitriles on being heated to about 250°.

The alkali cyanides are highly ionized,[18] so that no distinction can be made between cyanides and isocyanides. In the case of the cyanides of the heavy metals, however, the CN group is attached rigidly to the metal atom. The application of our principle leads to the inference that the carbon atom is adjacent to the metal atom; thus silver cyanide would have the formula AgCN or, if it is really the silver salt of the complex acid $HAg(CN)_2$, the formula $(Ag^+)(NCAgCN^-)$. These conceptions enable us to understand why an alkyl halide with an alkali cyanide forms the nitrile, and with a heavy metal cyanide, the isocyanide. In the first case reaction occurs between the cyanide ion and the halide; for example, $H_3CI + CN^- \longrightarrow H_3CCN + I^-$; and, inasmuch as the cyanide ion is free to attach itself by either end, the more stable compound, the nitrile, is formed. In the second case, however, the reaction is between molecules, and may occur in steps in the following way: the metal cyanide attaches itself to the alkyl halide, forming a double compound, from which the metal and halide atoms split off. The reaction between methyl iodide and silver cyanide would then be represented by the following steps.



$$Ag:\ddot{C}:\dot{N}: + H_3C:\ddot{I}: \rightarrow H_3C:\ddot{I}:\underset{Ag}{\ddot{C}}:\dot{N}: \rightarrow :\ddot{I}:\underset{Ag}{\ddot{C}}:\dot{N}:^{CH_3} \rightarrow AgI + :C:\dot{N}:^{CH_3}$$

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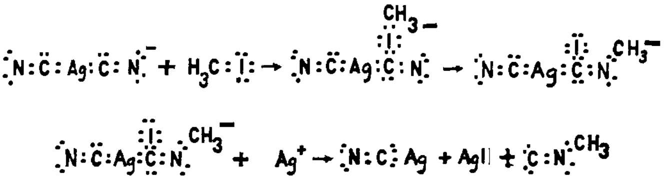
[16] Nef, *Ann.*, **270**, 328 (1892).

[17] Wade, *J. Chem. Soc.*, 81, 1596 (1902).

[18] Complete ionization of potassium cyanide in the solid state is indicated by its crystal structure. Bozorth, THIS JOURNAL, **44**, 317 (1922).

or, in case the formula $(Ag^+)(NCAgCN^-)$ is used,



It is evident that the cyanide group is prevented from presenting its carbon end to the alkyl carbon, and that the isocyanide is accordingly formed. The increasing formation of nitrile as the alkyl group becomes larger, and with aryl halides, is to be attributed to an increasing ease of rearrangement to the more stable form.

From potential considerations we also conclude that a proton would attach itself more easily to the carbon than the nitrogen end of the cyanide ion, and accordingly we assign to hydrogen cyanide the structure HCN.

Cyanogen.—There are three possible collinear isosteric formulas for cyanogen; NCCN, CNCN and CNNC. It is shown to have the first of these by its chemical properties, which lead to the conclusion that the two carbon atoms are linked together.[19] This structure is the one which would also be predicted to be the most stable on the basis of our principle.

Cyanogen Halides.—The compound cyanogen fluoride would be expected from our calculations to have the structure NCF, rather than the less stable one CNF. This compound is not known, but the chloride, bromide and iodide are known; the predicted formulas are, however, not those generally accepted. The evidence[20] upon which these substances are assigned formulas such as CNCl is the similarity in the reactions of the halogen in this substance and in others in which it is attached to nitrogen, such as the halogen-substituted amines and amides; but such evidence is not trustworthy, and other reactions point strongly to the formula NCCl. For example, cyanogen chloride with ammonia forms cyanamide: $NCCl + NH_3 \longrightarrow NCNH_2 + HCl$. Moreover, by the Friedel and Crafts reaction cyanogen chloride forms benzonitrile: $NCCl + C_6H_6 \longrightarrow HCl + C_6H_5CN$; this reaction cannot be explained with the use of the formula CNCl, but definitely requires NCCl; so in this case also chemical evidence verifies the predicted structures.

Summary

In order to calculate the relative stabilities of certain compounds, the principle is suggested that the differences in the free energies of isosteric compounds arise principally in the terms involving the mutual potential

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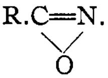
[19] Ref. 9, p. 196.

[20] Chattaway and Wadmore, *J. Chem. Soc.*, 81, 191 (1902).

energy of the positive kernels. This principle is applied in a number of cases, with the following results.

It is shown, in agreement with chemical evidence, that the structures of carbon dioxide and nitrous oxide are OCO and NNO rather than COO and NON.

The greater stability of the cyanate ion, NCO^-, as compared with the fulminate ion, CNO^-, is explained.

It is shown that theoretically the isocyanates, RNCO, should be more stable than the cyanates, ROCN. Furthermore, the postulate leads to the conclusion that fulminic acid and the inorganic fulminates have the formulas HCNO and MCNO, rather than the accepted ones HONC and MONC. Moreover, the nitrile oxides are considered to be esters of fulminic acid, and to have the formula RCNO, rather than  $R.C{=}N.$ All of the suggested structures are shown to be compatible with the experimental evidence.

The principle explains the observation that the nitriles, RCN, are more stable than the isocyanides, RNC. The structures HCN for hydrocyanic acid, $(M^+)(CN^-)$ for the cyanides of the alkali metals, and MCN for those of the heavy metals are shown to be in agreement with their chemical properties.

The accepted formula NCCN for cyanogen is shown to be the predicted one. The accepted formula CNX for the cyanogen halides is, however, rejected in favor of the predicted formula NCX.

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[CONTRIBUTION FROM THE LABORATORY OF PHYSICAL CHEMISTRY OF THE GEORGE WASHINGTON UNIVERSITY]

CRYSTAL ANGLES, MEASURED UNDER THE MICROSCOPE

BY EDWIN A. HILL

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For several years the writer in the courses in Chemical Microscopy given by him at the George Washington University, in which the "Elementary Chemical Microscopy" of Professor Chamot is used as a textbook, has given to his students a simple method whereby all of the solid angles in any crystal, however microscopically small, when found upon the object slide can be readily measured, with considerable accuracy, without removing the crystal from the slide upon which it has been formed, or without the use of any special piece of apparatus, a measurement which, so far as I am aware, could not hitherto be made, in cases where the crystal is too small, or too firmly attached, to be removed from the object slide and mounted in a goniometer.