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AN ATTEMPT TO ARRANGE SILICATES IN THE ORDER OF REACTION ENERGIES AT RELATIVELY LOW TEMPERATURES*

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INTRODUCTION

There have been attempts from time to time to arrange minerals, especially the silicates, in some order which would correspond to their sequence of formation, or their order of stability, or perhaps their order of energy of reaction under a given set of conditions. Bowen's Reaction Series, familiar to all of us, is an example. Many of us who are here today were inspired by the address of our past-president, Professor M. J. Buerger (1948), two years ago on the stability relations of minerals and by his recent contribution (1948) on silica-bridging in structures. Another very creditable attempt was made by H. W. Fairbairn (1943) in his paper on packing indexes of ions. His figures, while unquestionably important, did not lead to any mineral series of either formation or stability though probably were included in both and formed a part of each of them.

It should be possible, however, to arrange the minerals in some kind of an order which would tell us something of their reaction energies under a given set of conditions, let us say for illustration, weathering or at any rate, conditions under which water plays an important role at not a very high temperature, and proton transfer becomes important.

BONDING FORCES

We know very little about the magnitude of the forces which hold ions and atoms together in minerals. Some forces are covalent or electron-sharing as in diamond and most sulfides; others are ionic as in salts and oxides. For this reason these two classes cannot be directly compared. The silicates comprise a third class in which the forces are partly

* Address of the retiring president of the Mineralogical Society of America given at El Paso, Texas, on November 11, 1949, under the title "An Attempt to Arrange Silicates in the Order of Free Energies."

ionic and partly covalent. The cations with the exception of those with very high valence like Si and Ti, and possibly Al, are tied to oxygen largely ionically. An oxide like SiO_2 on the other hand is at least 50% covalent and for this reason the atomic percentage of Si in a silicate is of paramount importance in the behavior of the mineral.

It appears then that we may have to pay heed to two proportionality factors which for our purposes may be treated separately:

- (1) The factor dealing with ionic forces.
- (2) The factor dealing with covalent SiO_2 forces.

H								
2.1								
Li	Be	B	C	N	O	F		
1.0	1.5	2.0	2.5	3.0	3.5	4.0		
Na	Mg	Al	Si	P	S	Cl		
0.9	1.2	1.5	1.8	2.1	2.5	3.0		
K	Ca	Sc	Ti	Ge	As	Se	Br	
0.8	1.0	1.3	1.6	1.8	2.0	2.4	2.8	
Rb	Sr	Y	Zr	Sn	Sb	Te	I	
0.8	1.0	1.3	1.6	1.7	1.8	2.1	2.5	
Cs	Ba							
0.7	0.9							
			Fe'''	Fe'' assumed by the author				
			1.5	1.2				

FIG. 1. Electronegativities of the Elements. (After Linus Pauling)

Pauling has provided us with a means of estimating the order of magnitude of the ionic forces through his so-called Electronegativity Series which is shown in Fig. 1. He has assigned certain values to the most common ions which give their relationship to each other, that is, those farthest apart are the most electronegative with respect to each other, and have the greatest tendencies to form stable compounds with each other. In the silicates practically all cations are tied to oxygen which has a value of 3.5. We may, therefore, take some large power of the difference between 3.5 and the value of the respective cation as proportional to the bond strength. In our case, it is simpler and more expedient, as will be explained below, to take the numbers in Fig. 1 directly for our calculations, which will be illustrated by the following examples of quartz, considered here a silicate, and of orthoclase:

SiO_2 , $\text{Si} = 1.8$. The oxygen ions are not counted as it is assumed that

all cations are tied to oxygen. As there is only one cation in SiO_2 the electronegativity is 1.8 per cation.

KAlSi_3O_8 , $K=0.8$, $Al=1.5$, $3Si=3\times 1.8$. Adding $0.8+1.5+(3\times 1.8)=7.7$, which is the total for the formula. Dividing 7.7 by 5, the number of cations, gives 1.54 which is the electronegativity of orthoclase or microcline. If the differences between oxygen and the respective figures given in Fig. 1 had been taken, the result would have been 1.70 and 1.96, respectively, which are the complementary numbers to 1.80 and 1.54, each pair adding up to 3.50, the value for oxygen. For our purposes it is more convenient to use the numerical values 1.80 and 1.54 even though 1.54 actually stands for a greater electronegativity than 1.80, a numerical inversion to which one soon becomes accustomed.

Pauling's Table, Fig. 1, is not complete for our purposes as the element iron is left out, for the obvious reason that Fe has two possible valences. For the present purpose we may assume ferrous iron to have the value 1.2 the same as Mg, and ferric iron 1.5, like Al, though we may be certain that considerable errors are introduced by this assumption, even when as here oxidation is neglected as a factor. The value for hydrogen as given by Pauling is 2.1 which seems high to introduce into minerals containing hydroxyls or hydrogen bonds. The calculated values obtained with it appear to be of the correct order of magnitude, however.

If the silicates are arranged according to their calculated electronegativities (Table 1) it is observed that in general, the high temperature minerals of Ca and Mg are at one end and the hydrated ones are at the other. Closer examination, however, indicates plainly that these electronegativities alone do not give us the desired answer. Some important factor is still missing. It is obvious that electronegativity will not distinguish between two or more different manners of bonding of the same kinds of ions or atoms as, for example, SiO_4 framework and chain structures which are largely covalent anyway and for this reason should receive additional attention.

Such different bonding should introduce a proportionality factor in our calculations which we shall call the bridging factor, a name used first by M. J. Buerger (1948) who applied it to the manner and number of bridging between SiO_4 tetrahedra. For example, in quartz, cristobalite and tridymite all tetrahedra are directly tied to other tetrahedra of SiO_4 resulting in the highest number of bridges possible, a perfect framework, with a bridging factor, arbitrarily called 1.00. All other structures evidently will have smaller factors.

It immediately occurs to one that other atoms or ions besides Si must contribute to the bridging factor. AlO_4 tetrahedra commonly substitute

TABLE 1. ENERGY RELATIONS OF SILICATES BASED ON ELECTRONEGATIVITIES AND COORDINATIONS

Silicate	Cation Formula	Electro- Negativity	Bridging Factor	Energy Index
Quartz	Si	1.80	1.000	1.80
Montmorillonite	$H_4(Al, Mg)_{4-5}Al_2Si_6 \cdot 2H_2O$	1.80	0.948	1.77
Kaolinite	$H_8Al_4Si_4$	1.87	0.938	1.75
Pyrophyllite	$H_4Al_3Si_8$	1.80	0.962	1.73
Heulandite	$Ca_2Al_4Si_{14} \cdot 12H_2O$	1.90	0.878	1.67
Stilbite	$NaCa_2Al_3Si_{15} \cdot 15H_2O$	1.89	0.874	1.65
Lawsonite	$H_4CaAl_2Si_2$	1.78	0.928	1.65
Hydromicas	$H_4(H, K)Al_6Si_6$	1.73	0.950	1.64
Vermiculite	$H_2(Mg, Fe)_3(Si, Al)_4 \cdot 2H_2O$	1.77	0.922	1.63
Danburite	CaB_2Si_2	1.72	0.950	1.63
Euclase	$H_2Be_2Al_2Si_2$	1.73	0.938	1.62
Zircon	ZrSi	1.70	0.950	1.62
Bertrandite	$H_2Be_4Si_2$	1.73	0.926	1.60
Beryl	$Be_3Al_2Si_6$	1.66	0.964	1.60
Staurolite	$H_2FeAl_4Si_2$	1.69	0.940	1.59
Datolite	HCaBSi	1.72	0.926	1.59
Laumontite	$(Na_2, Ca)Al_2Si_4 \cdot 4H_2O$	1.80	0.876	1.58
Serpentine	$H_8Mg_6Si_4$	1.73	0.912	1.58
Chlorites	$H_8(Mg, Fe)_5Al_2Si_3$	1.73	0.912	1.58
Chloritoid	$H_4Fe_2Al_4Si_2$	1.70	0.928	1.58
Dumortierite	$HBAl_3Si_3$	1.65	0.958	1.58
Apophyllite	$KCa_4Si_3F \cdot 8H_2O$	1.82	0.862	1.57
Talc	$H_4Mg_6Si_8$	1.67	0.934	1.56
Petalite	$LiAlSi_4$	1.62	0.964	1.56
Muscovite	$H_4K_2Al_6Si_6$	1.65	0.942	1.55
Topaz	Al_2SiF_2	1.60	0.966	1.55
Tourmaline	$H_4NaFe_3B_3Al_6Si_6$	1.65	0.942	1.55
Sillimanite	Al_2Si	1.60	0.966	1.55
Kyanite	Al_2Si	1.60	0.966	1.55
Andalusite	Al_2Si	1.60	0.966	1.55
Margarite	$H_4Ca_2Al_8Si_4$	1.64	0.938	1.54
Analcime	$NaAlSi_2 \cdot H_2O$	1.70	0.898	1.53
Pumpellyite	$HCa_2Al_3Si_3 \cdot H_2O$	1.65	0.915	1.51
Cordierite	$Mg_2Al_4Si_5$	1.58	0.958	1.51
Mullite	Al_6Si_2	1.57	0.962	1.51
Natrolite	$Na_4Al_4Si_6 \cdot 4H_2O$	1.69	0.888	1.50
Axinite	$HCa_2BFeAl_2Si_4$	1.60	0.938	1.50
Albite	$NaAlSi_3$	1.56	0.956	1.49
Anthophyllite	$H_2Mg_7Si_3$	1.59	0.934	1.49
Glaucophanite	$H_2Na_2Mg_3Al_2Si_8$	1.58	0.940	1.49
Prehnite	$H_2Ca_2Al_2Si_3$	1.60	0.934	1.49
Phenakite	Be_2Si	1.60	0.934	1.49

TABLE 1—(continued)

Silicate	Cation Formula	Electro-Negativity	Bridging Factor	Energy Index
Pollucite	$H_2(Ca, Na)_2Al_2Si_5$	1.61	0.920	1.48
Orthoclase-Microcline	$KAlSi_3$	1.54	0.954	1.47
Zoisite	$HCa_2Al_3Si_3$	1.56	0.938	1.46
Epidote	$HCa_2(Al, Fe)_3Si_3$	1.56	0.934	1.46
Lithium mica	$H_4K_2Li_3Al_5Si_6$	1.57	0.924	1.45
Tremolite	$H_2Ca_2Mg_5Si_8$	1.56	0.932	1.45
Hornblende	tremolite-pargasite	1.56	0.928	1.45
Spodumene	$LiAlSi_2$	1.53	0.946	1.45
Anorthite	$CaAl_2Si_2$	1.52	0.950	1.44
Norbergite	H_2Mg_5Si	1.60	0.900	1.44
Marialite	$Na_4Al_3Si_9Cl$	1.52	0.938	1.43
Phlogopite-Biotite	$H_2KMg_3AlSi_3$	1.55	0.916	1.42
Jadeite	$NaAlSi_2$	1.50	0.946	1.42
Pyrope	$Mg_3Al_2Si_3$	1.50	0.938	1.41
Pargasite	$H_4Na_2Ca_4Mg_3Al_4Si_{13}$	1.51	0.924	1.40
Enstatite	Mg_2Si_2	1.50	0.934	1.40
Aegerine	$NaFeSi_2$	1.50	0.934	1.40
Almandite	$Fe_3Al_2Si_3$	1.50	0.930	1.40
Meionite	$Ca_4Al_6Si_6 \cdot CO_3$	1.49	0.935	1.39
Chondrodite	$H_2Mg_5Si_2$	1.54	0.904	1.39
Leucite	$KAlSi_2$	1.47	0.942	1.38
Pectolite	$H_2Na_2Ca_4Si_6$	1.49	0.920	1.37
Vesuvianite	$H_4Ca_{10}Mg_2Al_4Si_9$	1.48	0.918	1.36
Sapphire	Mg_2Al_3Si	1.46	0.934	1.36
Humite	$H_2Mg_7Si_3$	1.50	0.906	1.36
Titanite	$CaTiSi$	1.47	0.928	1.36
Diopside	$CaMgSi_2$	1.45	0.930	1.35
Augite	$(Ca, Mg, Fe)_2(Si, Al)_2$	1.45	0.930	1.35
Grossularite	$Ca_3Al_2Si_3$	1.43	0.934	1.34
Clinohumite	$H_2Mg_9Si_4$	1.48	0.906	1.34
Nepheline	$NaAlSi$	1.40	0.928	1.30
Wollastonite	$CaSi$	1.40	0.926	1.30
Andradite	$Ca_3Fe_2Si_3$	1.42	0.918	1.30
Forsterite	Mg_2Si	1.40	0.912	1.28
Fayalite	Fe_2Si	1.40	0.912	1.28
Kaliophilite	$KAlSi$	1.37	0.924	1.27
Gehlenite	Ca_2Al_2Si	1.36	0.920	1.25
Akermanite	Ca_2MgSi_2	1.36	0.914	1.24
Sodalite-Hauynite	$Na_4Al_3Si_3Cl$	1.35	0.904	1.22
Monticellite	$Ca.MgSi$	1.33	0.906	1.20
Merwinite	Ca_3MgSi_2	1.30	0.902	1.17
Cuspidine	$Ca_4Si_2F_2$	1.27	0.900	1.14
Larnite	Ca_2Si	1.27	0.900	1.14

for SiO_4 . What are their values? Each atom or ion ought to contribute to the bridging factor in proportion to the strength of its bonds. This strength is expressed by the charge on the ion divided by the number of coordinate bonds reaching this ion which corresponds to Pauling's "valence bond."

The coordinate numbers or bonds of the ions are well known and need no further discussion except for Al. Al has sometimes 4, in other structures 6, coordinates. In the present study we cannot distinguish between them. On the basis of packing, hardness, melting points and other less tangible properties it may be assumed that the two are of very similar valence bond strength. If there is any choice, I would assign a greater one to Al with 6 coordinates under the lower temperature conditions. A valence bond, therefore, of $3/4$ is assigned to all Al and ferric ions.*

An example may now be calculated. The sum of the valence bonds in KAlSi_3O_8 is $1/10 + 3/4 + 3 = 77/20$. This divided by the number of cations, 5, gives 0.77 a value which will be called the coordination coefficient.

If the total electronegativity of KAlSi_3O_8 were multiplied by this coefficient the wrong value would be obtained because this coefficient is introduced only to modify a portion, perhaps 20%, of the bonding forces contained in the electronegativity values. 80% of the electronegativity factor is used unchanged. Why are 20% and 80% respectively chosen? Because by trial and error it was found that this proportioning gives us the best empirical agreement. For KAlSi_3O_8 the bridging factor would be, therefore, $0.77 \times 0.20 + 0.80 = .954$, as compared with unity, which was selected for quartz. This factor when multiplied by the electronegativity 1.54 results in the value 1.47, which I shall call the energy index of KAlSi_3O_8 , that is of orthoclase-microcline.

About 90 silicates have been arranged according to their energy indexes in Table 1 and in subsequent tables containing special groups of minerals. In order to make the formulas as simple as possible, oxygen has been omitted except from water molecules. Isomorphous mixtures have been avoided as far as possible. The cations are given in the order of valence. Column 1 contains the electronegativity factor, column 2 the bridging factor. The coordination coefficients have been omitted. They may, however, be calculated from the information supplied in Table 1. The general spread between the indexes is not large but one may recall that they would have to be raised to some high, though unknown, power to give us comparable energy relations. Also the order is reversed in the tables, the highest energies having the lowest numerical values.

* Valence bonds of $1/8$ have been assigned to Ca and Na, $1/10$ to K and $1/2$ to H.

DISCUSSION OF ARRANGEMENT

An inspection of the order of the silicates shows:

(1) The presence of hydrogen, mostly as (OH), in the formulas puts these silicates largely in the upper half of the table.

(2) A water molecule is treated like two hydrogens in the electronegativity factor but its bonds do not figure in the coordination coefficient except in the sum of the ions or atoms used as divisor because the water molecule can hardly be said to be tied strongly to the other atoms, but it does occupy much space and in this manner weakens the structure materially. Without this treatment of H_2O , particularly in zeolites, these minerals would be quite out of line with respect to the other silicates. Even so, the zeolites behave in a manner (Stevens, 1948) which suggests several kinds of H_2O molecules (Gruner, E. 1948) in their structures. A somewhat similar role seems to be played by such radicals as Cl in marialite, meionite, and sodalite, for they, too, are largely responsible for the loose packing and weakening of structures. For this reason, they appear only in the divisors of the sums of the valence bonds.

(3) Quartz or its polymorphs, tridymite and cristobalite, occupy a unique position at the head of the silicates, though tridymite and cristobalite have relatively high energies and should not be placed with quartz, for which reason they were omitted entirely. Unfortunately, our scheme does not allow the proper placing of polymorphs like kyanite, sillimanite and andalusite, unless we include such properties as Fairbairn's "Packing Index," which would permit the separation of such trimorphs, placing kyanite, for example, somewhere before and andalusite after sillimanite in our table.

(4) Beryllium silicates are higher in the table than expected on first thought, while lithium silicates, like spodumene and lithium mica, are considerably lower than expected. If one recalls that spodumene decomposes very readily and the Li ion as a whole has a very great tendency to become hydrated this difficulty disappears. Calcium boron silicates, datolite in particular, are much higher in the table than would be expected. The high electronegativity of 2.0 for B is partly responsible for these positions. Boron compounds, admittedly, are not understood very well at present.

(5) Silicates which we associate with "dry" magmas have higher energies than those of "wet" magmas. For this reason, they alter easily under the assumed conditions to the minerals at the beginning of the table, apart from the decomposition to oxides and carbonates which are not discussed here.

Many silicates have been omitted because they present such complicated isomorphous mixtures that their energy indexes would be ques-

tionable, though they might be between those of the end members. There is no question, however, that any stress caused by mixing will be expressed by higher free energy.

GEOLOGIC APPLICATION

If the table is examined as a whole its usefulness is somewhat obscured by its length; but if it is broken down into smaller units either according to similarity in composition, or according to their paragenesis very interesting relations appear. Time does not permit detailed discussions, but a few selections are tabulated.

Hydrolytic silicates

In processes in which the temperature is not too high and H₂O is one of the reactants, the silicates of Table 2 are formed, commonly by decomposition of other silicates which were formed at higher temperatures and would possess higher reaction energies. Other minerals containing the hydroxyl could have been added to this table, for example, epidote, but we rarely think of them as connected with hydrolysis. It is evident that these hydrolytic silicates are among the lowest in energy.

TABLE 2. HYDROLYTIC PRODUCTS

	Energy Index		Energy Index
Quartz	1.80	Vermiculite group	1.63
Montmorillonite group	1.77	Serpentine group	1.58
Kaolinite group	1.75	Chlorites	1.58
Pyrophyllite	1.73	Talc	1.56
Hydromicas	1.64+	Muscovite	1.55
		Zeolites	1.50-1.67

Aluminum silicates connected with hydrothermal activity

The hydrolytic minerals just discussed are also included in the larger group of hydrothermal Al silicates. There are also some in this group which should be only among the pegmatite minerals, perhaps though occasionally found in veins as, for example, beryl. Table 3 is highly instructive because it shows the changes which are likely to occur with a drop in temperature in hydrothermal alterations and also in veins.

Biotite, for example, would go to vermiculite, and chlorite to hydromicas, and ultimately to the montmorillonite group. Other series would be: Feldspars to muscovite and/or to the montmorillonite group, or to kaolinite; feldspars to zeolites, to montmorillonite, or kaolinite as, for example, in Yellowstone Park. In all these series quartz would be

TABLE 3. ALUMINUM SILICATES CONNECTED WITH HYDROTHERMAL ACTIVITY

	Energy Index		Energy Index
Quartz	1.80	Tourmaline	1.55
Montmorillonite group	1.77	Muscovite	1.55
Kaolinite group	1.75	Topaz	1.55
Pyrophyllite	1.73	Margarite	1.54
Heulandite	1.67	Analcime	1.53
Stilbite	1.65	Pumpellyite	1.51
Hydromicas	1.64+	Natrolite	1.50
Vermiculite group	1.63	Albite	1.49
Bertrandite	1.60	Prehnite	1.49
Beryl	1.60	Orthoclase	1.47
Chlorites	1.58	Zoisite	1.46
Laumontite	1.58	Epidote	1.46
Apophyllite	1.57	Phlogopite-Biotite	1.42

one of the end products as the mineral of lowest energy having high bond stability under these conditions. Opal may be mentioned as a likely neighbor of quartz. The positions of the zeolites are somewhat doubtful.

Granitic Pegmatite Silicates

These silicates (Table 4) being high in Al, B and Be are high in bond strength and relatively low in energy. The apparent inconsistency of the Li silicates in this respect has been pointed out above. The high coordination of Be, which is 4, and its high electronegativity tend to put euclase, bertrandite and beryl a little higher in the list than expected. Alka-

TABLE 4. GRANITIC PEGMATITE SILICATES

	Energy Index		Energy Index
Quartz	1.80	Sillimanite	1.55
Zircon	1.62	Andalusite	1.55
Euclase	1.62	Cordierite	1.51
Bertrandite	1.60	Albite	1.49
Beryl	1.60	Phenakite	1.49
Dumortierite	1.58	Pollucite	1.48
Petalite	1.56	Orthoclase	1.47
Muscovite	1.55	Spodumene	1.45
Topaz	1.55	Lithium mica	1.45
Tourmaline	1.55	Phlogopite-biotite	1.42

lies, like Na, which are found in the composition of beryl naturally raise the reaction energy resulting in lower stability. This statement applies to the presence of any ill-fitting ions which may have entered the crystal lattice at higher temperatures when the tolerance for isomorphous mixing was much greater than at lower temperatures.

As a whole the energy indexes have a remarkably narrow range which reflects the similarity of conditions under which these silicates form. The positions of biotite and tourmaline being far apart in the table seem to be reason for the observation that the two are mutually exclusive, for tourmaline crowds out biotite. The high index of zircon indicates that besides having high stability this mineral may very well crystallize at a late stage at relatively low temperatures from wet magmas.

TABLE 5. CONTACT METAMORPHIC SILICATES

	Energy Index		Energy Index
Danburite	1.63	Meionite	1.39
Datolite	1.59	Chondrodite	1.39
Andalusite	1.55	Pectolite	1.37
Muscovite	1.55	Humite	1.36
Cordierite	1.51	Sphene	1.36
Axinite	1.50	Vesuvianite	1.36
Albite	1.49	Diopside	1.35
Orthoclase	1.47	Augite	1.35
Zoisite	1.46	Sapphirine	1.35
Epidote	1.46	Grossularite	1.35
Tremolite	1.45	Clinohumite	1.34
Hornblende	1.45	Wollastonite	1.30
Norbergite	1.44	Andradite	1.30
Marialite	1.43	Forsterite	1.28
Phlogopite-biotite	1.42	Gehlenite	1.25
Jadeite	1.42	Cuspidine	1.14
Almandite	1.40		

Contact metamorphic silicates

The contact metamorphic silicates of Table 5 have a great range as would be expected for we include here minerals which form directly in contact with the magma, as well as many which make use of or need mineralizers to form. The presence of these volatiles for example causes wollastonite to form at as low as 400° C. to 450° C. From its position in the table this would not have been expected. It may be stressed once more that the arrangement does not represent the relative temperatures of formation, but rather the ease with which the silicates may alter to

others under conditions as stated. The positions of silicates like marialite and cuspidine are not too certain for no particular allowance was made in the calculations for such an anion as F, and the value of a Cl radical is not certain as mentioned above.

DYNAMOMETAMORPHIC SILICATES

This is a very instructive group of silicates. With the exception of those very high in Al the structures are either framework, layers or double chains as may be seen in Table 6. This means high bridging factors except in serpentine, biotite, and hornblende where Mg and ferrous Fe lower this factor considerably. It is noteworthy that the only two open structures, albite and orthoclase-microcline have very high bridging factors as one should expect for alkali silicates in order to be stable under differential pressures.

TABLE 6. DYNAMOMETAMORPHIC SILICATES

	Energy Index		Energy Index
Quartz	1.80	Margarite	1.54
Pyrophyllite	1.73	Cordierite	1.51
Lawsonite	1.65	Glaucophane	1.49
Staurolite	1.59	Anthophyllite	1.49
Chlorites	1.58	Albite	1.49
Chloritoid	1.58	Orthoclase (microcline)	1.47
Serpentine	1.58	Zoisite	1.46
Talc	1.56	Epidote	1.46
Muscovite	1.55	Tremolite-hornblende	1.45
Tourmaline	1.55	Phlogopite-biotite	1.42
Sillimanite	1.55	Pyrope	1.41
Kyanite	1.55	Almandite	1.40

CONCLUSION

Similar tables may be prepared, for example, of acidic, alkali, and basic igneous rock silicates, and of Bowen's Reaction Series, all of which seem to agree with the order of energy indexes, except for the inverse positions of orthoclase and albite. If the proposition as outlined proves to be correct as a first approximation its ground-work is much simpler than could ever have been expected. Many criticisms from students of thermodynamics are anticipated.

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