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THE ELECTROSTATIC INTERLAYER FORCES OF LAYER STRUCTURE MINERALS

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Abstract—Using a simple ionic model, the energy necessary to expand a layer structure by a certain distance can be calculated. This has been done for a series of 15 structures including hydroxides, 2:1 and 1:1 structures of various types. Plots of energy versus separation distance show three major groups which have common bonding properties. For large separations, the group with the strongest interlayer bonds contains the brittle micas, the hydroxides, and the 1:1 structures. Intermediate bonding structures are the normal micas and the weakest bonds occur in the zero layer charge 2:1 structures. The relative energies needed for a given separation are not constant so that for small separations the zero layer charge structures such as talc and pyrophyllite are more strongly bonded than the normal micas. These groupings correlate very well with the expandability of the structures by water and other substances. It is proposed that this approach to the study of the layer structures will provide a simple theory explaining the expansion properties of layer silicates.

Key Words—Bonding, Charge, Energy, Expansion, Interlayer.

INTRODUCTION

The 2:1 layer silicates have a common crystal structure based on two sheets of tetrahedrally coordinated cations separated by a single sheet of octahedrally coordinated cations. Substitutions by differently charged cations result in a net negative charge on the layer which is balanced by placing additional cations between the layers. The stability of such structures is due to the electrostatic attraction between the negative layer and the positive interlayer cations. By substituting different cations in the three types of site, one obtains a great variety of minerals which exhibit a large diversity of physical and chemical properties. In an attempt to classify these minerals so as to, in a sense, explain the origin of their physical and chemical properties, the layer charge has proved to be the most useful criterion. Thus in 1966 the AIPEA agreed on a classification based on the total charge which contained five groups (Mackenzie, 1965; Brindley, 1966; Pedro, 1967). This appears to be a very natural system because the minerals when listed from high charge to low charge show a distinct and more or less gradual change in properties. The brittle micas have the largest charge (-2 per half unit cell) and their lack of flexibility compared to normal micas (-1) is easily attributed to the stronger electrostatic bonds between the layers resulting from the larger charge. A further reduction in layer charge produces the vermiculites (-0.9 to -0.6) which are normally expanded and contain water molecules between the layers. Presumably the larger charges and stronger interlayer bonding prevents the two types of mica from behaving in a similar fashion. Still smaller charges (-0.6 to -0.2) produce the smectites which also expand in the presence of water and many organic liquids but in a manner quite different from the vermiculites. The behavior of the smectites is related to the type of interlayer cation but in general one can say that they expand to a much greater degree than the vermiculites. The latter generally contain only one or two layers of water whereas the smectites with either Na or Li as the interlayer cations can expand to an unlimited degree. The final group in the classification contains the zero layer charge minerals talc and pyrophyllite.

The classification has three major problems which are pertinent to the present discussion. Firstly, the use of the total layer charge implies a continuity between all the groups. This point has been discussed extensively by Mering and Pedro (1969) who proposed that the separate groups, and in particular the vermiculites and smectites, are fundamentally distinct. Thus a low charge vermiculite and a high charge smectite may have the same layer charge but different properties of expansion. However, Suquet et al. (1977) have synthesized saponites which have layer charges varying between 0.33 and 1.0. They conclude that the "layer charge density cannot be considered as a criterion . . ." to distinguish smectites from vermiculites. Secondly, according to classification the smaller the layer charge, the easier it is to exchange the interlayer cation or to hydrate the mineral in question. There are apparently two conflicting influences involved here; namely the attraction between the external molecules and the interlayer ions as well as the interlayer bonding energy. The former provides the driving force for the expansion while the latter determines how easily the layers can be separated. The two are related so that the weaker the interlayer bonding (presumably allowing easy expansion), the fewer the number of interlayer cations (small driving force) and thus one finds little or no expansion. This is true if the primary interaction between the external molecules and the interlayer region is of the di-

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pole-cation type. For molecules such as water, acetone and 3-pentanone, Brindley and Ertem (1971) showed that reducing the layer charge reduced the expansion of montmorillonite. However, for other molecules such as ethanol, ethylene glycol, and morpholine, they found no relationship between layer charge and expansion. They suggested that for these latter molecules the cation-dipole attraction was augmented by hydrogen bonding to the layer surfaces. If the latter suggestion is correct, then talc and pyrophyllite having a very weak interlayer bond might expand with strong hydrogen bond forming molecules. Apparently this has not been observed (Hofman et al., 1956) so one is left with the conclusion that either some interlayer cations are necessary for expansion or perhaps there is something peculiar about the interlayer bonding of these two minerals which makes it very difficult to expand them. Thirdly, muscovite and biotite have the same layer charge but the latter is much more susceptible to exchange of the interlayer cation and subsequent expansion.

Recent work of several types has shed some light on these difficulties. Mering and Pedro (1969) suggested, and more recent experimental work has supported their idea, that the smectites have disordered ionic substitutions while the vermiculites and illites have a much higher degree of order (Besson et al., 1974). The evidence for this is the observation of diffuse streaks in electron diffraction patterns of vermiculites, illites, and muscovites while such streaks are absent in smectites. Thus there cannot be a continuity between the groups even though the layer charges may be similar or identical. More recently, Suquet et al. (1977) have observed such streaks in high-charge synthetic Na-saponites and Kodama (1975) has found them in talc. The distinction between smectites and vermiculites based on order/disorder of substituting cations is not clear.

Mering and Pedro (1969) did not address directly the nonexpandability of talc and pyrophyllite but simply treated all the nonexpanding structures as a group distinct from the intermediate charge expanding minerals. Giese (1975a) calculated the strength of the interlayer electrostatic attraction and found 6.5 kcal/mole and 4.1 kcal/mole for pyrophyllite and talc respectively. These are much smaller than for muscovite (32.2 kcal/mole) and phlogopite (22.4 kcal/mole for the hydroxy form and 27.6 kcal/mole for the fluoride form) (Giese, 1975b).

The third difficulty has been explained at least qualitatively (Giese, 1975b) by showing that the interlayer bonding of hydroxy-phlogopite is weaker than muscovite and moreover that substitution of F^- for OH^- increases the interlayer bonding energy as noted above. This is in agreement with the well-known influence of the OH orientation on the alteration of biotite (Bassett, 1960; Gilkes et al., 1972). The differences between muscovite, oxidized biotite and phlogopite/biotite are explained readily in terms of the electrostatic repulsion between the hydroxyl hydrogen and the interlayer cation.

Undoubtedly the layer charge plays a major role in the expanding properties of 2:1 structures but the precise relationship between charge and expansion is not clear. This contribution is an attempt to explore more fully the implications of the layer charge and how it may be related to the interlayer bonding and the expandability of layer structures.

BACKGROUND

The existence of a layer charge and interlayer cations of opposite charge implies the concept of interlayer electrostatic attraction or bonding. The electrostatic interactions between complex structures such as the layer silicates cannot be reduced to a single value, the net or global charge. This is illustrated clearly by the existence of a net electrostatic attraction between the neutral layers of talc and pyrophyllite. The present study was begun with the idea that perhaps the interlayer bonding strengths would be more useful than the layer charge in understanding the physical and chemical properties of the layer silicates (Brindley, 1970).

The calculations of the interlayer bonding have been discussed in detail elsewhere (Giese, 1974). Briefly, one calculates the potential energy for the crystal structure in question and then for a similar structure with identical interatomic distances and angles within the layers but with the layers separated by a large distance, usually 9 Å. The difference in energy is a measure of the strength of the interlayer bonding. For structures with a neutral charge on adjacent surfaces, such as the micas when the interlayer cations are distributed equally on both surfaces, the energy for the expanded mineral approaches rapidly an asymptotic value for separations beyond about 7 Å so that 9 Å is a good compromise between accuracy and length of computing time. For other structures, such as kaolinite with hydrogen ions on one surface and oxygen on the opposite surface, much larger distances are needed to reach a constant energy because in effect the two surfaces carry positive and negative charges which interact at greater distances whereas the micas have zero charge surfaces. In practice, the computations become too time-consuming to carry out completely for these minerals and the slope of the energy versus separation curve has been used as a measure of the relative energy (Giese and Datta, 1973).

As pointed out earlier, if we ignore the possible necessity of having interlayer cations for expansion, the interlayer bonding energies for talc, pyrophyllite, and the micas are not in agreement with the observed nonexpandability of the zero charge minerals. Therefore, the calculations presented here examined the change in potential energy as a function of increase in the layer separation for distances varying between 0.01 and 9.0

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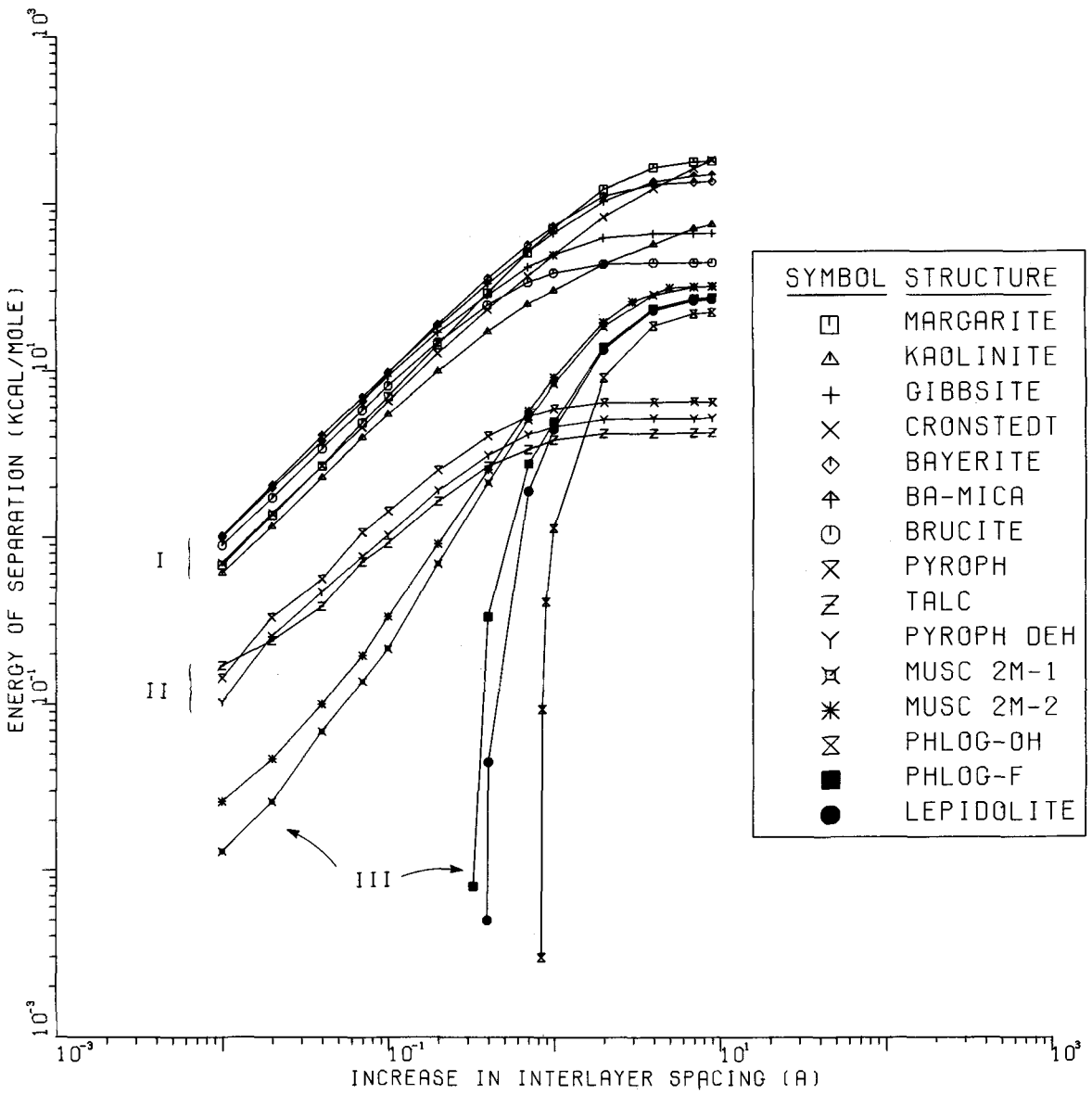


Fig. 1. The curves show the relationship between the expansion of layer structures and the energy required to separate the rigid layers from each other.

Å in the hope that the variation might reveal features not seen just by looking at the energy change caused by an expansion of 9 Å.

If interlayer bonding governs the expanding/nonexpanding properties of the 2:1 structures, then the bonding energy concept should be applicable to a much larger group of layer structures of which the 2:1 minerals form a subset. In order to examine this and at the same time to put the 2:1 minerals in a more general context, various layer structures such as the 1:1 minerals and hydroxides were included. For each type of structure, both di- and trioctahedral varieties were examined. Some important minerals such as the chlorites and vermiculites were not included because at present the hydrogen positions in these structures are not known and without this information, the calculations cannot be performed. Another very important group of minerals, the smectites, have not been studied because the disorder among the tetrahedral and/or octahedral sites cannot be described in the present form of the calculations. It should be emphasized that all the minerals included here are in the nonexpanded form. The 15 minerals are listed in Table 1 along with the reference giving the details of the crystal structure. A second reference, if given, reports the hydrogen positions for the minerals.

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Table 1. The structures used for the calculations shown in Figure 1 along with the ideal composition for each and the references describing the crystal structure.

STRUCTURE	IDEAL COMPOSITION	REFERENCE
pyrophyllite 1Tc	$Al_2Si_4O_{10}(OH)_2$	Wardle and Brindley (1972); Giese (1973b)
pyrophyllite dehydroxylate 1Tc	$Al_2Si_4O_{11}$	Wardle and Brindley (1972)
talc 1Tc	$Mg_3Si_4O_{10}(OH)_2$	Raynor and Brown (1973); Giese (unpublished)
muscovite $2M_1$	$KAl_2(Si_3Al)O_{10}(OH)_2$	Güven (1971); Giese (unpublished)
muscovite $2M_2$	$KAl_2(Si_3Al)O_{10}(OH)_2$	Zhoukhlistov et al. (1973); Giese (unpublished)
F-phlogopite 1M	$KMg_3(Si_3Al)O_{10}F_2$	Joswig (1972)
OH-phlogopite 1M	$KMg_3(Si_3Al)O_{10}(OH)_2$	Joswig (1972)
lepidolite $2M_2$	$KAl_{1.25}Li_{1.75}(Si_{3.5}Al_{0.5})O_{10}F_2$	Sartori et al. (1973)
margarite $2M_1$	$CaAl_2(Si_2Al_2)O_{10}(OH)_2$	Guggenheim and Bailey (1975)
Ba-mica 1M	$BaLiMg_2(Si_3Al)O_{10}F_2$	McCauley and Newnham (1973)
kaolinite 1T	$Al_2Si_2O_5(OH)_4$	Zvyagin (1967); Giese and Datta (1973)
cronstedtite	$Fe_2SiFeO_5(OH)_4$	Steadman and Nuttall (1963); Giese (unpublished)
bayerite	$Al(OH)_3$	Saalfeld and Wedde (1974)
gibbsite	$Al(OH)_3$	Saalfeld and Wedde (1975)
brucite	$Mg(OH)_2$	Zigan and Rothbauer (1967)

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RESULTS

Figure 1 is a log-log plot of the change in potential energy versus increase (from the known crystal structure) in the interlayer spacing for all 15 minerals. What strikes one immediately about the curves is their simplicity and general lack of variety. This is most surprising considering the great diversity of structure types included in the plot.

Looking at the large separation energies, we see three major groups labelled I to III (Figure 1) and for small separations, the group III structures can be further subdivided. The groups, subgroups, and their characteristics are as follows. The energies between parentheses are the approximate energy ranges for the group computed for the 9 Å separation.

Strong bonding (50 to 200 kcal/mole)

This includes 1) di- and trioctahedral 1:1 structures with long hydrogen bonds between layers, 2) hydroxides, both di- and trioctahedral with long hydrogen bonds, and 3) brittle micas both di- and trioctahedral.

Intermediate bonding (20 to 35 kcal/mole)

These are the normal di- and trioctahedral micas. The two subgroups differ greatly and are: A) Dioctahedral micas which have a concave curve; B) Trioctahedral micas whose energy difference becomes very small as the separation decreases. In fact for separations below approximately 0.5 Å there is a small electrostatic repulsion which must mean that for these small separations the van der Waals energy dominates.

Weak bonding (4 to 10 kcal/mole)

These are the neutral layer structures. All have a large plateau with essentially the same energy for separations larger than 2 Å. The energy decrease as the separation becomes small is less rapid than for the group III micas and below approximately 0.4 Å the interlayer bonding for the group II minerals is much greater than for the group III minerals.

One can divided the layer structures into two broad types: those with long hydrogen bonds between the layers and those without. It is clear from Figure 1 that all the hydrogen bonded minerals in the study belong to group I. By analogy with the studies of the role played by long hydrogen bonds in the kaolinite group minerals (Giese, 1973), one can conclude that the stability of all hydrogen bonded layer structures is due almost solely to these bonds. Therefore one would expect a similar behavior for any well-ordered layer structure containing either interlayer water or interlayer hydroxyls such as the chlorites and hydrated vermiculites.

Non-hydrogen bonded layer structures are much more complex in their behavior and this complexity is the result of the interplay of two independent variables each of which has an influence on the interlayer bonding. The first of these is of course the magnitude of the layer charge. If this charge is sufficiently large (−2 for the 2:1 structures), it dominates the interlayer bonding and one has a group I mineral, or is sufficiently small (0 for talc and pyrophyllite) one has a group II mineral. For charges of −1, and perhaps smaller, the di- versus trioctahedral character of the mineral dominates. The reasons for this are not obvious but may be related to the fact that in the trioctahedral case the positive octahedral cation charge is distributed over three sites instead of two thereby "diluting" the influence these sites have on the ions of the adjacent layers.

DISCUSSION

The implications of this study can best be discussed in terms of a simple mechanical model of the expansion of a layer structure and formation of an intercalated or hydrated compound. Starting with the unexpanded structure, the steps in the process are 1) the opening, very likely at the edge of the crystal, of one or more layers; 2) the insertion of molecules of the intercalating or hydrating compound; 3) the subsequent diffusion of the expanding molecules throughout the crystal. In order to expand a layer structure, according to this model, the system must meet two criteria: 1) the initial expansion must be possible under the conditions of the experiment; 2) the final product must be thermodynamically more stable than the starting materials.

Since this study has examined the energies necessary to separate the layers of unexpanded structures, the results are directly applicable to the first step in the intercalation process. It seems clear moreover that what is important in the process is not the energy required to separate the layers of a given structure by a large distance but, on the contrary, to separate them by a very small distance. In other words, if the expansion process cannot be initiated, it cannot occur. Therefore the important differences in expandability of layer structures should be explained by the relative energies for separating the layers by a small distance. Figure 1 indicates that for an arbitrary separation of 0.01 Å the structures are ordered, beginning with the most strongly bonded, in the sequence: group I, group II, dioctahedral micas, and trioctahedral micas. This sequence is in much better agreement with the known expansion properties than are the layer charges. The nonexpansion of talc and pyrophyllite is not surprising since they are relatively more difficult to expand by small distances than are trioctahedral or dioctahedral micas. The fact that in general the trioctahedral 2:1 structures are easier to expand than the dioctahedral varieties is easily understood and may also explain why the vermiculites are trioctahedral. Finally, the origin of the generally observed difficulty of expanding the group I structures is clear.

The energies necessary for small expansions indicate general trends in expansion properties but are only part of the process of expansion. One should not conclude that a particular structure in, for example, group I can-

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not be expanded, only that it is more difficult than other structures of lower energy. The sole exception to the nonexpandability of the structures in group I is kaolinite. Various salts can be intercalated in kaolinite by a mechanical grinding process (Wada, 1961) or treatment with a concentrated solution of an appropriate salt (Weiss et al., 1963; Smith et al., 1966). The calculations reported here are for perfect, infinite crystals and thus represent ideal behavior. Real structures, which deviate greatly from perfection, such as kaolinite may exhibit properties different from those consistent with their bonding energies.

The vermiculites when dehydrated are probably similar to a trioctahedral mica with a reduced layer charge. One might expect them to behave in a similar manner to the group I trioctahedral structures but generally with the curve shifted to smaller energies. The smectites differ from the vermiculites basically in three ways; 1) they have in general a lower layer charge; 2) the ionic substituents are disordered; 3) the site of the layer charge may be either octahedral, tetrahedral or both, while vermiculites have their charges in the tetrahedral sites. If we exclude the question of disorder, the smectites fall somewhere between the group II and III structures in terms of layer charge, but these groups are so completely different that it is impossible at this time to predict with any certainty what sort of curve one would find for a dehydrated smectite. It is almost certain to be different from a dehydrated vermiculite because the latter will not rehydrate but the former will.

CONCLUSIONS

The energy required to expand layer structures is related to the layer charge but in an indirect manner. A plot of the energy as a function of the amount of the interlayer expansion is characteristic of the particular layer structure. From this standpoint many different structures can be placed in a few distinctive groups. This grouping is more successful in explaining the expansion properties than the layer charge because it indicates 1) that talc and pyrophyllite should be difficult to expand and this difficulty is not related to the absence of interlayer cations; 2) that high layer charge structures as well as those having interlayer hydroxyl bonds should be difficult to expand; 3) that the trioctahedral 2:1 structures with moderate layer charge should be easier to expand than equivalent dioctahedral structures.

It is suggested that similar plots for vermiculites and smectites including the order/disorder of the octahedral and tetrahedral cations may explain their very different expansion properties. This subject is being actively pursued.

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Резюме- Используя простую ионную модель,можно вычислить энергию,необходимую для расширения слоистой структуры на определенное расстояние. Это было проделано для серии из 15 структур,включая гидроокиси,структуры 2:1 и 1:1 различных типов. Графики зависимости энергии от расстояния разделения указывают на 3 главных группы,которые имеют характерные связующие свойства. При большом разделении группа с сильнейшими межслойными связями включает хрупкие слюды,гидроокиси и структуры 1:1. Структурами с промежуточными связями являются структуры нормальных слюд, и слабейшими связями обладают структуры 2:1 со слоями,имеющими нулевые заряды. Относительные величины энергии,необходимые для данного разделения,не являются постоянными.Так при небольших разделениях структуры со слоями,имеющими нулевые заряды,такие как тальк и пирофиллит,связаны сильнее,чем нормальные слюды. Это группирование очень хорошо коррелируется со способностью структур к расширению водой и другими жидкостями. Предполагается использовать этот метод для изучения слоистых структур,что обеспечит простую теорию для объяснения свойств расширения слоистых силикатов.

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