

MIXED-LAYER MINERAL ASSOCIATIONS IN MUSCOVITE-CELADONITE AND MUSCOVITE- CHLORITE JOINS

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EXTENDED ABSTRACT

Using the accumulated data on illite as a basis, it is proposed that this mineral or mineral group can be treated as a phase in a petrologic system. By doing this, the problems of origin and metamorphic behavior can be determined. This involves making some generalizations about illite that hold closely enough to the real situation to allow translation of data from natural minerals to a synthetic system and vice versa.

Illite, or material that is generally called illite, is rather heterogeneous as it occurs in rocks and sediments; however, most illitic material found in *rocks* has the following characteristics: (1) a dioctahedral micallike structure that is disordered in the c sin β crystallographic direction, i.e. a 1Md polymorph; (2) a composition similar to that of muscovite but containing more Mg, Fe, and Si with less K and Al; (3) varying amounts of interlayering with a basically 14Å material (the amount of 14Å material is usually small, < 15 per cent); (4) a common association in rocks with chlorite and quartz with or without feldspar, montmorillonoid, or kaolinite; (5) an origin (at least partial) in the depositional or diagenetic environment, i.e. low temperature and pressure conditions. If it can be assumed that these generalizations bear a reasonable relation to the true situation, a simplified model of the illite mineral or mineral group can be made in terms of a petrologic system.

The system chosen is a modification of the $\frac{1}{2} K_2O-R^{2+}O-R_2^{3+}O_3$ co-ordinates, considering SiO_2 and H_2O in excess of any requirement during a chemical reaction. The two assumptions are quite applicable to most sediments and thus could be considered valid for sedimentary rocks. In such a system, the variables of composition can be chosen to represent minerals commonly occurring in sediments: KAl = feldspar, R^{3+} = kaolinite or pyrophyllite, R^{2+} = talc or a chlorite (Fig. 1).

Natural illite compositions fall between muscovite (KR_3^{3+}) and chlorite ($R^{2+}R^{3+}$ to R^{2+}) in the system. Their bulk composition is close to that of muscovite but varies as f_{O_2} changes the $Fe^{3+} : Fe^{2+}$ ratio. This change may only occur as new phases are produced.

The micas muscovite (KR_3^{3+}) and biotite ($KR^{3+}R_2^{2+}$) form a boundary in the system above which potassium is present in sufficient quantity to form a mica, if it is stable, from all the R^{3+} and R^{2+} components present. Illites fall below this line and are therefore considered potassium deficient, i.e. below the content necessary to form a single-phase mica product from the bulk composition.

If illite is not in a "mica" system (above the line), it is in a join between muscovite plus another component because natural illite does nevertheless greatly resemble the mica muscovite in structure and bulk composition. The most likely join would be muscovite-chlorite. This is not too unreasonable because the two minerals are commonly

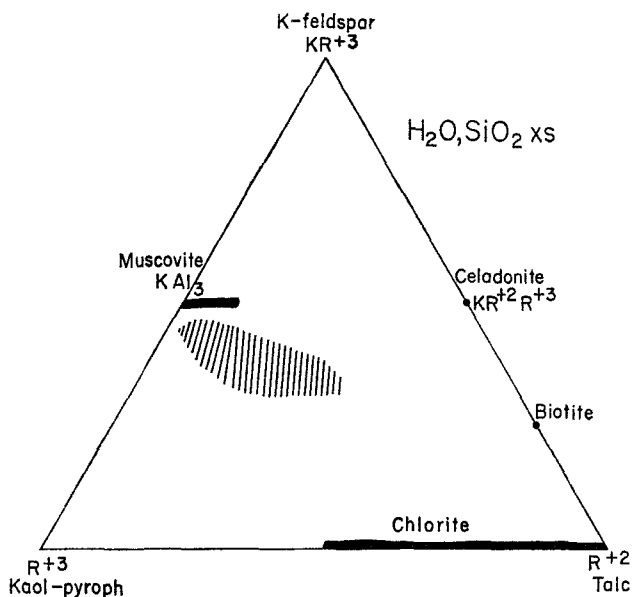


FIG. 1. Compositions of illite (shaded area) as plotted in the KR^{3+} - R^{3+} - R^{2+} system.

associated in sedimentary rocks. The potassium "deficiency" of the bulk composition and the common presence of interlayered 14\AA phases add to the plausibility of such a consideration. The very regular and rather uniform occurrence and habit of illite in indurated sedimentary rocks suggests that an equilibrium situation has been reached for some rather general P-T-X conditions.

The nonbinary systems muscovite-celadonite were studied in order to determine the amount of solid solution possible in a muscovite that has a true mica composition, i.e. $K \pm .1$ atom per unit cell. Eight such joins were studied, and the results showed very limited Mg and Fe substitutions at conditions of low-grade metamorphism. It was concluded that a mica, with a dioctahedral, potassic 10\AA structure, would have a composition near muscovite at conditions experienced by most sedimentary rocks. No continuous solid solution exists between muscovite and celadonite-glaucanite.

There were, however, interesting results in all these joins. The muscovite- $(Mg_{1.0}Fe_{0.5}^{3+}Al_{0.5})$ -celadonite join (Fig. 2) is used as an example. It must be remembered that the bulk composition of each point contains enough potassium to create a theoretical single-phase mica product (K-saturated). The products at low temperatures were muscovite, muscovite-montmorillonoid mixed-layer, and montmorillonoid plus quartz and K-feldspar in all runs. At higher temperatures and higher pressures the products were muscovite, biotite, sanidine, and quartz. Runs lasted for 30 days.

Here we have products that closely approximate phases found in natural samples—mixed-layering of two clay minerals. In order to demonstrate that such a product is not a metastable phase formed upon quenching of the run, the products formed at higher temperature—muscovite + biotite—were subjected to conditions that had produced mixed-layer minerals. The products of this run were mixed-layered. However, the amount of mixed layering did not reproduce that of the original materials synthesized under the same conditions. As a consequence it must be stated that the boundary as drawn between

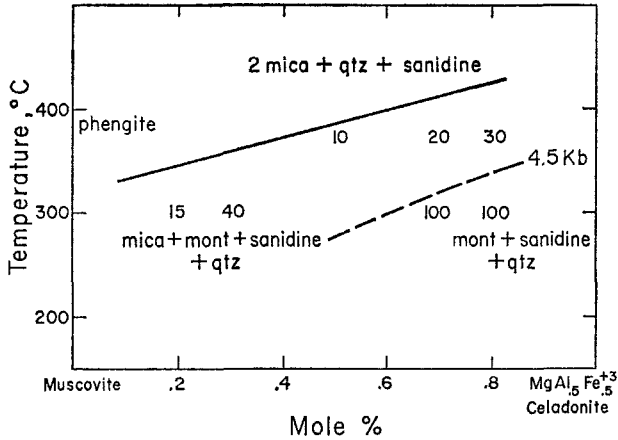


FIG. 2. Phase relations in the muscovite–celadonite composition join at 2 kb pressure. Numbers indicate per cent montmorillonoid in clay–mineral phase. Dashed line shows mica–montmorillonoid boundary of synthesis products at 4.5 kb pressure.

mica and mica-mixed-layer products cannot be fixed exactly but the reaction of mica + mixed-layer \rightarrow two micas was reversed, indicating equilibrium between the two assemblages at some temperature above the "reversal run" (2 kb, 400°–440°C). Thus, muscovite mixed-layer micas are stable under some real P–T conditions. Similar mineral associations found in sedimentary rocks should be considered as stable phases.

Of petrologic interest are the P–T– x relations of the mixed-layer products. It was found that an increase of temperature or pressure decreased the amount of mixed-layering in the product. Also, as could be expected, when the bulk composition approached muscovite, less mixed layering was found, because less material was available to form a montmorillonoid. There are conditions of low temperature where even the muscovite will form an expandable mineral that could be called a montmorillonoid. Therefore the boundary between mica and mica–montmorillonoid phases will run completely across the system for some P–T conditions.

Relating this to sedimentary rocks, the materials that represent the highest grade of metamorphism or diagenesis (Paleozoic shales and metamorphosed shale) would be expected to contain the least montmorillonoid or mixed layer material. This has been previously observed by several authors. Also a montmorillonoid phase was formed in the presence of excess K^+ ; the run product K-feldspar indicates this, as would the bulk composition of the system. Thus it is difficult to maintain that all montmorillonoids will become micas given enough K^+ . This opens a possibility previously closed by some authors.

A summary of the muscovite–celadonite systems shows that under conditions of high K content montmorillonoid and mica–montmorillonoids will form. These phases form a two-mica assemblage at higher temperatures. The mixed-layer phases were shown to have a stability range and to represent a stable assemblage for their compositions. Comparison of the assemblages in the mica–celadonite system with the K-bentonites of sedimentary rocks seems warranted. The mineral assemblages and bulk compositions are rather similar: a high potash content and a mixed-layer type mineralogy—muscovite and montmorillonite.

Considering the initial argument, it seems reasonable to treat illite as a product of

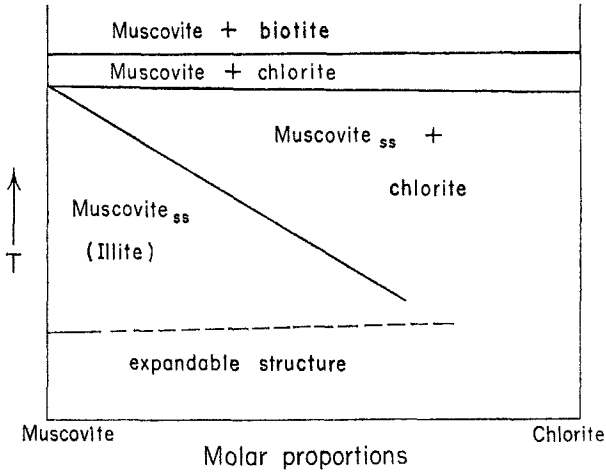


FIG. 3. Proposed phase relations in muscovite-chlorite composition join. Experiments on natural and synthetic materials indicate that the muscovite solid solution-chlorite relations may be useful to explain the nature of illite.

mineral associations in a muscovite-chlorite join. In this system (Fig. 3) illite is considered to be a solid solution of chlorite in muscovite. Synthetic products at about 20 per cent chlorite substantiate the interpretation of the muscovite end of the diagram. mixed-layer 14-10Å and 14Å chlorite was formed. The mixed layering was small (~15 per cent or less) as is generally the case for illite.

Several series of experiments were run using natural clay minerals as starting materials. Illite and an illite-chlorite assemblage from a sedimentary rock were subjected to a range of temperatures and 2 kb pressure. The results substantiate the interpretation of the diagram at higher temperatures. It would appear that, given a bulk composition at low temperatures, one could have illite or illite-chlorite mixtures as stable phases with quartz and water in excess. At high temperatures illite has less of the chlorite component in solid solution. Eventually muscovite and chlorite are stable. Reactions involving other phases occur as chlorite disappears and biotite comes in as a stable phase. The sequence illite → chlorite + muscovite → biotite + muscovite was reported by Winkler (1964) in hydrothermal experiments using natural illite.

The main conclusion drawn is that illite is a solid solution mineral-muscovite + chlorite. This solid solution is dependent upon bulk composition and P-T conditions of the environment. Illite bulk compositions represent those of the potash-poor area of the $KAl-R^{3+}-R^{2+}$ diagram. A thorough investigation of the system in which illite occurs will give the particular stability range and amounts of solid solution that will define the mineral illite.

REFERENCE

- WINKLER, H. G. F. (1964) Das T-P Feld der Diagenese und niedrigtemperierten Metamorphose aufgrund von Mineralreaktionen, *Beiträge zur Mineralogie und Petrographie* 10, 70-93.