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## ACCELERATED WEATHERING OF FELDSPARS

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### INTRODUCTION

This investigation was undertaken with the purpose of developing a method for the alteration of feldspars, which, except for the acceleration by higher temperatures, would more nearly simulate natural conditions than had previous methods. In order to carry this out, it was decided to supply fresh liquid to the sample, and to carry away the soluble reaction products in one continuous process. It was proposed at first to attempt the decomposition of certain types of feldspars with carbonic acid; and, in addition, to identify the reaction products and determine the temperature limits in order to throw more light on the origin of clays.

### PREVIOUS WORK

Considerable work has been carried on in the past by geologists on the synthesis of clays and the decomposition of feldspars. In general two methods have been followed. First, the co-precipitated gels of alumina and silica are placed in a reaction chamber and heated for some time, usually with an acid. In this way finely crystallized kaolinite has been formed and detected by *x*-ray methods. The work of Noll,<sup>1</sup> van Nieuwenburg, Pieters,<sup>2</sup> Schwarz,<sup>3</sup> Ewell and Insley<sup>4</sup> have been along this line.

The second method consists in a hydrothermal treatment of feldspar in a reaction chamber with a rather high concentration of acid. Collins,<sup>7</sup> and later Badger and Ally,<sup>8</sup> have reported the production of kaolinite in this way by the use of hydrofluoric acid. Schwarz and Trageser<sup>5,6</sup> have reported the formation of both kaolinite and pyrophyllite by the hydrothermal treatment of feldspar with hydrochloric acid. Collins<sup>7</sup> attempted the decomposition of feldspar with carbonic acid at a low temperature but obtained no positive results.

In none of these methods has mention been made of a continual supply of fresh solute or the removal of the soluble reaction products as occurs in nature.

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## PLAN OF THE INVESTIGATION

In this investigation the minerals orthoclase, anorthite, and albite were used as the original materials. These minerals after fine grinding were subjected to the action of water and carbon dioxide, the reaction being accelerated by a moderately high temperature. Arrangements were made for applying to the sample a supply of fresh carbonic acid solution and for removing the soluble reaction products in one continuous process. The altered feldspars were then studied by means of the petrographic microscope and the x-ray powder method to determine the resultant minerals present.

## ORIGINAL MATERIALS

A potash feldspar from West Paris, Maine, containing a large proportion of orthoclase was selected for the first test. This material had the following analysis:

SiO <sub>2</sub> .....	65.5
Al <sub>2</sub> O <sub>3</sub> .....	19.6
Fe <sub>2</sub> O <sub>3</sub> .....	0.8
CaO.....	0.4
MgO.....	Trace
K <sub>2</sub> O.....	11.2
Na <sub>2</sub> O.....	3.3

A soda feldspar, consisting principally of albite, was also used. The analysis showed:

SiO.....	58.53
Al <sub>2</sub> O <sub>3</sub> .....	19.59
Fe <sub>2</sub> O <sub>3</sub> .....	0.06
CaO.....	0.71
MgO.....	—
K <sub>2</sub> O.....	1.29
Na <sub>2</sub> O.....	9.52

The lime feldspar consisted of perfect crystals of anorthite from Miakejima, Japan. Because of the excellence and purity of the crystals no chemical analysis was deemed necessary.

The feldspar crystals were crushed and then ground with water in a porcelain pebble mill for three weeks, which reduced the fragments to a maximum size not greater than one micron. This very fine grinding was carried out in order to expose a maximum surface, and thus accelerate the rate of reaction.

## APPARATUS

The reaction chamber developed for this work is shown in Fig. 1. The chamber itself is made from a piece of heavy seamless steel tubing

with heads held in place by a ring of six cap crews. The chamber was made tight by means of a copper gasket about 1/16 in. thick which covered the entire ends of the reaction chamber. Into these gaskets were soldered the various copper connecting tubes. The sample, weighing about ten grams, was placed in a very thin, porous cup made of alumina,

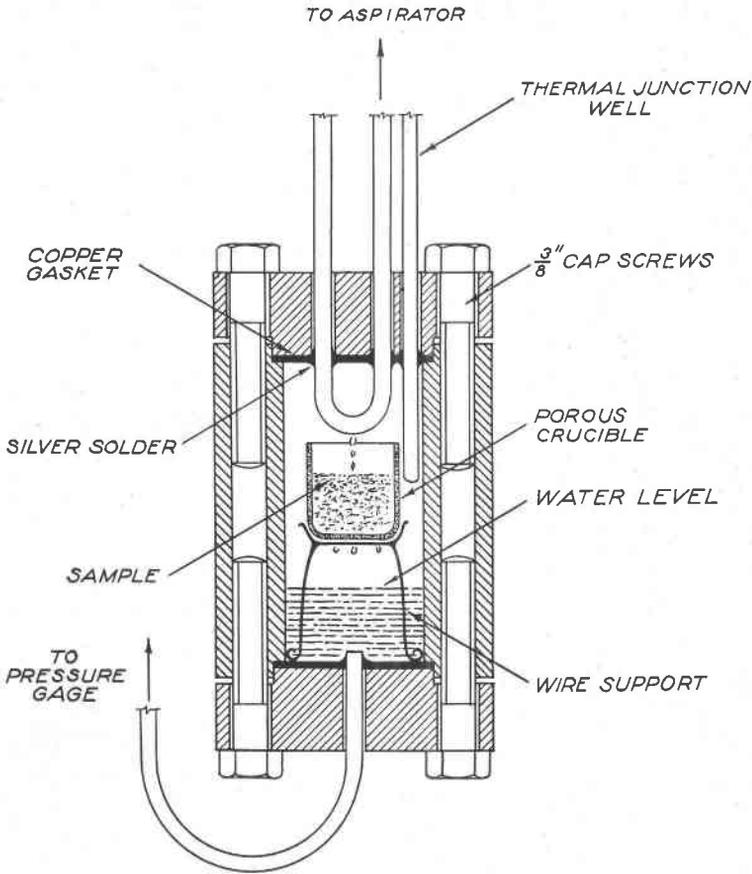


FIG. 1. Reaction chamber.

which is supported on a wire framework to elevate it just above the water level. A copper tube leads from the bottom of the chamber to a pressure gauge which indicates at all times the pressure in the chamber. The temperature in the chamber is measured by a copper-constantine thermo-couple inserted in a well near the center of the reaction chamber. The cold junction of the couple is kept in ice.

The important feature of this reaction chamber is the "U" shaped condenser tube which is silver-soldered into the top gasket. Air is drawn through this tube to condense the vapors on the outer surface from where it drips down onto the sample. The rate of condensation can be varied by the amount of air drawn through the tube. It is generally adjusted so that the rate of forming condensate is about equal to the amount of liquid which can flow through the sample and porous cup.

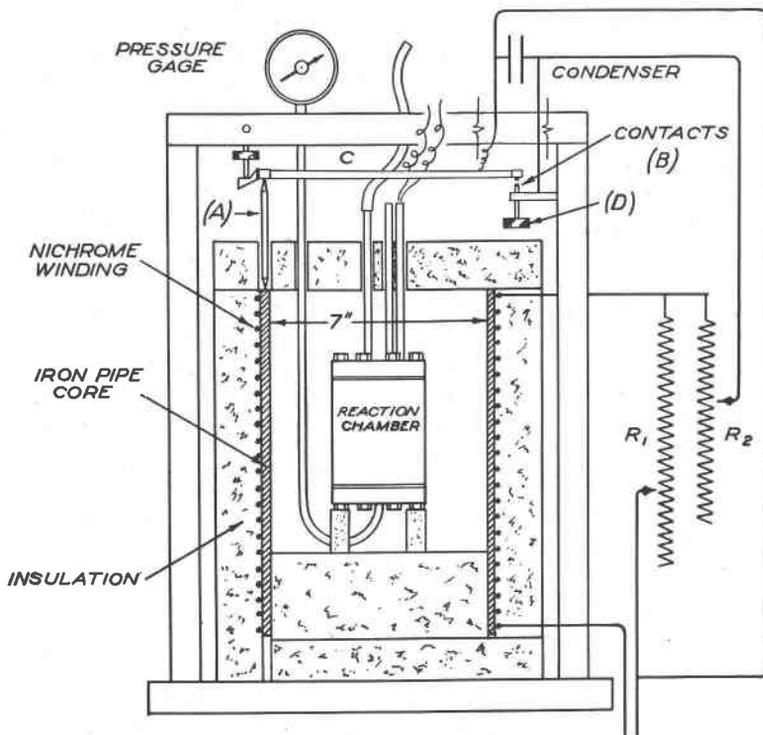


FIG. 2. Heating furnace.

In some of the first runs condensation was so rapid that the cup continuously overflowed.

One of the chief advantages of this reaction chamber is the ease with which it can be kept tight. After the first trial runs, there was little trouble with leakage, for in nearly every case the pressure when cold, at the end of a long run, was exactly the same as when the run was started. This reaction chamber before being used was tested under a hydrostatic head of 5000 pounds per square inch.

The furnace for heating the reaction chamber is shown in Fig. 2 and consists of a simple cylindrical chamber with a nichrome winding. The furnace is well insulated and requires only a few amperes to maintain the required temperature. The novel feature of this furnace is the temperature control which is shown diagrammatically in Fig. 2. An expansion of the iron furnace core acting through the strut *A* opens the contact *B* with about a thirty to one multiplication by a lever *C*. The opening of these contacts cuts out the portion of the current flowing through the parallel resistance  $R_2$  which decreases the total current flowing through the windings by about 10%. After the furnace cools slightly the contacts are closed and the current is again increased. Rheostat  $R_1$  is used for controlling the inflow for the particular temperature desired. A close adjustment of the temperature is obtained by the screw *D*. This very simple mechanism requires no relays and is entirely reliable over long periods of time. Under good conditions the temperature in the reaction chamber is maintained within plus or minus 1°C. for the duration of the run, although in these particular tests variations in room temperature occasionally allowed the temperature to go beyond these limits.

#### METHOD OF MAKING THE RUN

A run is made by placing the sample in the porous cup and adding about 75 c.c. of distilled water to the bottom of the chamber. Lumps of dry ice are then placed in the chamber, surrounding and on top of the porous cup, until it is nearly filled; then the top of the chamber is bolted down as quickly as possible. The bolts are not tightened up completely until all the carbon dioxide has vaporized and the pressure has gradually settled to the correct value. Then by tightening down the bolts it is very easy to arrive at exactly the desired gas pressure. This method eliminates the usual system of pipes, valves, and connections that would be necessary if carbon dioxide was supplied from a tank.

The reaction chamber is then placed in the furnace, covered, and gradually brought up to the desired temperature. The condenser tube is connected to an aspirator which draws air through it constantly. After the temperature is once established, the only attention required during the run is a daily check of the pressure and temperature.

#### RESULTS

##### *Orthoclase*

A number of runs were made with orthoclase under various conditions of temperature and carbon dioxide pressure, as shown in the following table:

TABLE 1

Run No.	Temp. C.	CO <sub>2</sub> Pressure Cold lbs./sq. in.	Total Pressure Hot lbs./sq. in.	Total Time at Temp.	Conversion
2	225°	15	390	11 days	Very slight alteration
4	320	200	2250	10	Slight alteration
6	280	265	1800	10	Slight alteration
6a	280	265	1800	21	Considerable change
6b	280	265	1800	33	Nearly complete change
7	320	525	2950	15	Nearly complete change
8	350	500	4000	11	Almost no alteration
9	250	500	2000	12	Slight leak

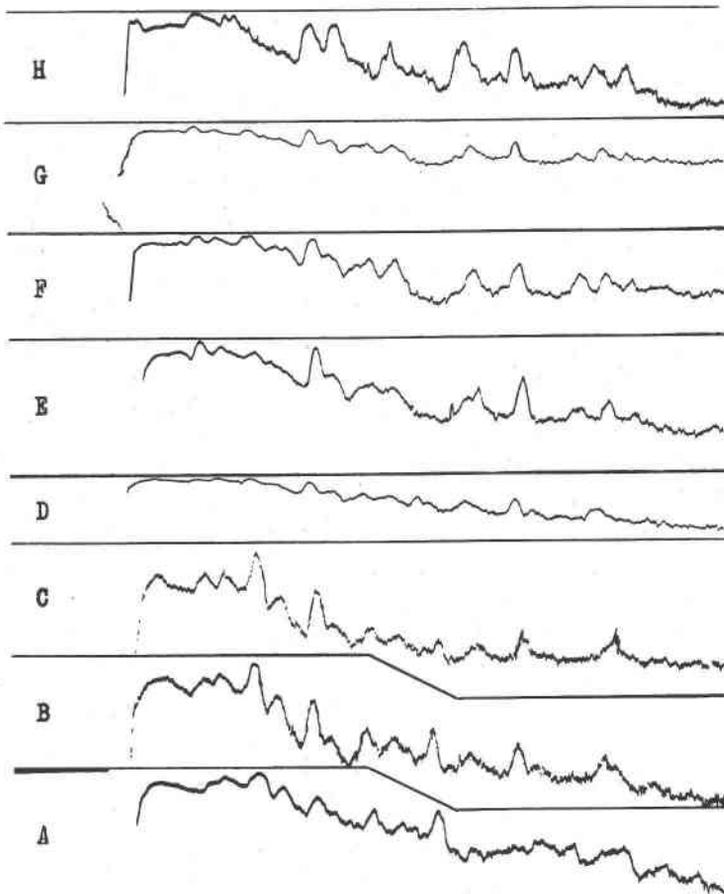


FIG. 3. Micro-photometer curves of x-ray patterns.

In order to determine the identity of the reaction product, *x*-ray patterns were made using the Cu  $K\alpha$  radiations from a Muller tube with a metal filter. The sample was mounted on a thread with collodion and placed in a standard Debey type camera with 57.3 mm. diameter. A .08 mm. aluminum screen was used to prevent haze in the background. As the pattern showed continuous lines, density curves were run on the Moll micro-photometer.

Curve *A*, in Fig. 3, is representative of the unaltered orthoclase feldspar. Curve *B* represents the treated sample from run No. 4, indicating a slight alteration from the original sample. Curve *C* is the product from run No. 6*a* and shows a slightly greater change from the original material. Curve *D*, from run No. 6*b*, indicates a still closer approach to the end product, although the high density of this particular negative somewhat obscures the details of the curve. Curve *E*, from run No. 7, is almost completely altered to sericite, as can readily be seen by comparing with the curves *F* and *G*, which represent muscovite and sericite. These latter curves are practically identical, although the variation in density of the negative somewhat decreases the amplitude of the peaks in curve *G*. Curve *H* represents an English china clay or kaolinite, shown in order to indicate the considerable difference from the decomposition product obtained in these runs. In fact, patterns were made from most of the hydrated aluminum silicates on the same apparatus in order to be sure that sericite alone satisfied the conditions.

An interesting fact brought out in Table 1 is that apparently no decomposition took place at 350°C., while comparatively rapid conversion occurred at 320°C. An upper temperature limit may lie between these values, above which the decomposition cannot proceed, but more work must be carried out before a definite statement can be made. As yet no definite conclusion can be drawn in regard to the influence of CO<sub>2</sub> pressure on the reaction.

Another check on the identity of the mineral formed can be obtained from the amount of combined water stable in the sample above 110°C. The following table shows this:

TABLE 2

Material	Ignition Loss
Orthoclase as finely ground	2.1%
Run #6 (10 hours)	4.0
Run #6 <i>a</i> (21 hours)	5.5
Run #6 <i>b</i> (33 hours)	5.9
Muscovite	6.2

It is apparent that the ignition loss increases regularly with the conversion, from 2.1% for the original feldspar to 6.2 for the sericite. These figures indicate fairly well the extent of the change.

We can also obtain some indication of the type of mineral formed in the decomposition by studying the thermal curves shown in Fig. 4. The first curve represents kaolinite with a heat absorption at 580°C.

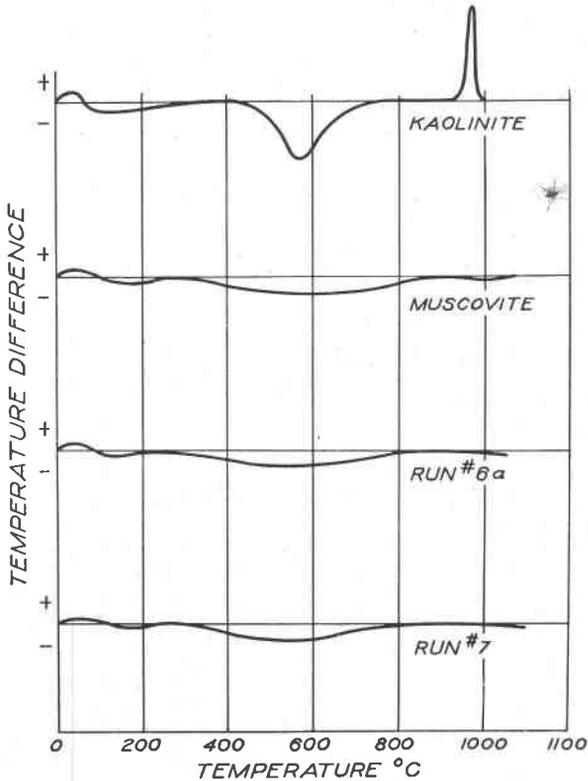
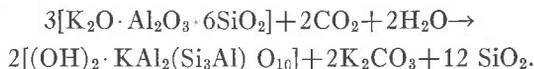


FIG. 4. Thermal curves.

and an evolution of heat at about 970°C. The second curve is for muscovite which probably has the same crystal structure as sericite, and it will be seen that there is only a slight heat absorption around 600°C., but no heat evolution to correspond with the kaolinite. The curves for run No. 6a and run No. 7 indicate a considerable similarity to the muscovite, and here again there is no trace found of the heat evolution at 970°C.

Therefore we may conclude from the confirming evidence of the *x*-ray diagrams, the heating curves, and the ignition loss that orthoclase is converted quite readily to the mineral sericite at temperatures up to 320°C. We may write the following equation to express the reaction occurring in this decomposition process:



The potassium carbonate and silica resulting from this change are taken away from the sample by the percolating water, leaving a concentration of potassium carbonate in the water at the bottom of the chamber. This silica was in the form of a hard, shiny crust firmly attached to the metal at the surface of the water. An *x*-ray film of this crust gave a definite pattern for quartz. Although no chemical analysis was made of the water left in the reaction chamber, it gave a fairly strong alkaline reaction, undoubtedly due to the dissolved potassium carbonate. In this particular feldspar, there was, of course, a certain proportion of soda, and this partially replaced the potash in the above equation.

#### *Anorthite*

Runs made on the anorthite are shown in the table below:

TABLE 3

Run No.	Temp. C.	CO <sub>2</sub> Pressure Cold	Total Pressure	Total Time at Temp.	Conversion
11	300	260	1950	18 days	No change
12	250	300	1230	23	Slight change
14	275	300	1650	24	Considerable change

An *x*-ray pattern of the original anorthite and the resultant product of run No. 14 is shown in Fig. 5.

It will be noted that there are two sets of lines; one dotted and one continuous. Therefore, a micro-photometer curve was not particularly satisfactory for use in analysis. The petrographical examination of the decomposition products showed a definite crystallization of calcite, and if we therefore take from the original *x*-ray pattern the lines of calcite,

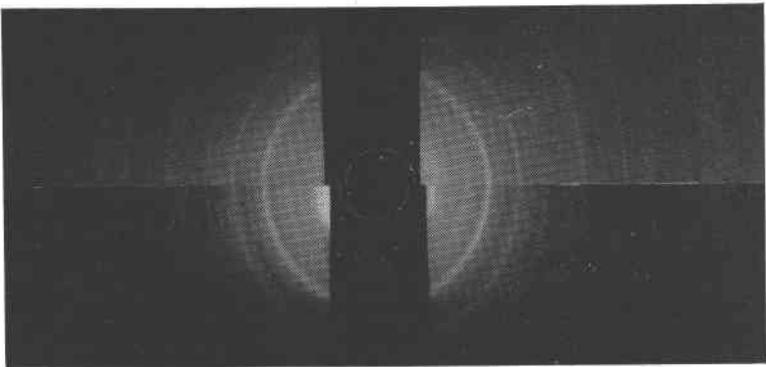
which are spotted due to the large size of the crystals, we find the resultant pattern agrees with that of the mineral pyrophyllite, except for one faint line which corresponds to a line from the quartz pattern. It is believed that a little quartz may have come from a slight disintegration of the bond in the porous crucible. The lines as measured on the film are shown in Table 4.

The reaction occurring in this decomposition may be expressed as follows:



In this case there was no deposit of quartz around the lining of the reaction chamber as was true of the orthoclase, and there was very

X-ray diagram of untreated anorthite.



X-ray diagram of treated anorthite.

FIG. 5

little evidence of the resulting calcite being carried away with the percolating water. As calcite is undoubtedly slightly soluble in water containing carbon dioxide, it seems strange that the crystals of calcite grow to such a large size in the porous cup. This may be due to the fact that the carbon dioxide was in such high concentration that it prevented any solution of the calcium carbonate.

The few runs that have been made seem to indicate that this reaction will not take place above 300°C., but will take place fairly rapidly at 275°C.

TABLE 4. ANORTHITE (Run #14)

Original Anorthite		Treated Anorthite		Pyrophyllite		Calcite	
Dist. in cms. on film	Intensity						
		2.00	2	1.95	8		
2.30	2	2.30	3				
		2.73	1	2.75	8	2.76	0.5
2.88	10	2.84	10	2.85	8		
3.14	2	3.10	1			3.06	8
3.28	2	3.27	1				
		3.44	1	3.42	6		
3.68	5	3.64	4	3.62	6	3.76	2
3.90	0.5						
4.10	0.5	4.00	1	4.05	2	4.09	3
4.34	3	4.33	3				
4.64	3	4.60	1				
		4.82	1	4.70	1	4.89	5
		5.02	2			4.93	5
5.30	2	5.27	2				
		5.42	1	5.38	2		
5.84	0.5	5.80	1			5.79	0.5
		6.13	2	6.10	2	6.24	1
6.40	2	6.34	2	6.38	0.5		
6.56	1	6.50	1			6.47	1
6.84	1	6.84	2				
7.04	2	7.02	2				
7.56	1	7.56	1				
		7.64	1				
8.06	0.5	8.00	1				
8.40	0.5	8.33	1			8.27	1

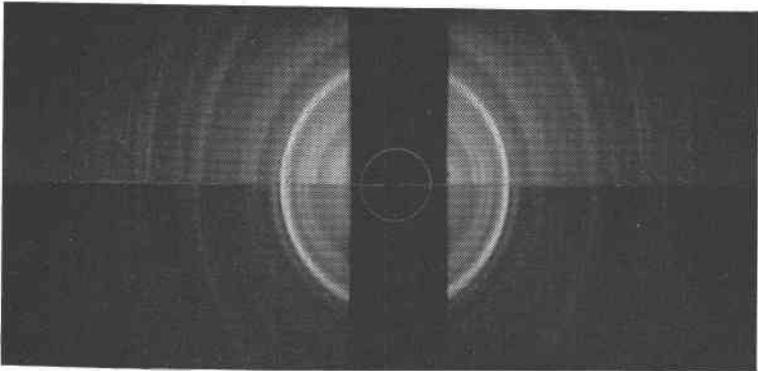
*Albite*

The mineral albite was tested in the reaction chamber as indicated in the table below.

TABLE 5

Run No.	Temp. C.	CO <sub>2</sub> Pressure Cold lbs./sq. in.	Total Pressure Hot lbs./sq. in.	Total Time at Temp.	Conversion
10	315	200	2100	19 days	No change
13	250	300	1230	23	Slight change
15	275	300	1650	24	Slight change

X-ray diagram of untreated albite.



X-ray diagram of treated albite.

FIG. 6

The *x*-ray patterns for the original and altered material shown in Fig. 6 indicate a slight structural change, but not as great as for the other minerals. The reason for this slight change is not certain, as it would be supposed that this mineral would be less stable than the potash feldspar. It is possible that we have not as yet selected a temperature giving the maximum rate of conversion. Also the reaction chamber always showed a light film of oil on the water, which came from the gauge and this may have retarded the reaction. Indications of the types of reaction products are not particularly clear, but seem to suggest an alteration to the mineral paragonite, a soda mica analogous to sericite. In Table 6 are given the *x*-ray film measurements. Only sericite, of the minerals checked, showed agreement with the new lines formed, and here a number of lines are missing. No pure paragonite was available for comparison. Longer runs to decompose the albite more completely will be necessary before definite identification is possible.

TABLE 6. ALBITE (Run #15)

Original Albite		Treated Albite		New Lines		Sericite	
Dist. in cms. on film	Intensity						
		2.04	6	2.04	6	2.00	10
2.24	5	2.24	6			2.20	8
2.44	5	2.44	6				

TABLE 6—Continued

		2.70	3	2.70	3	2.73	8
2.80	10	2.80	10				
3.09	4	3.07	4			3.18	0.5
3.56		3.56	6	3.56	6	3.53	10
3.74	3	3.71	3				
3.96	1	3.90	3				
4.32	3	4.30	2				
4.66	2	4.66	2			4.60	2
5.10	6	5.04	6				
		5.20	0.5	5.20	0.5		
5.58	2	5.60	4			5.62	3
5.90	1	5.92	0.5				
6.13	1	6.09	0.5				
6.27	1	6.27	6			6.32	8
6.42	2	6.44	2				
6.62	2	6.60	2				
6.87	3	6.85	2				
7.06	2	7.06	1			7.05	2
7.26	0.5	7.28	0.5				
		7.49	0.5				
		7.80	1	7.80	1	7.77	2
8.10	0.5	8.10	0.5				

DISCUSSION OF RESULTS

The upper temperature of 320°C. for the decomposition reaction found in these experiments is materially lower than that determined by Schwartz and Trageser. They state that pyrophyllite is a high temperature product, formed above 400°C., while below that temperature kaolin is produced. In the present investigation no kaolin was noted under any of the conditions of our tests. This difference may be due entirely to the comparatively low concentration of carbonic acid.

Sericite is sometimes an associate of kaolin in weathered feldspar, and by some geologists is considered an intermediate stage in the weathering process. The present tests would indicate that sericite was a final product of feldspar alteration at comparatively high temperatures; while kaolinite may be formed at lower temperatures or under more acid conditions. Much more work is necessary to get a clear understanding of the changes involved.

The reverse process, that is the change of kaolin or clay to sericite to form slates, is a common occurrence in nature. A study of this type of alteration would be of interest as it must entail the assimilation of potash from ground waters. We have at present very little information on the temperature range of this change.

## CONCLUSIONS

In this investigation there has been developed a reaction chamber for carrying on the accelerated alteration of feldspars under constant temperature conditions. The reaction simulates natural conditions because fresh liquid is constantly supplied to the sample and the soluble reaction products are constantly removed.

The three feldspars tested can be decomposed with water and carbon dioxide, the orthoclase changing to sericite, and the anorthite to pyrophyllite under the particular conditions of our tests. Maximum temperature limits for this reaction have been tentatively established at 350°C. for orthoclase, and 300°C. for anorthite. Albite was partially decomposed, but the end products are not as yet identified.

## FURTHER WORK

It is realized that the present work is only a preliminary survey of the alteration of feldspar, and further investigation should be continued with other types of feldspar, and over a wider range of temperature, to obtain a quantitative value for the rate of reaction. It is quite probable that the tests carried out at lower temperatures, or with lower concentrations of carbon dioxide, will form other products. It is believed that an increase in the speed of the reaction will take place by using still finer particles of feldspar, which can be obtained by separating in a centrifuge. It would also be advisable to interrupt the run periodically and regrind the reacting material to break up the rather dense crust that forms on the surface of the specimen. This regrinding would expose fresh surfaces and give the sample more permeability.

This work has been made possible by the generosity of the Geological Society of America who have allotted funds to carry out this investigation. The author takes this occasion to express his appreciation for the assistance in this matter.

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