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INTRODUCTION.

The investigation of the ternary system MgO-Al₂O₃-SiO₂ was undertaken in order to determine all the crystalline substances formed from pure melts of these oxides, and to establish the equilibrium relations of such of the crystals as are stable in contact with the liquid. Stated briefly, this involved a microscopical study to identify the different crystalline substances, and a thermal study to determine the melting temperatures of these substances when alone or in mixtures.

THE CRYSTALLINE PHASES.

The crystalline substances found to be stable in contact with liquid are here enumerated: the three components, MgO (periclasé), Al₂O₃ (corundum), SiO₂ (tridymite, cristobalite); the four binary compounds, 2MgO.SiO₂ (forsterite), MgO.SiO₂ (clinoenstatite), MgO.Al₂O₃ (spinel), Al₂O₃.SiO₂ (sillimanite); and a ternary compound, 2MgO.2Al₂O₃.5SiO₂ (which may be considered an end member in the series of cordierites). Since the properties of the components and binary compounds have been described in previous papers,¹ they will be treated

¹ The following papers summarize and amplify what had been done previously: The Binary System MgO-SiO₂, N. L. Bowen and Olaf Andersen, this Journal, 37, 478, 1914; The Ternary System CaO-Al₂O₃-SiO₂, G. A. Rankin and F. E. Wright, *ibid.*, 39, 1-79, 1915; The Ternary System CaO-Al₂O₃-MgO, G. A. Rankin and H. E. Merwin, J. Am. Chem. Soc., 38, 568, 1916; and The Ternary System Diopside-forsterite-silica, N. L. Bowen, this Journal, 38, 207, 1914.

only incidentally here,² but the equilibrium relations of these forms in ternary mixtures of $\text{MgO-Al}_2\text{O}_3\text{-SiO}_2$ will be considered in detail. On the other hand, cordierite of such composition, and an unstable form having similar composition which appears at lower temperatures, have not been described before,³ therefore their properties and mode of formation will be taken up.

GENERAL PROCEDURE.

In this account only such additional apparatus, experimental methods and applications of the phase rule will be described as seem to be necessary for understanding this particular problem.

The first step in this investigation consisted in the preparation from especially pure MgO , Al_2O_3 and SiO_2 of a few furnace charges whose composition represented random intervals over the entire range of ternary mixtures. It may be said concerning these: (1) that compositions high in SiO_2 , being viscous, require repeated fusions with subsequent fine grinding in order to insure chemical homogeneity throughout the charge. This condition is established in melts which do not crystallize readily on cooling by the determination of the uniformity in the refractive index of the glass obtained from such melts; (2) that those melts containing so high a percentage of either MgO , or Al_2O_3 that they melt but little if any below the melting point of platinum,⁴ about 1750, further heated in the iridium furnace; this insures the formation of sufficient liquid to permit complete combination to take place; (3) that in many of the compositions, particularly those high in alumina, the compounds are readily formed from their components at temperatures at which there is but little if any liquid present.

Having prepared a number of chemically homogenous charges of different compositions, the next step was the

² In these melts clinoenstatite is seldom twinned. Needles, prisms and tables have been found in the same quench.

³ J. Morozewicz, "Experimentelle Untersuchungen über die Bildung der Minerale im Magma," p. 68, briefly described crystals obtained from silicate melts, which are similar to this high temperature form, and which he called cordierite. The relation of various anhydrous crystals to cordierite will be discussed under another heading.

⁴ All charges are fused in platinum crucibles placed in a Fletcher gas-blast furnace.

determinations of their primary crystalline phases; that is, the determinations of the crystalline substance which is the first to crystallize from a completely melted cooling charge, or the last to melt in a crystalline charge when heated. The determination of the primary crystalline phase in a given composition is carried out by heating a small charge of the material to various constant temperatures for a period of time sufficient to establish equilibrium, and suddenly cooling (quenching) so as to prevent the crystallization of any liquid which may have formed. By proceeding systematically in this way one eventually obtains a quench which contains a single crystalline substance embedded in glass.

In this manner the primary phases which could be obtained below 1600°⁵ were determined in the first composition prepared. These data give one an idea not only as to the different crystalline compounds which will be found in the system MgO, Al₂O₃, SiO₂ but also an idea as to the melting temperatures throughout this system. With the preliminary data, thus obtained, as a working basis, other compositions were made up and the primary phases determined. Proceeding systematically in this way to prepare charges, whose compositions are such as to give further information as to the crystalline substances found in this system, the range of compositions in which each such substance occurs as a primary phase was determined. In other words, using phase rule terms, the equilibrium field of each such crystalline substance was established.

For the study of the composition of the ternary compound, glasses were required which contained no crystals. Many of these glasses required preparation at temperatures considerably above 1600°. Small charges were melted in the gas blast furnace and subsequently rapidly chilled by immersing in water.

LIMITS OF THE FIELDS.

Data which determine the limits of each field are given in tables 1-7 inclusive. These data are presented graphically in the triangular diagram, figure 1.

⁵ 1600° C is about the upper limit of the platinum-resistance furnace from which the charges are quenched by dropping into a dish of mercury.

TABLE 1.

Quenchings which determine the limits of the field of spinel ($\text{MgO} \cdot \text{Al}_2\text{O}_3$)¹; see also Table 8.

Composition	<i>Phases present, glass + spinel.</i>			Time in hours
	Weight MgO Al_2O_3	Per cent SiO_2	Temp.	
22.0	73.0	5.0	1900 ²	
15.0	80.0	5.0	1900 ²	
15.0	60.0	25.0	1750 ²	
55.0	18.0	27.0	1750 ²	
49.0	21.0	30.0	1700 ²	
37.0	23.0	40.0	1600	.25
39.0	21.0	40.0	1600	.25
17.0	38.0	45.0	1475	.5
30.0	25.0	45.0	1450	.5
25.0	24.0	51.0	1375	.5

¹ Formulas written thus are ideal. The actual crystals formed in parts of the field may have quite a different composition. In the case of spinel solid solution is extensive.

² Iridium furnace melt.

TABLE 2.

Quenchings which determine the limits of the field of forsterite ($2MgO.SiO_2$); see also Table 9.

<i>Phases present, glass + $2MgO.SiO_2$.</i>				
Composition MgO	Weight Al_2O_3	Per cent SiO_2	Temp.	Time in hours
31.0	22.0	47.0	1450	.5
30.0	20.0	50.0	1450	.5
26.0	22.0	52.0	1370	.5
28.0	20.0	52.0	1400	.5
26.0	20.0	54.0	1377	.5
32.0	11.0	57.0	1465	.5

TABLE 3.

Quenchings which determine the limits of the field of clinoenstatite ($MgO.SiO_2$); see also Table 10.

<i>Phases present, glass + $MgO.SiO_2$.</i>				
Composition MgO	Weight Al_2O_3	Per cent SiO_2	Temp.	Time in hours
25.0	18.0	57.0	1360	.5
30.0	10.0	60.0	1465	1.0

TABLE 4.

Quenchings which determine the limits of the field of SiO_2 ;
see also Table 11.

<i>Phases present, SiO_2 + glass.</i>				
Composition MgO	Weight Al_2O_3	Per cent SiO_2	Temp.	Time in hours
20.0	17.0	63.0	1370	.5
15.0	20.0	65.0	1390	.5
27.0	8.0	65.0	1475	.5
10.0	23.0	67.0	1445	.5
9.0	22.0	69.0	1450	.5

TABLE 5.

Quenchings which determine the limits of the field of sillimanite ($Al_2O_3.SiO_2$); see also Table 12.

<i>Phases present, glass + $Al_2O_3.SiO_2$.</i>				
Composition MgO	Weight Al_2O_3	Per cent SiO_2	Temp.	Time in hours
12.0	46.0	42.0	1575	.5
16.0	38.0	46.0	1490	.5
16.0	35.0	49.0	1465	2.0
15.0	35.0	50.0	1500	.5
15.0	34.0	51.0	1465	1.0
12.4	31.6	56.0	1490	.5
10.4	26.6	63.0	1425	.5
9.0	24.0	67.0	1450	.5
7.0	22.0	71.0	1475	.5

TABLE 6.

Quenchings which determine limits of field of cordierite
($2\text{MgO} \cdot 2\text{Al}_2\text{O}_3 \cdot 5\text{SiO}_2$); see also Table 13.

Phases present, cordierite + glass.

Composition MgO	Weight Al_2O_3	Per cent SiO_2	Temp.	Time in hours
16.0	34.0	50.0	1450	1.0
17.0	33.0	50.0	1460	.5
16.0	33.0	51.0	1450	1.0
21.0	28.0	51.0	1430	1.0
16.5	32.0	51.5	1450	1.0
18.0	30.0	52.0	1400	1.5
16.0	31.0	53.0	1450	1.0
25.0	22.0	53.0	1370	.5
16.0	30.3	53.7	1450	1.0
15.0	30.0	55.0	1440	.5
20.0	19.0	61.0	1350	.5

TABLE 7.

Quenchings which determine the limits of the field of Al_2O_3 .

Phases present, glass + α Al_2O_3 (corundum).

Composition MgO	Weight Al_2O_3	Per cent SiO_2	Temp.	Time in hours
10.0	70.0	20.0	1750 ¹	
10.0	65.0	25.0	1750 ¹	
14.0	47.0	39.0	1585	.5
15.0	44.0	41.0	1575	.5

¹ Iridium furnace melt.

While it is not our present purpose to discuss each of these fields at length, it may be well to state here certain special facts concerning the fields and their respective crystalline phases.

Fields of Stable Compounds.

The components MgO , Al_2O_3 , SiO_2 and the compounds $2\text{MgO} \cdot \text{SiO}_2$, $\text{MgO} \cdot \text{Al}_2\text{O}_3$ and probably $\text{Al}_2\text{O}_3 \cdot \text{SiO}_2$ do not dissociate when melting, therefore the composition of each necessarily lies within its respective field. The accurate determinations of the limits of these fields were in most cases made without difficulty. The limits of the field of MgO and certain portions of the fields of Al_2O_3 , $\text{Al}_2\text{O}_3 \cdot \text{SiO}_2$, $\text{MgO} \cdot \text{Al}_2\text{O}_3$ and $2\text{MgO} \cdot \text{SiO}_2$, however, could not be determined with extreme accuracy for the reason that the temperatures required are beyond the range of the platinum quenching furnace, 1600° . Due to the pecu-

liar circumstance, however, that the excess glass from compositions containing between 65% Al_2O_3 and 85% Al_2O_3 did not crystallize readily even when slowly cooled from the high temperature (2100°) obtained in the iridium furnace, it was possible to obtain Al_2O_3 (corundum) and spinel⁶ each alone imbedded in glass in certain compositions. This enabled us to determine more accurately than would otherwise have been possible the limits of certain portions of the fields of Al_2O_3 and spinel. Aside from these data the limits of the portions of the fields mentioned which are at temperatures too high for direct study were determined indirectly from certain theoretical reasonings based on experimental evidence. This evidence and theory will be taken up later when we come to consider the boundary curves and quintuple points.

Fields of Unstable Compounds.

Clinoenstatite, $MgO.SiO_2$, and cordierite have no definite melting points, each being a crystalline substance of such a nature that when pure it dissociates when melting begins, therefore their compositions lie outside of their respective fields. The determinations of the limits of these two fields involved no especial difficulty since each lies wholly within the range of temperatures possible in the platinum quenching furnace. The determination of the composition of such unstable crystalline substances may be, however, a matter of extreme difficulty. Of the two such substances which we have to consider in this ternary system one, $MgO.SiO_2$, is a definite compound of constant composition; the other, cordierite, is affected by solid solution, and is so fine-grained when crystallized from a glass at temperature below that at which it begins to melt, that its homogeneity cannot be accurately tested. (See p. 316.)

MELTING TEMPERATURES WITHIN THE FIELDS.

Of the eight fields of the diagram two have melting temperatures all of which may be determined in the platinum quenching furnace. These are the fields of $MgO.SiO_2$ and the ternary compound (cordierite).

⁶ Microscopical examination of various iridium furnace melts containing only spinel and a small percentage of glass would seem to indicate that spinel in ternary mixtures takes up a small amount of SiO_2 in solid solution. This together with the extent of the solid solution between Al_2O_3 and $MgO.Al_2O_3$ in the binary system is indicated in fig. 1.

Part of the melting temperatures of all the other fields, except the field of MgO, may be thus determined. Tables 8-13 show these ascertained melting temperatures for various compositions within each field. From these temperatures and others referred to later the isotherms of fig. 6 were drawn.

TABLE 8.

Quenchings which determine melting temperatures of various compositions within field A-6-4-3-7-B; Primary phase is $(\text{MgO} \cdot \text{Al}_2\text{O}_3)$ spinel.

Composition MgO	Weight Al_2O_3	Per cent SiO_2	Temp.	Time in hours	Phases present
17.5	42.5	40.0	1600	.25	Glass + tr. spinel
25.0	32.0	43.0	1583	.5	Glass
			1570	.5	Glass + spinel
29.0	23.0	48.0	1450	.5	Glass
			1430	.5	Glass + spinel
20.0	30.0	50.0	1450	.5	Glass
			1435	.5	Glass + spinel
25.0	25.0	50.0	1410	.5	Glass
			1390	.5	Glass + spinel
26.0	23.0	51.0	1380	1.0	Glass
			1376	1.0	Glass + spinel

TABLE 9.

Quenchings which determine melting temperatures of various compositions within the field G-6-4-5-F; Primary phase is $2\text{MgO} \cdot \text{SiO}_2$.

Composition MgO	Weight Al_2O_3	Per cent SiO_2	Temp.	Time in hours	Phases present
35.0	20.0	45.0	1555	.5	Glass
			1550	.5	Glass + tr. $2\text{MgO} \cdot \text{SiO}_2$
35.0	15.0	50.0	1560	.5	Glass
			1530		Glass + tr. $2\text{MgO} \cdot \text{SiO}_2$
26.0	21.0	53.0	1375	.5	Glass
			1365	.5	Glass + $2\text{MgO} \cdot \text{SiO}_2$
27.0	18.0	55.0	1415	.5	Glass
			1390	.5	Glass + $2\text{MgO} \cdot \text{SiO}_2$

TABLE 10.

Quenchings which determine melting temperatures of various compositions within field is F-5-1-E; Primary phase is $\text{MgO} \cdot \text{SiO}_2$.

Composition MgO	Weight Al_2O_3	Per cent SiO_2	Temp.	Time in hours	Phases present
25.0	20.0	55.0	1375	.5	Glass
			1365	.5	Glass + $\text{MgO} \cdot \text{SiO}_2$
24.0	17.0	59.0	1383	1.5	Glass
			1370	1.0	Glass + $\text{MgO} \cdot \text{SiO}_2$
35.0	5.0	60.0	1515	.5	Glass
			1510	.5	Glass + $\text{MgO} \cdot \text{SiO}_2$
21.0	18.0	61.0	1370	.5	Glass
			1360	.5	Glass + $\text{MgO} \cdot \text{SiO}_2$
25.0	12.0	63.0	1440	.5	Glass
			1420	1.0	Glass + $\text{MgO} \cdot \text{SiO}_2$
30.0	6.0	64.0	1500	.5	Glass
			1498	.5	Glass + $\text{MgO} \cdot \text{SiO}_2$
25.0	11.0	64.0	1450	1.0	Glass
			1425	.5	Glass + $\text{MgO} \cdot \text{SiO}_2$

TABLE 11.

Quenchings which determine melting temperatures of various compositions within field E-1-2-D; Primary phase is SiO_2 .

Composition MgO	Weight Al_2O_3	Per cent SiO_2	Temp.	Time in hours	Phases present
20.0	15.0	65.0	1455	.5	Glass
			1450	.5	Glass + SiO_2
11.0	22.0	67.0	1445	.5	Glass
			1435	.5	Glass + SiO_2
20.0	18.0	62.0	1355	.5	Glass
			1352	.5	Glass + SiO_2
15.0	15.0	70.0	1530	.5	Glass
			1500	.5	Glass + SiO_2

TABLE 12.

Quenchings which determine melting temperatures of various compositions within field D-2-3-7-C; Primary phase is $Al_2O_3.SiO_2$.

Composition MgO	Weight Al_2O_3	Per cent SiO_2	Temp.	Time in hours	Phases present
15.0	32.0	53.0	1460	1.0	Glass
			1457	1.0	Glass + $Al_2O_3.SiO_2$
10.0	24.0	66.0	1438	1.0	Glass
			1435	1.0	Glass + $Al_2O_3.SiO_2$
16.0	40.0	44.0	1550	.25	Glass
			1525	.25	Glass + $Al_2O_3.SiO_2$
14.5	36.5	49.0	1550	.25	Glass
			1525	.25	Glass + $Al_2O_3.SiO_2$
10.0	30.0	60.0	1550	.25	Glass
			1525	.25	Glass + $Al_2O_3.SiO_2$

TABLE 13.

Quenchings which determine melting temperatures of various compositions within field 1-2-3-4-5; Primary phase is the ternary compound (cordierite).

Composition MgO	Weight Al_2O_3	Per cent SiO_2	Temp.	Time in hours	Phases present
16.0	34.0	50.0	1460	1.0	Glass
			1457	1.0	Glass + cordierite
20.0	23.0	57.0	1400	.5	Glass
			1375	.5	Glass + cordierite
18.0	30.0	52.0	1450	.5	Glass
			1425	.5	Glass + cordierite
25.0	23.0	52.0	1370	.5	Glass
			1365	.5	Glass + cordierite
20.0	25.0	55.0	1415	.5	Glass
			1400	.5	Glass + cordierite
15.0	30.0	55.0	1460	.5	Glass
			1450	.5	Glass + cordierite
23.0	20.0	57.0	1375	.5	Glass
			1350	1.0	Glass + cordierite
15.0	27.0	58.0	1450	.5	Glass
			1425	.5	Glass + cordierite
20.0	20.0	60.0	1365	1.0	Glass
			1340	.5	Glass + cordierite
15.0	23.0	62.0	1425	.5	Glass
			1400	.5	Glass + cordierite
11.0	23.0	66.0	1450	1.0	Glass
			1440	.5	Glass + cordierite

THE BOUNDARY CURVES.

The boundary lines in fig. 1 represent the projection upon the horizontal plane of the lines on the solid concentration-temperature diagram. In order to make clear the way in which the temperature varies along these

FIG. 2.

lines, we have made projections upon a vertical plane, obtaining in this way the series of boundary curves reproduced in fig. 2, in which the numbers and letters used correspond to those given in fig. 1. The ordinates represent temperatures which were determined experimentally.

These temperatures which are ascertained by the quenching method are contained in Table 14. The temperatures at the ends of the boundary curves are given later as quintuple and quadruple point temperatures.

Of the 14 boundary curves in this system, all of which are given in fig. 2, there are three which are represented by dotted lines to indicate that the compositions and temperatures along these lines could not be accurately determined, because direct methods could not be used. The indirect methods employed will be described when quintuple point 6 is considered. Two other boundary curves, 2-3 and 1-5, are so flat at their summits that from temperatures of complete melting alone it has not been feasible to determine whether or not either contains a maximum. However, from crystallization curves a maximum is evident in 2-3 (see Table 14a), and must lie between 53% and 60% SiO_2 . Several crystallization curves for compositions near point 5 were run, but they did not definitely indicate a maximum on line 1-5.

TABLE 14.

Quenchings which determine temperatures for points on the boundary curves.

Comp. Weight		Per cent	Temp.	Time	Phases present	Bound-
MgO	Al_2O_3	SiO_2		in hours		ary
30.0	6.0	64.0	1482	.5	Glass + $MgO.SiO_2$	E-1
			1460	2.0	Glass + $MgO.SiO_2$ + SiO_2	
27.0	8.0	65.0	1475	.5	Glass + SiO_2	1-2
			1450	1.0	Glass + SiO_2 + $MgO.SiO_2$	
20.0	19.0	61.0	1365	1.0	Glass + cordierite	
			1350	.5	Glass + cordierite + SiO_2	
15.0	20.0	65.0	1400	.5	Glass	D-2
			1390	.5	Glass + SiO_2 + cordierite	
7.0	22.0	71.0	1485	.5	Glass + $Al_2O_3.SiO_2$	
			1475	.5	Glass + $Al_2O_3.SiO_2$ + SiO_2	
10.0	24.0	66.0	1435	1.0	Glass + $Al_2O_3.SiO_2$	2-3
			1430	1.0	Glass + $Al_2O_3.SiO_2$ + cordierite	
15.0	32.0	53.0	1460	.5	Glass + $Al_2O_3.SiO_2$	3-7
			1455	.5	Glass + $Al_2O_3.SiO_2$ + cordierite	
16.0	35.0	49.0	1465	2.0	Glass + $Al_2O_3.SiO_2$	
			1460	1.0	Glass + $Al_2O_3.SiO_2$ + cordierite	
16.0	38.0	46.0	1490	.5	Glass + $Al_2O_3.SiO_2$	3-4
16.0	40.0	44.0	1550	.5	Glass	
17.0	33.0	50.0	1525	.5	Glass + $Al_2O_3.SiO_2$ + spinel (tr.)	4-6
			1458	.5	Glass + cordierite	
20.0	30.0	50.0	1456	1.0	Glass + cordierite + spinel	
			1435	.5	Glass + spinel	
25.0	25.0	50.0	1425	.5	Glass + spinel + cordierite	4-6
			1390	.5	Glass + spinel	
21.0	28.0	51.0	1375	1.0	Glass + spinel + cordierite	
			1430	1.0	Glass + cordierite	
26.0	23.0	51.0	1425	1.0	Glass + cordierite + spinel	4-6
			1376	1.0	Glass + spinel	
31.0	22.0	47.0	1375	.5	Glass + spinel + $2MgO.SiO_2$	
			1450	.5	Glass + $2MgO.SiO_2$	
37.0	21.0	42.0	1435	.5	Glass + $2MgO.SiO_2$ + spinel	4-6
			1600	.25	Glass	

TABLE 14 (continued).

Quenchings which determine temperatures for points on the boundary curves.

Comp.	Weight	Per cent	Temp.	Time	Phases present	Bound-
MgO	Al ₂ O ₃	SiO ₂		in hours		ary
			1585	.25	Glass + tr. 2MgO.SiO ₂ + tr. spinel	
37.0	23.0	40.0	1600	.25	Glass + spinel	
			1550	.5	Glass + spinel + 2MgO.SiO ₂	
26.0	23.0	51.0	1376	1.0	Glass + spinel	4.5
			1370	1.0	Glass + spinel + 2MgO.SiO ₂ + [cordierite	
			1365	.5	Glass + 2MgO.SiO ₂ + cordierite	
18.0	30.0	52.0	1390	.5	Glass + cordierite	
			1360	1.0	Glass + cordierite + 2MgO.SiO ₂	
22.0	25.0	53.0	1362	.5	Glass + cordierite	
			1359	1.0	Glass + cordierite + 2MgO.SiO ₂	
32.0	11.0	57.0	1465	.5	Glass + 2MgO.SiO ₂	F.5
			1450	.5	Glass + 2MgO.SiO ₂ + MgO.SiO ₂	
27.0	18.0	55.0	1390	.5	Glass + 2MgO.SiO ₂	
			1370	.5	Glass + MgO.SiO ₂	
26.0	20.0	54.0	1377	.5	Glass + 2MgO.SiO ₂	
			1370	.5	Glass + 2MgO.SiO ₂ + MgO.SiO ₂	
25.5	21.0	53.5	1361	.5	Glass	
			1360	.5	Glass + 2MgO.SiO ₂ + MgO.SiO ₂	
26.0	21.0	53.0	1365	.5	Glass + 2MgO.SiO ₂	
			1360	.5	Glass + 2MgO.SiO ₂ + MgO.SiO ₂	
25.0	20.0	55.0	1355	.5	Glass + MgO.SiO ₂	5.1
			1350	.5	Glass + MgO.SiO ₂ + cordierite	
23.0	20.0	57.0	1352	1.0	Glass + cordierite	
			1350	.5	Glass + cordierite + MgO.SiO ₂	
21.0	18.0	61.0	1360	.5	Glass + MgO.SiO ₂	
			1350	.5	Glass + MgO.SiO ₂ + cordierite	
12.0	46.0	42.0	1575		Glass + Al ₂ O ₃ .SiO ₂ + <i>a</i> Al ₂ O ₃ (tr.)	C-7
15.0	44.0	41.0	1575		Glass + spinel (tr.) + <i>a</i> Al ₂ O ₃	B-7
14.0	47.0	39.0	1600		Glass + spinel (tr.) + <i>a</i> Al ₂ O ₃	

TABLE 14a.

Quenchings which indicate a maximum on the boundary curve between sillimanite and the ternary compound (cordierite).

Comp.	Weight	Per cent	Temp.	Phases present
MgO	Al ₂ O ₃	SiO ₂		
16.0	33.0	51.0	1460	Glass + <i>a</i> ternary compound
			1400	Glass + <i>a</i> ternary compound + spinel
15.0	34.0	51.0	1465	Glass + Al ₂ O ₃ .SiO ₂
			1460	Glass + Al ₂ O ₃ .SiO ₂ (not yet resorbed) + ter-
				[nary compound + spinel
18.0	30.0	52.0	1400	Glass + ternary compound
			1360	Glass (?) + ternary compound + 2MgO.SiO ₂
				[+ ?
14.4	35.6	50.0	1525	Glass + Al ₂ O ₃ .SiO ₂
			1400	Glass + Al ₂ O ₃ .SiO ₂ (not yet resorbed) + spinel

THE QUINTUPLE POINTS.

Of the seven quintuple points in this system, one (point 1, fig. 1) is a well-established eutectic; another

(point 6, fig. 1), at a temperature beyond the reach of the platinum resistance furnace, is probably a eutectic; and a third (point 5, fig. 1) at the end of a flat boundary curve where it has been found impossible to be certain of equilibrium within a reasonable time, may be a eutectic.

Point 6 has been located and characterized as a eutectic according to theoretical reasoning based on such experimental evidence as could be obtained. If this point is a eutectic its composition must necessarily lie within the triangle MgO , $\text{MgO} \cdot \text{Al}_2\text{O}_3$, $2\text{MgO} \cdot \text{SiO}_2$, but if it is not a eutectic its composition must lie outside of this triangle. In either case, however, it is necessary that the three boundary curves which meet at this point be of such a nature that each one will intersect the angle formed by the other two. From these considerations and a glance at points 4, A, and G (fig. 1), the starting points of these three boundaries, 4-6, A-6 and G-6 respectively, it is evident that if point 6 is a eutectic it will probably be of a composition near the line joining $\text{MgO} \cdot \text{Al}_2\text{O}_3$ and $2\text{MgO} \cdot \text{SiO}_2$ and inside the triangle $\text{MgO} \cdot \text{Al}_2\text{O}_3$ - MgO - $2\text{MgO} \cdot \text{SiO}_2$, since the general direction of boundary 4-6 is fixed by actual data, and theoretically boundary G-6 will only intersect the angle formed by A-6 and 4-6 if such is the case. Granting for the moment that point 6 is a eutectic, let us consider the nature of indirect experimental evidence which would prove this to be so. If point 6 is thus a eutectic the composition line $\text{MgO} \cdot \text{Al}_2\text{O}_3$ - $2\text{MgO} \cdot \text{SiO}_2$ will be the projection of the concentration-temperature diagram for the binary eutectic system of these two compounds, and the binary eutectic composition will be at the intersection of this line and the boundary 4-6. This point of intersection represents a composition which is the maximum melting temperature on the boundary 4-6. That $\text{MgO} \cdot \text{Al}_2\text{O}_3$ - $2\text{MgO} \cdot \text{SiO}_2$ is a binary eutectic system was shown experimentally by microscopical examination of mixtures of these two compounds which had previously been fused in the iridium furnace. These examinations showed such material to contain only these two compounds, while if $\text{MgO} \cdot \text{Al}_2\text{O}_3$ - $2\text{MgO} \cdot \text{SiO}_2$ did not form a true eutectic series, well-developed crystals of MgO undoubtedly would have been present. Also such melting temperatures as could be determined for these mixtures in the iridium furnace would tend to show

that the intersection point P represents a maximum temperature on the boundary 4-6.

It is upon such indirect evidence that point 6 is designated as a eutectic.

TABLE 15.

Quenchings which show the course of the crystallization curves which pass through quintuple points (not eutectics) 2 and 4, and that cross the field of the ternary compound (cordierite) near point 3.

Comp.	Weight	Per cent	Temp.	Time in hours	Phases	Point
MgO	Al ₂ O ₃	SiO ₂				
10.0	24.0	66.0	1438	1.0	Glass	(2)
			1435	1.0	Glass + Al ₂ O ₃ .SiO ₂	
			1428	1.0	Glass + Al ₂ O ₃ .SiO ₂ + cordierite	
			1425	1.0	Glass + SiO ₂ (tridymite) + cordierite	
16.0	35.0	49.0	1465	2.0	Glass + Al ₂ O ₃ .SiO ₂	(3)
			1457	1.0	Glass + Al ₂ O ₃ .SiO ₂ + cordierite	
			1450	4.0	Glass + spinel + cordierite	
26.0	23.0	51.0	1378	1.0	Glass	(4)
			1376	1.0	Glass + spinel	
			1375	.5	Glass + 2MgO.SiO ₂ + cordierite	

Following is a list of the 7 quintuple points giving composition in weight percentages of MgO, Al₂O₃, SiO₂ and the corresponding temperatures as determined by quenches. The numbers assigned to each quintuple point correspond to the numbers in fig. 1.

Point 1 is the eutectic for MgO.SiO₂, SiO₂ and the ternary compound (MgO 20.3, Al₂O₃ 18.3, SiO₂ 61.4). Its melting point (1345°±5°) is based on the values obtained from various compositions as given in Table 16.

TABLE 16.

Melting temperature: Quintuple point 1 for MgO.SiO₂, SiO₂ and ternary compound as determined from various compositions.

Composition, weight, per cent	{ MgO	20.0	18.9	25.0	20.0	25.0
	{ Al ₂ O ₃	19.0	24.1	18.0	18.0	20.0
	{ SiO ₂	61.0	57.0	57.0	62.0	55.0
Temperature		1348	1340	1340	1345	1345

Point 2 is the quintuple point for SiO₂, ternary compound and Al₂O₃.SiO₂ (MgO 10.0, Al₂O₃ 23.5, SiO₂ 66.5). Its melting temperature (1425°±5°) is based on the values obtained from compositions given in Table 17.

TABLE 17.

Melting temperature: Quintuple point 2 for SiO₂, ternary compound and Al₂O₃.SiO₂ as determined from various compositions.

Composition, weight, per cent	{ MgO	12.4	10.0	10.0	9.0
	{ Al ₂ O ₃	31.6	30.0	24.0	24.0
	{ SiO ₂	56.0	60.0	66.0	67.0
Temperature		1425	1425	1425	1425

Point 3 is the quintuple point for ternary compound, $Al_2O_3 \cdot SiO_2$ and spinel (MgO 16.1, Al_2O_3 34.8, SiO_2 49.1). Its melting temperature ($1460^\circ \pm 5^\circ$) is based on the values obtained from various compositions as given in Table 18.

TABLE 18.

Melting temperature: Quintuple point 3 for ternary compound, $Al_2O_3 \cdot SiO_2$ and spinel as determined from various compositions.

Composition, weight, per cent	MgO	15.3	16.0	16.0	16.5
	Al_2O_3	38.8	38.0	35.0	34.5
	SiO_2	45.9	46.0	49.0	49.0
Temperature		1460	1455	1465	1460

Point 4 is the quintuple point for spinel, ternary compound, and $2MgO \cdot SiO_2$ (MgO 25.7, Al_2O_3 22.8, SiO_2 51.5). Its melting temperature ($1370^\circ \pm 5^\circ$) is based on the values obtained from compositions as given in Table 19.

TABLE 19.

Melting temperature: Quintuple point 4 for ternary compound, spinel and $2MgO \cdot SiO_2$ as determined from various compositions.

Composition, weight, per cent	MgO	26.0	29.0	25.0
	Al_2O_3	23.0	23.0	24.0
	SiO_2	51.0	48.0	51.0
Temperature		1370	1370	1363

Point 5 is the quintuple point (eutectic?) for $2MgO \cdot SiO_2$, $MgO \cdot SiO_2$, and ternary compound (MgO 25.0, Al_2O_3 21.0, SiO_2 54.0). Its melting temperature ($1360^\circ \pm 5^\circ$) is based on the values obtained from various compositions as given in Table 20.

TABLE 20.

Melting temperature: Quintuple point 5 for ternary compound, $MgO \cdot SiO_2$, and $2MgO \cdot SiO_2$ as determined from various compositions.

Composition, weight, per cent	MgO	22.0	25.0	26.0	25.5	25.0
	Al_2O_3	25.0	22.0	21.0	21.0	21.0
	SiO_2	53.0	53.0	53.0	53.5	54.0
Temperature		1357	1360	1355	1353	1363

Point 6 is the eutectic point for $2MgO \cdot SiO_2$, MgO , and spinel (MgO 56.0, Al_2O_3 16.0, SiO_2 28.0). Its melting temperature ($1700^\circ \pm 25^\circ$) is based on values obtained from several compositions melted in the iridium furnace, temperature measurements being made with an optical pyrometer.

Point 7 is the quintuple point for Al_2O_3 , spinel and $Al_2O_3 \cdot SiO_2$ (MgO 15.2, Al_2O_3 42.0, SiO_2 42.8). Its melt-

ing temperature ($1575^{\circ} \pm 5^{\circ}$) is based on values obtained from various compositions as given in Table 21.

TABLE 21.

Melting temperature: Quintuple point 7 for spinel, $\text{Al}_2\text{O}_3 \cdot \text{SiO}_2$, and $\alpha\text{-Al}_2\text{O}_3$ as determined from various compositions.

Composition, weight, per cent	$\left\{ \begin{array}{l} \text{MgO} \\ \text{Al}_2\text{O}_3 \\ \text{SiO}_2 \end{array} \right.$	10.0	15.0	15.0	14.0	15.0
		70.0	60.0	44.0	47.0	48.0
		20.0	25.0	41.0	49.0	37.0
Temperature		1575	1585	1570	1568	1568

THE COMPOSITION AND PROPERTIES OF THE TERNARY COMPOUND (CORDIERITE).

The temperatures in the field of the ternary compound are highest along the boundary toward sillimanite, but

FIG. 3.

there is no maximum within the field, therefore the composition of the crystals of the compound which begin separating near the point 3 must be between sillimanite and the maximum in the boundary curve 2-3, that is within the triangle III, IV, V of fig. 3. But the courses of the crystallization curves of Table 14a show that the composition must lie within about the limits IV, V, VI, VII. The simplest formulas that can be written for compositions within this quadrilateral are $\text{MgO} \cdot \text{Al}_2\text{O}_3 \cdot 3\text{SiO}_2$ and $2\text{MgO} \cdot 2\text{Al}_2\text{O}_3 \cdot 5\text{SiO}_2$.

If the crystals separating in the other parts of the field of the ternary compound have compositions thus represented then there should be a maximum on boundary curve 1-5. If the composition of the crystals of the ter-

nary compound at this maximum is near 1:1:3 the maximum should be easily located; on the other hand if near 2:2:5 it would be so near point 5 that it might not be definitely established. As already stated, melting temperatures gave no decisive information concerning such a maximum, therefore a supposedly favorable sample (MgO 25, Al₂O₃ 20, SiO₂ 55) was heated 18 hours at 1345° to 1355° to determine if forsterite would appear in recognizable form, as a means of settling the question of a maximum. No forsterite was detected. Thus 2:2:5 must be approximately the composition of the ternary compound separating near point 5. But before a more definite conclusion is arrived at the properties of the crystals of this compound separating in various parts of the field must be considered, and such properties compared with optical properties of crystalline aggregates obtained by crystallizing at lower temperatures glasses having the supposed composition of the compound.

Faceted crystals of cordierite are best obtained from melts in which it (or sillimanite) is the primary phase. Such crystals can be studied to best advantage when they occur in glass of considerable higher refractive index, *i. e.*, in melts containing less than 55 percent SiO₂. All the crystals observed, with the exception of those in one quench, were stout six-sided prisms with basal termination and negative elongation. The prism angle, within the limits of error of measurement, was 120°, and basal sections gave a nearly or quite uniaxial negative interference figure. In the one exceptional case several crystals were observed which gave unmistakable evidence of orthorhombic symmetry, although their general habit was just described. These crystals had the 120° prism and the side and basal pinacoids corresponding to the faces on all the other crystals; they had also the front pinacoid, and a second prism which bevels the edge between the side pinacoid and the unit prism.

The double refraction on the base is too small to give the orientation of β and γ but $\alpha \parallel c$. The refractive indices of crystals from melts highest in SiO₂ (66%) are $\alpha = 1.519$, β and $\gamma = 1.522$. From melts containing about 50% SiO₂ $\alpha = 1.524$, β and $\gamma = 1.528$. The differences are not much greater than the error of determination, ± 0.002 , but yet they may represent considerable

solid solution.⁷ The birefringence, $\gamma - \alpha$ as measured independently on several prisms is .003-.004.

The optical method of determining the composition of an unstable compound consists in obtaining from a glass of proper composition at a temperature below that of dissociation, an optically homogeneous crystalline aggregate having optical properties corresponding to the optical properties of crystals which are stable in the field of the compound. The desired aggregate should be obtainable, provided minute porosity does not develop during crystallization. In the case in hand we assume that there is no porosity, for there appears to be expansion instead of contraction during crystallization (the crystals have lower refraction than the glass). Glasses having the compositions MgO 11-13, Al₂O₃ 31-29, SiO₂ 57 crystallize to aggregates with a mean refractive index of $1.521 \pm .003$, that is, like crystals formed in the most siliceous melts within the field of the ternary compound. Likewise glasses of compositions MgO 13-15, Al₂O₃ 35-33, SiO₂ 52 give $n = 1.527 \pm .003$, like crystals from the least siliceous melts. The only simple ternary compounds approximating these compositions are MgO.Al₂O₃.3SiO₂ (MgO 12.4, Al₂O₃ 31.6, SiO₂ 56) and 2MgO.2Al₂O₃.5SiO₂ (MgO 13.7, Al₂O₃ 34.6, SiO₂ 51.7).

What were apparently very nearly homogeneous crystalline aggregates were prepared from glasses of a considerable range of composition extending on the composition diagram as a thin lens-shaped area, with its long axis the silica-spinel line, and one curved boundary nearly coinciding with the boundary curves 7-3-2. This area covers a range of silica content from about 45 to 70 per cent. In fact, if homogeneity of such aggregates had been the sole method of determining the composition of this unstable crystalline phase, the conclusion that it was a solid solution over a long range of compositions would have been reached.

There are three chief points to be considered in the interpretation of such aggregates: (1) the absolute sizes of grain of the crystals, (2) the shapes of the crystals, (3) the relative refractions of the crystals and possible glass. Obviously, whatever the other conditions, if the constituents have nearly the same refractive index, or if

⁷ Compare the case of spinel, *J. Am. Chem. Soc.*, **38**, 568, 1916.

the crystals have one dimension or more below microscopic size, the aggregate will appear homogeneous. The substance present in largest amount may crystallize in a meshwork which limits the size of the particles of the other substances. In such a texture diffusion would be so hindered that a very long heating might be required to allow the excess constituent to grow to appreciable size.

Aggregates in which the ternary compound was the chief constituent were prepared by heating for periods of 15 minutes to 5 hours at 900° to 1400°. While the glass was heating the μ , or unstable, form of the ternary compound began crystallizing at 900° or less as fine fibers which radiated from points on the surface of the grains. At temperatures of 925° to 1150° this form inverted to the α -form. The inversion in different portions of the same charge, and in different aggregates from glasses of the same composition, had an extreme temperature range of about 200°. This range is mostly accounted for in a lag in the starting of the inversion. When once started in a grain the inversion proceeded so rapidly that only in a few instances was a grain found, after quenching, to contain both forms. It is significant that the apparent homogeneity of aggregates containing the low temperature, or μ -form, covered a greater range of composition than that of the high temperature form. Finer grained and therefore more homogeneous-appearing aggregates would presumably be formed at the lower temperatures. The refractive indices of the aggregates decrease practically linearly with increase of silica. The average refractive index of clear aggregates containing the μ -form ranges from 1.535 at 68% SiO₂ to 1.560 at 40% SiO₂; and of aggregates of the α -form, 1.515 at 64% SiO₂ to 1.540 at 44% SiO₂.

The form appearing at the lower temperatures is called unstable because repeated attempts to produce it by heating the stable form at temperatures below those at which the stable form could be produced from the unstable met with no success. Thus there appears to be no true inversion point in this case, but the temperatures at which the rate of inversion becomes notable may be different for aggregates differing in silica content. An apparent rise of this temperature amounting to about 100° for an increase of 30 per cent of silica was observed.

Table 22 contains other details of the observations on these forms.

Thus the optical comparison of crystals from various parts of the cordierite field with crystal aggregates formed from glasses having compositions in which MgO , Al_2O_3 and SiO_2 range from 1:1:3 to 2:2:5, indicate that the possibility of solid solution of nearly this extent should be considered. Already it has been shown that the crystallization curves, the temperatures, and the optical properties of the crystals over about one-third of the cordierite field are satisfactorily explained by assuming the existence of a 2:2:5 compound. Solid solution over the range just indicated would account for the optical properties of the rest of the crystals. A diagrammatic representation of change of composition of crystals and course of crystallization curves in this case is shown in fig. 4. A case of more extensive changes is shown in fig. 5.

TABLE 22.

Quenchings showing the characteristics of aggregates of the α - and μ -ternary compound. ¹				
Composition	Weight	Per cent	Temp.	Phases
MgO	Al_2O_3	SiO_2		
17.5	42.5	40.0	915	μ
			925	$\mu + \text{MgO} \cdot \text{Al}_2\text{O}_3 + ?$
			1000	$\alpha + \text{Al}_2\text{O}_3 \cdot \text{SiO}_2 + ?$
16.0	40.0	44.0	925	$\mu + \alpha + ?$ (tr.)
			950	$\mu + \alpha + ?$ (tr.)
			975	$\alpha + \mu + \text{MgO} \cdot \text{Al}_2\text{O}_3$ (tr.)
14.5	40.0	45.5	950	μ
			1000	$\alpha + ?$
			1010	$\alpha + \text{Al}_2\text{O}_3 \cdot \text{SiO}_2 + ?$
			1025	Indefinable aggregate
15.3	38.8	45.9	1000	$\mu + \alpha$
			1050	$\mu + \alpha + \text{Al}_2\text{O}_3 \cdot \text{SiO}_2$
			1100	$\alpha + \text{Al}_2\text{O}_3 \cdot \text{SiO}_2 + ?$
14.5	36.5	49.0	950	μ
			1050	α
			1300	$\alpha + \text{Al}_2\text{O}_3 \cdot \text{SiO}_2 + ?$
14.4	35.6	50.0	950	μ
			1050	α
			1300	$\alpha + \text{Al}_2\text{O}_3 \cdot \text{SiO}_2 + \text{MgO} \cdot \text{Al}_2\text{O}_3 ?$
14.0	35.0	51.0	1450	α
			1460	Glass + $\text{Al}_2\text{O}_3 \cdot \text{SiO}_2$
16.0	33.0	51.0	1360	$\alpha + \text{MgO} \cdot \text{Al}_2\text{O}_3$
			1450	$\alpha + 10\%$ (?) glass
16.5	32.0	51.5	1430	$\alpha + 5\%$ (?) glass
15.0	32.0	53.0	1450	$\alpha + 5\%$ (?) glass
15.0	30.0	55.0	900	$\mu + \text{glass}$
			1000	$\mu + \alpha + ?$ (tr.)

¹ The $\text{MgO} \cdot \text{Al}_2\text{O}_3$ and other binary compounds reported here were usually in small amounts and in very small crystals, so that their identification was not always certain.

TABLE 22 (continued).

Quenchings showing the characteristics of aggregates of the α - and μ -ternary compound.

Composition MgO	Weight Al_2O_3	Per cent SiO_2	Temp.	Phases
			1100	$\alpha + ?$
			1350	$\alpha + MgO \cdot Al_2O_3 (?)$
12.4	31.6	56.0	1000	μ
			1100	α
			1425	α
			1435	$\alpha + tr. Al_2O_3 \cdot SiO_2$
15.0	27.0	58.0	950	Not homogeneous
			1100	Not homogeneous
10.0	30.0	60.0	1100	μ (a little cloudy)
			1170	$\mu + ?$
			1400	$\alpha + ?$
10.4	26.6	63.0	1100	μ
			1250	$\mu + \alpha + Al_2O_3 \cdot SiO_2$
			1350	$\alpha + Al_2O_3 \cdot SiO_2 + ?$
			1400	$\alpha + Al_2O_3 \cdot SiO_2 + SiO_2 (?)$
11.0	23.0	66.0	1400	$\alpha + ?$
			1425	$\alpha + ?$
9.	24.0	67.0	1400	$\alpha + ?$

FIG. 4.

FIG. 4. General relations as in fig. 6. In the area A-A-A-A crystals have composition A' and the crystallization curves are straight lines. In the other parts of the field, because of solid solution, the crystallization curves are not straight lines, and crystals at C, E, etc., have compositions C', E', etc. The maxima are between the arrows.

FIG. 5.

FIG. 5. General relations as in fig. 4 except that solid solution is more extensive.

THE CONCENTRATION-TEMPERATURE SOLID MODEL.

By interpolation from the foregoing melting temperatures together with those for the binary systems $\text{MgO-Al}_2\text{O}_3$, $\text{Al}_2\text{O}_3\text{-SiO}_2$, and MgO-SiO_2 , which were obtained from studies previously published, constant-temperature lines or isotherms may be drawn. This has been done for every 100° from 1400° to 1900° and the resulting

FIG. 6.

diagram is given in fig. 6, which shows moreover the melting temperature of each compound, component,⁸ quadruple and quintuple point. Those isotherms represented by full lines (isotherms 1400° - 1600°) are based on accurate data, those represented by dotted lines (isotherms 1700° - 1900°) are approximations based on data largely obtained from extrapolation on the high melting

⁸ The melting point of SiO_2 (cristobalite) is taken from recent unpublished determinations by J. B. Ferguson and H. E. Merwin.

boundary curves. A solid model corresponding to fig. 6 is shown in fig. 7, and the structure⁹ of the model before filling with plaster can be seen in fig. 8.

NATURAL CORDIERITE.

So little is known concerning natural cordierite that very definite comparisons between it and the ternary

FIG. 7.

compound just described can not be made. The ternary compound has been called cordierite for various reasons which appear in the text, but which need not be enumerated here. Possibly as much as 2 or 3 per cent of

⁹ For description see The Ternary System $CaO-Al_2O_3-SiO_2$, this Journal, 39, 16, 1915.

water may be present in certain unaltered cordierites,¹⁰ others may be nearly or quite anhydrous.¹¹ After the expulsion of water the residues of all the cordierites analyzed approach very closely the formula $2(\text{MgO} \cdot \text{FeO}) \cdot 2\text{Al}_2\text{O}_3 \cdot 5\text{SiO}_2$. A hydrous iron-bearing cordierite after heating 30 minutes at 1440° consisted of glass and

FIG. 8.

sillimanite. Dehydration for 5 minutes at 600° caused a decrease of .010 in the average refractive index of the same material. Probably the escape of water was not accompanied by corresponding shrinkage, and the liquid used to immerse the grains for the refractive index

¹⁰ See "Untersuchungen an Cordierite." Leopold Oppenheimer, Vehr. Heidelb. Nat. Med. Ver., N. F. 13, 257-302, 1914.

¹¹ H. Bücking, Neues Jahrb. Min. Geol., 1901, I, 72 (ref.).

determination did not at once penetrate the pores.¹² Such behavior has prevented the determination of a possible inversion in cordierite corresponding to that of the μ -form of the ternary compound into the α -form.

FeO IN MAGNESIAN MINERALS AND ROCKS.

The magnesia compounds of this ternary system are represented among rock-forming minerals by compounds in which ferrous oxide plays the role of magnesia to a considerable extent. Although it is probable that FeO can thus replace MgO without causing the formation of new phases, yet the temperature relations must be so different that fig. 1 cannot represent the shapes of the fields of these phases. It is conceivable—but not probable unless FeO largely predominates—that the field of cordierite might disappear, and that the field of corundum might approach or even border the field of olivine, but even then corundum would not be expected as an early phase in the crystallization of a dry magma near olivine in composition.

SUMMARY.

The temperature-concentration relations of the various crystalline phases in equilibrium with liquid in the ternary system MgO-Al₂O₃-SiO₂ have been investigated, and are represented by diagrams and a model. A ternary compound, 2MgO.2Al₂O₃.5SiO₂, unstable at its melting point and considerably affected by solid solution has been observed in two forms. The μ -form (unstable) crystallizes from glass at temperatures below about 950°, and transforms at somewhat higher temperatures into the α -form (stable). The properties of both forms, but especially the α -form, are close to those of the mineral cordierite. The effects of solid solution in flattening two of the boundary curves of the field of this compound are discussed.

The preparation and properties of crystalline aggregates of substances which disassociate when melting are considered.

Crystals of natural cordierite disassociate when melting. Brief consideration is given to the effects of FeO in magnesian rocks and minerals.

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¹² The progressive refilling of pores of crystals from which volatile matter has been expelled can often be observed thus in a viscous liquid under the microscope.