

AFM LIQUIDUS PROJECTIONS FOR GRANITIC MAGMAS, WITH SPECIAL REFERENCE TO HORNBLLENDE, BIOTITE AND GARNET

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ABSTRACT

The liquidus relationships projected in the AFM system ($A = Al_2O_3-K_2O-Na_2O-CaO$, $F = FeO$, $M = MgO$) have been described by Abbott & Clarke (1979) for silicate liquids (Liq) saturated with respect to quartz, alkali feldspar, plagioclase and one or more of the AFM minerals, Als (andalusite or sillimanite), garnet (Gar), biotite (Bio) and cordierite (Cdt). Three improvements are presented in this paper: (1) The liquidus relationships in part of the metaluminous region ($A < 0$) are deduced by considering the equilibrium $Liq-Bio-Hnb$ ($Hnb = hornblende$). The liquidus boundary for this equilibrium is believed to extend from the metaluminous region, where the reaction is even ($Hnb + Bio = Liq$), into the peraluminous region, where it is odd ($Bio = Hnb + Liq$). Liquids on the $Hnb-Bio-Liq$ equilibrium may change from metaluminous to peraluminous during normal fractional crystallization. (2) A plausible sequence of changes in the AFM liquidus topology is presented for the disappearance of a liquidus field for FeBio from the AF join. The breakdown of FeBio results in the appearance of a liquidus field for fayalite. (3) The effects of adding MnO to the AFM system are examined. Whereas at low $Mn/(Fe+Mg+Mn)$ the equilibrium $Bio-Gar-Liq$ is interpreted to be even ($Bio+Gar = Liq$), at high $Mn/(Fe+Mg+Mn)$ this equilibrium is believed to be odd ($Gar = Bio+Liq$). This behavior may account for the disappearance of Bio during the final stages of fractional crystallization, leading to garnetiferous aplites and pegmatites.

Keywords: granite, peraluminous, AFM, magma genesis.

SOMMAIRE

Les relations projetées sur le liquidus dans le système AFM ($A = Al_2O_3-K_2O-Na_2O-CaO$, $F = FeO$, $M = MgO$) ont été décrites (Abbott & Clarke 1979) pour liquides silicatés (Liq) saturés en quartz, feldspath alcalin, plagioclase et au moins une des phases du système AFM: Als (andalusite ou sillimanite), Gar (grenat), Bio (biotite) et cordiérite (Cdt). Nous apportons ici trois précisions. 1. On peut déduire les relations sur le liquidus de la partie méta-alumineuse du système ($A < 0$) en considérant l'équilibre Liq, Bio et

Hnb (hornblende). L'équation d'équilibre s'appliquerait de la région méta-alumineuse, où elle est d'ordre pair ($Hnb + Bio = Liq$), jusqu'à la région hyperalumineuse, où elle est impaire ($Bio = Hnb + Liq$). Les liquides impliqués changeraient donc de méta-alumineux à hyperalumineux pendant une cristallisation fractionnée normale. 2. Une séquence vraisemblable dans la topologie du liquidus AFM vise à expliquer la disparition du champ de stabilité de FeBio sur le liquidus à partir du côté AF du système. La décomposition de FeBio est à l'origine de la présence de la fayalite sur le liquidus. 3. L'addition de MnO au système semble transformer la réaction d'équilibre $Bio-Gar-Liq$ de paire ($Bio + Gar = Liq$) aux faibles teneurs de Mn, à impaire ($Gar = Bio + Liq$) pour un rapport $Mn/(Fe + Mg + Mn)$ élevé. Ce comportement expliquerait l'élimination de la biotite lors des stades terminaux de cristallisation fractionnée, et la formation d'aprites et de pegmatites à grenat.

(Traduit par la Rédaction)

Mots-clés: granite, peralumineux, AFM, pétrogenèse magmatique.

INTRODUCTION

The hypothetical liquidus relationships for the AFM system ($A = Al_2O_3-K_2O-Na_2O-CaO$, $F = FeO$, $M = MgO$; Thompson 1957) projected from quartz, alkali feldspar and plagioclase have been presented by Abbott & Clarke (1979). The liquidus relationships show the projected loci of silicate liquids coexisting with two or more of the AFM minerals biotite (Bio), garnet (Gar), cordierite (Cdt) and Als (andalusite or sillimanite), where the silicate liquid is saturated with respect to quartz, alkali feldspar and plagioclase.

The liquidus relationships for various activities of H_2O were proposed on the following theoretical basis: (1) For a given activity of water $a(H_2O)$, the temperature of the liquid coexisting with quartz, two feldspars (an alkali feldspar and oligoclase or andesine) and one or more of the AFM minerals is only slightly lower than the temperature of the liquid coexisting with just quartz and the same two

feldspars but no AFM minerals (Abbott & Clarke 1979). (2) Where a silicate liquid becomes possible, the subsolidus AFM mineral assemblages may be treated like alkemade relationships so that, for each two-phase subsolidus AFM mineral assemblage, there is a liquidus boundary marking the locus of projected liquids coexisting with the two AFM minerals. The three-phase equilibria (Liq + 2 AFM minerals) may be odd (peritectic) or even (cotectic) or partly odd and partly even, using the terminology of Ricci (1951). For each three-phase subsolidus AFM assemblage, there is a point on the liquidus surface representing the projected composition of the liquid coexisting with three AFM minerals. The four-phase equilibria (Liq + 3 AFM minerals) may be odd or even as well.

Whether the various liquidus equilibria are odd or even depends on the composition of the liquid relative to the coexisting AFM minerals. The odd-even ambiguity can be removed by taking into account a small number of assumptions discussed in Abbott & Clarke (1979): (1) There is never more than one liquidus minimum in the AFM projection, *i.e.*, during ideal fractional crystallization all liquids proceed to the same liquidus minimum. (2) The ratio F/M is higher in the silicate liquid than in the coexisting AFM mineral assemblage. This may not always be true, especially for liquids coexisting with garnet (Green 1977, Miller *et al.* 1981). (3) For any given set of conditions in P and $a(\text{H}_2\text{O})$, the composition of the liquid having the lowest temperature (projected position of the liquidus minimum) has a high ratio of F/A. Other assumptions listed in Abbott & Clarke (1979) pertain to the appearance and disappearance of liquidus fields for garnet and cordierite, and need not be restated here.

In this paper, three improvements are presented. The first involves relationships in the metaluminous region ($A = \text{Al}_2\text{O}_3 - \text{K}_2\text{O} - \text{Na}_2\text{O} - \text{CaO} < 0$) and the existence of a liquidus field for hornblende. The second improvement involves the breakdown of biotite at low $a(\text{H}_2\text{O})$ and high T and the disappearance of a liquidus field for biotite from the AF join. The third improvement involves the effects of adding MnO to the system, particularly the disappearance of biotite and subsequent appearance of garnet in late aplites and pegmatites owing to partition of MnO into the silicate liquid.

The discussion is restricted to temperatures outside the stability field for muscovite; thus, muscovite-bearing assemblages are not considered. The stability limit for muscovite used in this paper (*e.g.*, in Fig. 3) is from Thompson

& Algor (1977). All reactions are written in terms of the AFM minerals only.

HORNBLENDE

The association of hornblende and biotite is common in many granitic intrusions. In a series of comagmatic intrusions related by differentiation, the biotite almost always appears later than the hornblende, in agreement with the reaction series of Bowen (1928). Certainly the sequence hornblende, biotite + hornblende, biotite in differentiated granites is sufficiently common that there is little need here for thorough documentation.

The Maine coastal plutons (Chapman 1962) are a set of circular to elliptical composite granite bodies (in most cases). The plutons are Devonian, post-Acadian orogeny. The earliest intrusions are commonly hornblende granite, succeeded by comagmatic hornblende-biotite-bearing or biotite-bearing granite (Chapman 1974, Karner 1968, Wones 1974). At some stage during crystallization, hornblende stopped forming, so that in the latest intrusions biotite is the only ferromagnesian silicate mineral. In the Red Beach granite (Abbott 1977, 1978, Amos 1963), the transition from early hornblende granophyre to later biotite granite may be due in part to assimilation of aluminous sediments, but this is still not satisfactorily documented. In other Maine coastal plutons, the latest biotite granites are situated near the centres of the plutons, farthest from the margins of the plutons, hence, farthest from any possible influence by wall-rock contamination. Because the transition from hornblende to biotite occurs in a number of bodies that intrude a wide range of older country rocks, the hornblende-biotite transition is believed to be the result of normal differentiation by crystal fractionation.

In the Red Beach granite (Abbott 1977), biotite coexisting with hornblende in the early intrusions and biotite in the later intrusions are distinctly peraluminous, $\text{Al}_2\text{O}_3 / (\text{CaO} + \text{Na}_2\text{O} + \text{K}_2\text{O}) = 1.27-1.43$, whereas the hornblende is metaluminous, $\text{Al}_2\text{O}_3 / (\text{CaO} + \text{Na}_2\text{O} + \text{K}_2\text{O}) = 0.27-0.32$. Evidently, by crystallizing hornblende, successively younger liquids became more aluminous; in fact, the latest liquids were slightly peraluminous.

Figure 1a shows the compositional ranges possible for biotites and hornblendes projected onto the AFM diagram. The corresponding liquidus equilibrium (Liq-Hnb-Bio) is shown in Figure 1b. The hornblende liquidus field is

shown extending to more aluminous compositions than the least aluminous biotites. The reaction on the low-temperature end of the liquidus boundary is believed to be odd:



This reaction is consistent with the observed crystallization sequence hornblende, biotite + hornblende (on the liquidus boundary), biotite alone (as the result of reaction 1). Cawthorn & Brown (1976) have suggested a similar behavior to explain the transition from metaluminous to peraluminous liquids.

Granite-melting experiments done by Gibbon & Wyllie (1969) provide additional support for reaction 1. When samples of biotite granite (sample FC-C, containing no hornblende initially) from the Farrington complex (North Carolina) were melted, hornblende appeared in charges with as little as 10% melting. This demonstrates that at least under some conditions biotite melts incongruently to hornblende plus liquid.

In the southern Appalachian Piedmont there are a number of hornblende-biotite granitic plutons (Speer *et al.* 1980). In some cases, the biotite appears to have started crystallizing before the hornblende; in other cases, the hornblende preceded the biotite. Speer *et al.* (1980) suggested, on the basis of experimental work by Wones & Dodge (1977), that the order of crystallization of biotite and hornblende depends on the activity of H_2O . In the present context (Fig. 1), the order of crystallization (hornblende followed by biotite or biotite followed by hornblende) depends on the projected AFM composition of the liquid. In the metaluminous region, the initial liquid may lie on either side of the biotite-hornblende liquidus boundary. If initially the liquid is in the primary liquidus field for hornblende, hornblende will be followed by biotite; if the liquid is in the primary liquidus field for biotite, biotite will be followed by hornblende. Once on the two-phase liquidus boundary, the liquid will change composition along the boundary and may become peraluminous where a portion of the two-phase boundary is odd. Under these circumstances, there could be two periods during which biotite would be the only AFM mineral crystallizing. The sequence of crystallization might be biotite (in the metaluminous region), biotite + hornblende (on the two-phase boundary), biotite alone (in the peraluminous region as the result of reaction 1). The writer is not aware of any granitic intrusions showing this sequence of crystallization.

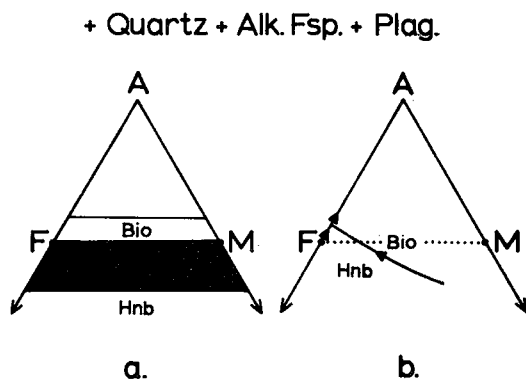
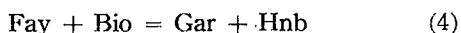
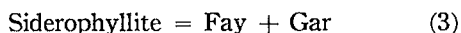
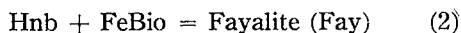


FIG. 1. (a) Schematic phase relationships between hornblende (Hnb) and biotite (Bio) in the AFM projection. (b) Projected liquidus equilibria involving biotite and hornblende. The arrows on the two-phase liquidus boundary (Liq-Hnb-Bio) and on the AF join indicate the direction of falling temperature. The high-temperature end of the equilibrium Liq-Hnb-Bio is even ($\text{Hnb} + \text{Bio} = \text{Liq}$); the low-temperature end is odd ($\text{Bio} = \text{Hnb} + \text{Liq}$).

The biotite-hornblende liquidus boundary would be expected to move in response to changing $a(\text{H}_2\text{O})$. Increasing $a(\text{H}_2\text{O})$ should result in expansion of the primary liquidus field for the more hydrated phase, biotite. Hence, the two-phase liquidus boundary should shift away from A with increasing $a(\text{H}_2\text{O})$ in Figure 2. A metaluminous liquid, which at low $a(\text{H}_2\text{O})$ is in the primary field for hornblende, could, at a higher $a(\text{H}_2\text{O})$, be in the primary field for biotite. Of course, this would determine the order of crystallization of biotite and hornblende.

BIOTITE

Figures 2 and 3 show the presumed phase relationships for the disappearance of a liquidus field for biotite from the AF join, $a(\text{H}_2\text{O}) \leq 1$. In order of increasing temperature, the proposed subsolidus equilibria are as follows, assuming almandine to be stable:



The nature of the breakdown reactions for

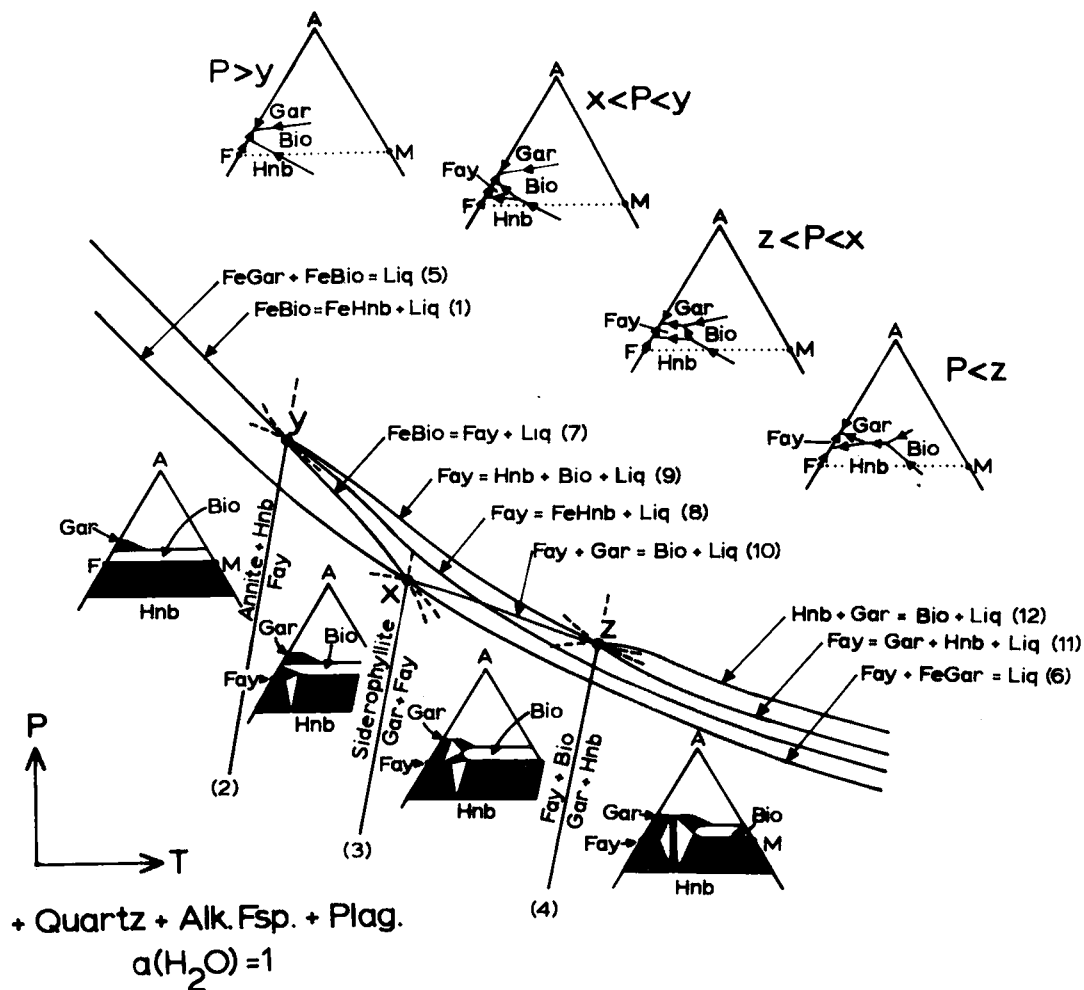


FIG. 2. Schematic phase relationships involving biotite (Bio), fayalite (Fay), garnet (Gar), hornblende (Hnb) and liquid (Liq) for $a(\text{H}_2\text{O}) = 1$. Four distinct subsolidus AFM topologies correspond, respectively, to four distinct AFM liquidus topologies. On the AFM liquidus diagrams, the arrows indicate falling temperature on the two-phase liquidus boundaries and on the binary AF join.

biotite and positions of the reactions in P - T space are strongly dependent on the fugacity of oxygen (Eugster & Wones 1962). It is assumed here that the fugacity of oxygen is sufficiently low that Fe_2O_3 is not present. Eugster & Wones (1962) have shown that annite, $\text{KFe}_3\text{AlSi}_3\text{O}_{10}(\text{OH})_2$, breaks down in the presence of quartz to sanidine, fayalite and an aqueous vapor phase at temperatures as high as 700°C at $P = 2020$ bars. Reaction 2 involves hornblende because, as noted earlier, the biotite coexisting with hornblende is slightly more aluminous than a mica on the annite-phlogopite join. Biotite coexisting with garnet contains an appreciable siderophyllite-eastonite component.

$\text{K}(\text{Fe},\text{Mg})_{2.5}\text{Al}_{0.5}(\text{Si}_{2.5}\text{Al}_{1.5})\text{O}_{10}(\text{OH})_2$. Siderophyllite is assumed to be the most refractory of the Fe end-member biotites, breaking down according to reaction 3. The temperature difference between reactions 2 and 3 is believed to be small. Both reactions are shown in Figure 3 within the range of conditions suggested by Fyfe (1969) for the breakdown of biotite. The three-phase assemblage Fay-Gar-Bio, which is possible at higher temperatures than reaction 3, has been observed in granulite-facies metamorphosed pelitic rocks (Korzzhinskii 1936).

Reaction 4 was located arbitrarily in Figure 3 at a higher temperature than reaction 3. The assemblage Gar-Bio-Hnb, which is possible at

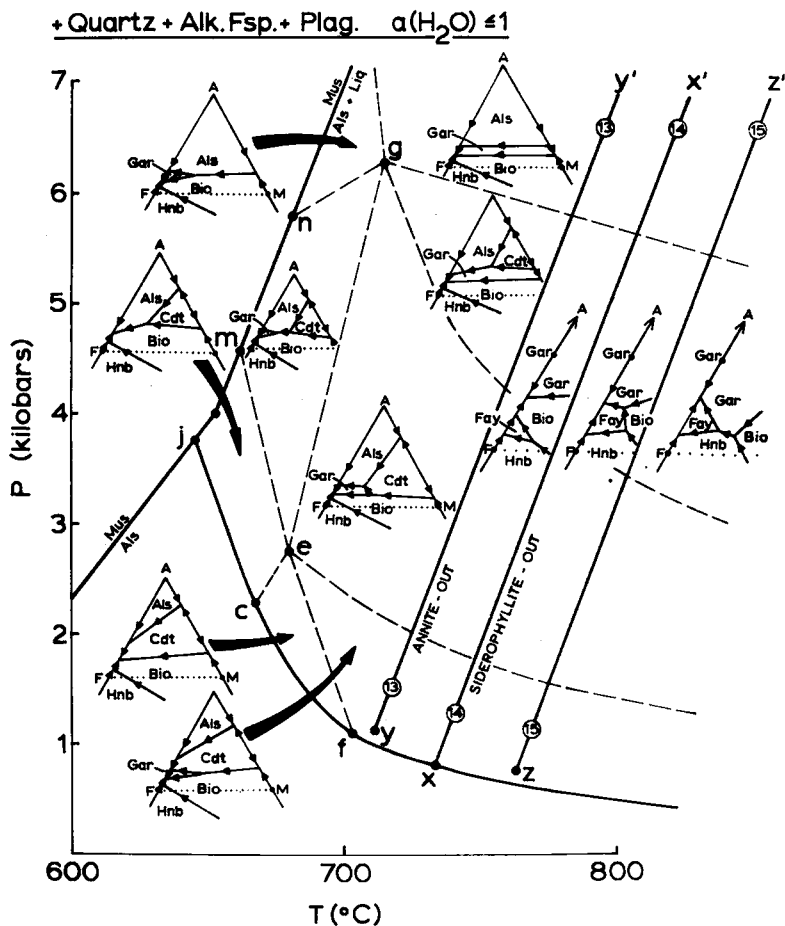
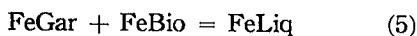


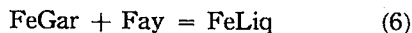
FIG. 3. The invariant points x , y and z at $a(\text{H}_2\text{O}) = 1$ (Fig. 2) are shifted to higher pressures as $a(\text{H}_2\text{O})$ is decreased, defining the three loci xx' , yy' and zz' . These lines (xx' , yy' and zz') divide P - T - $a(\text{H}_2\text{O})$ space into four regions for liquids coexisting with quartz, alkali feldspar, plagioclase and one or more of the AFM minerals garnet, biotite, hornblende and fayalite. Each region is characterized by a distinct AFM liquidus topology. Also shown are the AFM liquidus relationships involving garnet, biotite, cordierite and Als (andalusite or sillimanite) from Abbott & Clarke (1979). Reactions involving muscovite (Mus) are from Thompson & Algor (1977).

higher temperatures than reaction 4, has been observed by Ferry (1979) in granites from central Maine.

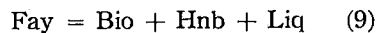
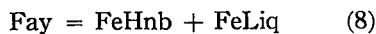
The three subsolidus reactions (2, 3, 4) meet the solidus at three invariant points, x , y and z (Figs. 2, 3). If $P > x$, the liquidus minimum reaction is:

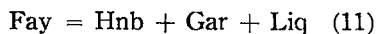
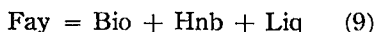


If $P < x$, the liquidus minimum reaction is:



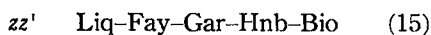
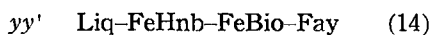
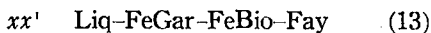
Various other binary AF liquidus reactions and ternary AFM liquidus reactions are encountered at higher temperatures, depending on P :





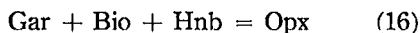
The liquidus reactions were determined according to the assumptions cited earlier and the rules of Schreinemakers.

As $a(\text{H}_2\text{O})$ decreases from one, the points x , y and z describe the loci xx' , yy' and zz' (Fig. 3), assumed to be approximately parallel to the breakdown reaction for muscovite (from Thompson & Algor 1977). The lines xx' , yy' and zz' correspond to the following equilibrium AFM assemblages,



which divide the P-T space into four regions. Each region has a characteristic AFM liquidus topology for liquids coexisting with one or more of the minerals Gar, Fay, Hnb and Bio. Liquidus reactions involving Als, Cdt, Bio and Gar are also shown (Abbott & Clarke 1979). It should be noted that the positions for xx' , yy' and zz' (equilibria 13, 14 and 15) are strongly influenced by the fugacity of oxygen.

At some higher temperature than zz' , a liquidus field for orthopyroxene will appear in conjunction with the appearance of orthopyroxene in the subsolidus region. For instance, one possibility is that orthopyroxene may appear according to the following reaction:



Ferry (1979) reported the assemblage Gar-Bio-Hnb in Siluro-Devonian granite stocks of central Maine. The crystallization pressure was calculated to be approximately $P(\text{total}) = 3500$ bars (Ferry 1978). At this pressure the assemblages is consistent with a comparatively high low $a(\text{H}_2\text{O})$. Ferry (1978) also reported the assemblage Bio-Gar-Als (sillimanite) from apparently more differentiated granites in the same area. At 3500 bars pressure, this assemblage is consistent with a comparatively high $a(\text{H}_2\text{O})$, in the region *emng* of Figure 3 (see

Abbott & Clarke 1979). The change in the AFM assemblage reflects a progressive increase in $a(\text{H}_2\text{O})$ during crystallization.

EFFECTS OF MnO

The very latest intrusions in many granite plutons commonly contain spessartine-rich garnet as the only ferromagnesian AFM mineral (Cawthorn & Brown 1976). Garnets in aplites and pegmatites from the Shelburne batholith, southern Nova Scotia, are essentially almandine-spessartine ($\text{Alm}_{35}\text{Spe}_{42}\text{Py}_{15}$), as determined by microprobe analyses. In the pegmatites and aplites in which garnet is the only ferromagnesian AFM mineral, the bulk ratio of $\text{Mn}/(\text{Fe} + \text{Mn} + \text{Mg})$ is high, essentially equal to the ratio for the garnet, about 0.4. In the Shelburne batholith and also in the Spruce Pine district of North Carolina, the spessartine-rich garnet-bearing aplites and pegmatites were preceded by biotite-garnet granites. In the Shelburne batholith, garnet coexisting with biotite has approximately the same composition as the garnet occurring alone in the late pegmatites and aplites. However, the bulk ratio of $\text{Mn}/(\text{Fe} + \text{Mn} + \text{Mg})$ must be lower in the early granites because biotite contains only minor MnO. Evidently, differentiation from biotite-garnet granite to garnet-bearing, biotite-free pegmatites and aplites proceeds with increasing $\text{Mn}/(\text{Fe} + \text{Mn} + \text{Mg})$ in the silicate liquid.

Hall (1965) cited enrichment in MnO as the cause for the absence of biotite and appearance of spessartine-rich garnet in the latest intrusions associated with the Donegal granites of northern Ireland. Cawthorn & Brown (1976) and Miller & Stoddard (1978) have suggested that biotite is eliminated because the equilibrium Gar-Bio-Liq is odd:



The author can find no obvious *textural* evidence for this reaction in those biotite-garnet granites in which the bulk ratio of $\text{Mn}/(\text{Fe} + \text{Mn} + \text{Mg})$ is low. However, when the ratio of $\text{Mn}/(\text{Fe} + \text{Mn} + \text{Mg})$ is high, the absence of biotite in late-stage intrusions makes it very difficult to deny reaction 17.

The probable effects of increasing $\text{Mn}/(\text{Fe} + \text{Mn} + \text{Mg})$ are shown schematically in Figure 4. Where $\text{Mn}/(\text{Fe} + \text{Mn} + \text{Mg})$ is low, the equilibrium Gar-Bio-Liq is interpreted to be even, as suggested by Abbott & Clarke (1979). At high $\text{Mn}/(\text{Fe} + \text{Mn} + \text{Mg})$, this equilibrium is interpreted to be odd (reaction 17).

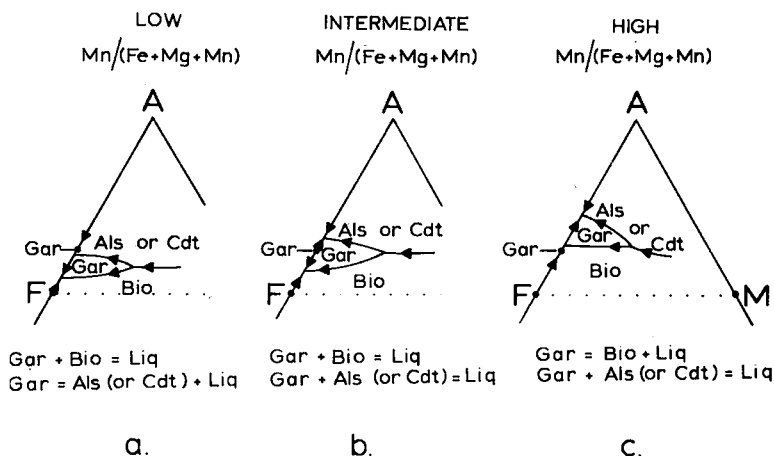


FIG. 4. Hypothetical changes in the AFM liquidus topology as a function of the ratio $Mn/(Fe+Mg+Mn)$. At low $Mn/(Fe+Mg+Mn)$, the equilibrium Liq–Bio–Gar is believed to be even ($Gar+Bio = Liq$); at high $Mn/(Fe+Mg+Mn)$, this equilibrium is believed to be odd ($Gar = Bio+Liq$).

DISCUSSION

The hypothetical liquidus relationships in the AFM projection presented in Abbott & Clarke (1979) and in this paper represent a *petrogenetic grid for silicate liquids saturated with respect to quartz, alkali feldspar, plagioclase and one or more of the AFM minerals*. Originally (Abbott & Clarke 1979), the model was conceived for peraluminous compositions only. In this paper the model has been extended to include metaluminous compositions by considering the liquidus field for hornblende. The model has also been extended by taking into account the breakdown of biotite and the possible effects of increasing $Mn/(Fe+Mn+Mg)$. It should be emphasized that the various liquidus equilibria are strongly influenced by the fugacity of oxygen. It has been assumed throughout that the fugacity of oxygen is sufficiently low that an Fe_2O_3 component need not be considered.

The appearance and disappearance of different AFM minerals during the course of fractional crystallization take place according to the well-recognized principles governing ternary liquidus equilibria as long as the liquid is saturated with respect to quartz, alkali feldspar and plagioclase. One- and two-phase AFM mineral assemblages are common in granitic rocks and correspond, respectively, to crystallization in one-mineral AFM liquidus fields ($Liq + 1$ AFM mineral) or on two-mineral AFM liquidus boundaries ($Liq + 2$ AFM minerals). Three-

phase AFM mineral assemblages are less common and correspond to various possible peritectic points on the AFM liquidus surface. An AFM mineral may be resorbed or simply stop growing during the course of fractional crystallization whenever the liquid encounters a two-mineral liquidus boundary ($Liq + 2$ AFM minerals) that is odd, or a three-mineral liquidus reaction point ($Liq + 3$ AFM minerals) that is odd. Hornblende can be eliminated as the result of reaction 1. If the ratio of $Mn/(Fe+Mn+Mg)$ is high, biotite can be eliminated by reaction 17. Other examples are described in Abbott & Clarke (1979).

Systematic changes in the AFM mineral assemblage in differentiated intrusions reflect normal crystal–liquid equilibria, are characteristic of specific AFM liquidus topologies (Fig. 3), and hence are characteristic of the conditions in P , T , $a(H_2O)$ and other factors.

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