SHORTER COMMUNICATIONS

THE BIOTITE-CORDIERITE-ALMANDITE SUBFACIES OF THE HORNBLENDE-GRANULITE FACIES: A DISCUSSION.*

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De Waard has proposed several subdivisions of the granulite facies of regional metamorphism. The latest of these—the biotite-cordierite-almandite subfacies (de Waard, 1966)—is based in part on the valuable work of Wynne-Edwards and Hay (1963).

De Waard's technique is to interpret naturally occurring assemblages in terms of (a) intensive physical variables, and (b) hypothetical reactions. The purpose of this note is to show to what extent his methods are arbitrary and questionable.

It is necessary at the outset to say a word about terminology. Following a common practice, de Waard uses the ambiguous term P_{H_2O} (P_w in de Waard 1965a) without defining his usage. Two definitions he might have employed are:

1. the partial pressure of a component in a mixture of gases occupying a definite volume, is that pressure the component would exert if it alone filled the volume (e.g. Prutton & Marron, 1951, p. 15);
2. the partial pressure of a component is the product of total gas pressure and the mole fraction of that component in the gas mixture, (e.g. Lewis & Randall, 1961, p. 214).

Only for ideal gases are these equivalent (Denbigh, 1966, p. 116), hence the ambiguity.

The notation used in this critique is:

T	temperature
V	volume
S	entropy
P_{H_2O}	in direct quotation or paraphrase of de Waard, precise meaning unknown
P_{load}	de Waard's term, taken to be equivalent to P_s
P_s, $P_{E_{H_2O}}$	Greenwood's usage (1961, p. 3924).

THE PHYSICAL VARIABLES

De Waard's choice of T, P_{load} and P_{H_2O} as definitive variables, is bound to be an oversimplification in any system containing iron as a

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component. To ignore the possible effect of oxygen fugacity on stability fields of assemblages in a system of this kind, is tantamount to treating the hypothetical reactions as being of simple dehydration type. In the absence of experimental data this may be justified as a rough, first approximation; but no justification seems possible for de Waard's subsequent treatment of his chosen variables.

For example, apropos his reaction 1 de Waard (1966, p. 483) states:

> "This condition [the coexistence of sillimanite-cordierite-almandite-biotite-orthoclase-quartz] . . . . may be considered to reflect univariant equilibrium for an open system, or divariant equilibrium for a closed, H_2O deficient system."

In other words the variance of a closed system is claimed to be greater than that of the equivalent[1] open system. This is wrong, for if the closed system were indeed divariant, then an equivalent open system would have a variance of $m + 2$, where m is the number of perfectly mobile components (see for example Thompson, 1955, p. 66).

It is possible that by 'open' de Waard meant to indicate that a separate aqueous phase was present i.e. in contradistinction to Yoder's term H_2O-deficient. In this way he could get his required univariancy, but only provided the system were closed.

Further observations are most readily made graphically. A simple dehydration reaction, independent of variations in oxygen fugacity, of the general form:

$$\text{hydrous phase} \pm \text{other phases} = \text{anhydrous phase(s)} + H_2O$$

can be represented as a three dimensional surface, having P_s, $P_{E_{H_2O}}$ and T as rectangular coordinates. In the idiom of metamorphic petrology the assemblages stable on the low temperature side of the surface may be called low grade, while those stable on the high temperature side may be referred to as high grade.

Greenwood (1961, figure 1a) gives a diagram of the complete surface. In figure 1a (this paper) it has been projected onto the P_s-T plane and contoured with respect to $P_{E_{H_2O}}$. In figure 1b (this paper) the surface has been projected onto the P_s-$P_{E_{H_2O}}$ plane and contoured with respect to T.

The slope of the contours in figure 1a is given by

$$\left(\frac{\partial P}{\partial T}\right)_{P_{E_{H_2O}}} = \frac{\Delta S}{\Delta Vs}$$

For the type of reaction under consideration, this will in general be negative (Greenwood, 1961, p. 3926), so that it will be possible to obtain a high grade assemblage from a low grade one by both increasing P_s or

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[1] Equivalent in having the same total number of components, and the same coexisting phases.

by increasing T, if $P_{E_{H_2O}}$ remains constant. This condition in itself is unlikely—implying as it does a situation "wherein the rock pressure is variable but the fluid pressure remains constant" (Thompson, 1955, p. 85). However, it is highly likely that a dehydration curve for realistic conditions within the earth's crust, will have dP_s/dT negative, and the constant $P_{E_{H_2O}}$ contours of figure 1a represent a virtual limit for such a curve (see figure 2 in Thompson, 1955, p. 90). Therefore, for a generalized dehydration reaction taking place in the earth's crust, and marking a boundary between subfacies, it is unlikely to be true that

> "the transition from one subfacies to another may thus be produced under similar P conditions by a change of T in the opposite sense from a change of P" (de Waard, 1966, p. 487).

Consider now figure 1b (this paper), which has a direct bearing on figure 2 (de Waard, 1966), figure 1 (de Waard, 1965a) and figure 12 (de Waard 1965b). The last two mentioned are described as being "essentially isothermal" so that this is presumably true of the first of the three also though its caption is confusing on this point.

If de Waard's diagrams are in fact taken to be isothermal, then his isograds are comparable to the isotherms of figure 1b. The slope of these is given by:

$$\left(\frac{\partial P_{E_{H_2O}}}{\partial P_s}\right)_T = -\frac{\Delta V_s}{\Delta V_{H_2O}}$$

which in general will be positive so that the isotherm will be concave towards the $P_{E_{H_2O}}$ axis (see Thompson, 1955, p. 84). In other words de

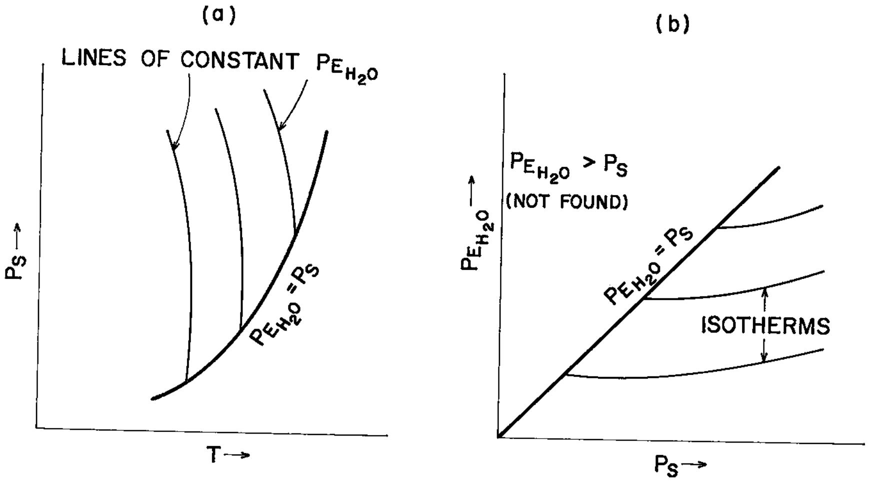

FIG. 1. Projections of a dehydration reaction surface, (slightly modified after Greenwood, 1961, fig. 1).

Waard's isograds should also be concave towards his P_{H_2O} axis and not orthogonal as he shows them.

It is not clear what part of de Waard's diagrams, if any corresponds to the naturally unknown part of figure 1b. This region can be avoided of course, by choosing an origin within the naturally feasible part, though some note should be made to this effect. As they stand de Waard's diagrams suggest in a qualitative way, that it is possible to have P_{H_2O} much higher than P_{load} in granulite facies terrain.

The Hypothetical Reactions

De Waard (1966) formulates five reactions, three explicitly and two in a vaguer manner. Consider first reaction 1:

$$10K_2(Fe,Mg)_{5\frac{1}{2}}Al_3Si_{5\frac{1}{2}}O_{20}(OH)_4 + 28Al_2SiO_5 + 65SiO_2 =$$

$$11(Fe,Mg)_2Al_4Si_5O_{18} + 11(Fe,Mg)_3Al_2Si_3O_{12} + 20KAlSi_3O_8 + 20H_2O$$

This may properly be called hypothetical, since no evidence, petrographical, experimental or otherwise, is advanced in its support.

According to de Waard (1966, pp. 485–486):

> "the left hand side of the reaction represents the almandite-amphibolite facies, characterised by coexisting biotite and sillimanite . . . The right hand side represents the anhydrous assemblage of the pyroxene-granulite facies".

Again:

> "the combination of reactants and products of the reaction characterize the hornblende-granulite facies", (p. 486).

The last quotation resembles similar remarks concerning other reactions made by de Waard elsewhere (e.g. 1965a, p. 456, p. 457; 1965b, p. 167). It is examined here in the light of a P_s-T projection of the reaction curve drawn for de Waard's implicit assumption that fugacity of oxygen is not a significant variable, (figure 2).

The unbroken curve is what is generally referred to as 'the upper stability curve'. The broken line is one of the infinite number of curves that can be drawn for the condition $P_{E_{H_2O}}$ = a constant (e.g. the contours of figure 1a, which have already been discussed). The next paragraph applies to either type of curve.

Figure 2 shows all the divariant six phase assemblages stable on high grade and low grade sides of the reaction curve. A significant feature is that all of these assemblages involve the coexistence of "reactants and products" of reaction so that this criterion cannot be used to recognize a subfacies intermediate between the high grade and low grade regions of

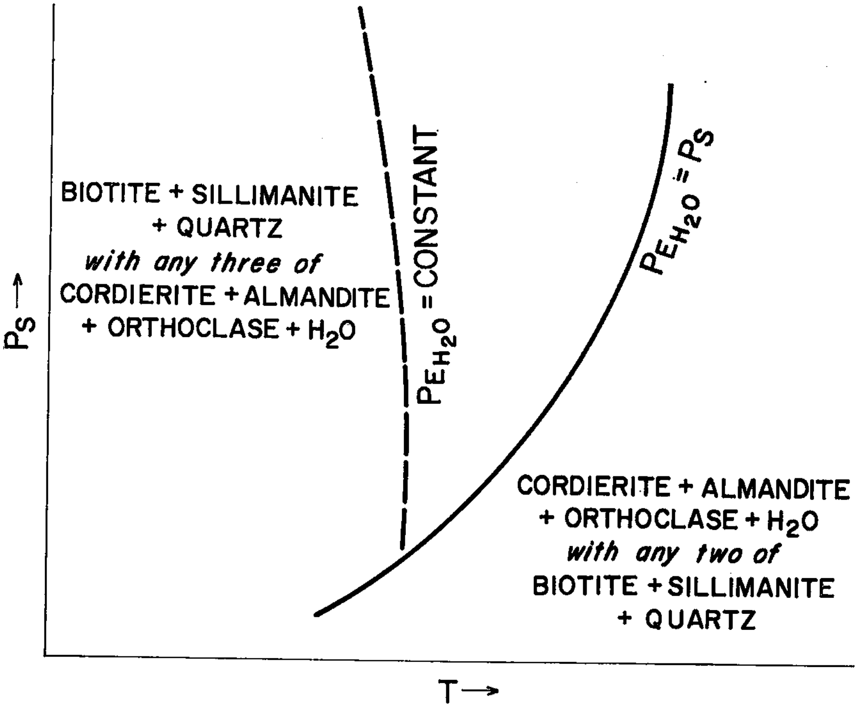

FIG. 2. P_s–T projection of de Waard's hypothetical reaction 1.

figure 2. In other words, if we take the last quotation from de Waard literally, we would have to consider the whole of the P_s-T field of figure 2 as belonging to his hornblende granulite facies.

A further point can be made with reference to reaction 3 (de Waard, 1966):

$$NaCa_2(Fe,Mg)_4Al_3Si_6O_{22}(OH)_2 + (Fe,Mg)_3Al_2Si_3O_{12} + 5SiO_2$$
$$= 7(Fe,Mg)SiO_3 + NaAlSi_3O_8 + 2CaAl_2Si_2O_8 + H_2O$$

De Waard uses this reaction as the basis for an orthopyroxene isograd (1966, p. 488 figure 2) marking the high P_{H_2O} limit of his biotite-cordierite-almandite subfacies. According to de Waard's diagram parageneses on the low grade side of the isograd lack orthopyroxene, while those on the high grade side, if of appropriate composition, may contain it.

A diagram analogous to figure 2 (this paper) can be drawn for reaction 3. On the low grade side the reaction curve, in a closed system, orthopyroxene can coexist in divariant equilibrium with hornblende-almandite-quartz and *either* plagioclase *or* an aqueous fluid. On the high grade side of the curve, again in a closed system, orthopyroxene is stable in divariant equilibrium with plagioclase, an aqueous fluid and *any two* of the phases hornblende, almandite and quartz. Reaction 3 therefore cannot be used to define an isograd marking the absolute first or last appearance of orthopyroxene, as de Waard's figure 2 clearly implies. The other isograds are open to the same general criticism.

CONCLUSION

The basis of de Waard's subdivision of the granulite facies is questionable for the following reasons:

1. the choice of P_{load}, T and 'P_{H_2O}' as definitive variables is arbitrary, and de Waard's use of them is not consistent with the physicochemical behaviour of real systems:
2. the subdivision is based partly on hypothetical reactions, for which no evidence is offered; and in the manner stated by de Waard, the simple criterion of coexistence of products and reactants of a particular reaction, cannot be used to recognize a subfacies. Furthermore, de Waard's isograds are not uniquely defined by the reactions that he uses.

ACKNOWLEDGMENTS

I would like to thank Professor C. Wayne Burnham for reading this manuscript, and for suggesting ways in which the presentation might be improved.

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Manuscript received June 26, 1967