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THOLEIITIC AND CALC-ALKALIC SERIES IN RELATION TO THE BEHAVIORS OF TITANIUM, VANADIUM, CHROMIUM, AND NICKEL

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ABSTRACT. In typical tholeiitic (TH) series of volcanic rocks, the FeO* (total iron as FeO), TiO_2, and V contents increase first and, then passing a maximum, begin to decrease with advancing fractional crystallization (that is, with increasing FeO*/MgO), whereas in typical calc-alkalic (CA) series, the FeO*, TiO_2, and V contents decrease monotonically and rapidly with advancing fractional crystallization. Fe, Ti, and V are elements particularly concentrated in magnetite. So, the similar behavior of these three elements in relation to the distinction between the TH and CA series is consistent with the view that the difference between the two series is controlled mainly by the crystallization of magnetite. However, it is noted here that the rock series classification is based on the patterns of compositional variation of magmas with fractional crystallization and not on the origin of their parental magmas. It is conceivable that parental magmas of the CA series, for example, are generated by two or more different processes.

The Cr content of volcanic rocks decreases with increasing FeO*/MgO ratio approximately in the same way in both TH and CA series. Though the Ni content also tends to decrease with increasing FeO*/MgO, TH series rocks tend to have higher Ni contents than CA series rocks with the same FeO*/MgO ratios. An analogous tendency exists but is very weak for Cr contents.

INTRODUCTION

The tholeiitic (TH) and calc-alkalic (CA) series are the two major rock series in the non-alkalic (subalkalic) igneous rocks. In each series, the composition of rock (or magma) should vary with advancing fractional crystallization. So, the major and trace element compositions of igneous rocks could be understood in relation to the rock series and the stage of fractional crystallization. Some trace elements show much higher abundance in one series than in the other of the two. Other trace elements show no such preference. Such relations should give a clue to a better understanding of the nature and origin of igneous rock series.

In this paper, we are dealing with the behaviors of vanadium, chromium, and nickel in the TH and CA series to show how their behaviors cast light on the nature of the series.

In the past, there was considerable ambiguity in the concept of igneous rock series, because intrinsic and more casual characteristics of series were not clearly distinguished. So, for example, Kuno's (1950, 1959) hypersthenic series of Hakone Volcano was classed as a TH series by Nockolds and Allen (1956), whereas it was regarded as a typical CA series by Kuno himself. Miyashiro (1974) tried to clarify the concept of rock series by formulating the intrinsic characteristics of the TH and CA series. There, the TH series was defined by a slower rate of increase of SiO_2 content and higher enrichment of FeO* (= total iron as FeO) with advancing fractional crystallization in basaltic magmas than the CA series as was shown in figure 1 of Miyashiro (1974). The advance in fractional crystallization was measured by increase in FeO*/MgO ratio. The boundary between the two series was represented by the following equation (Miyashiro, 1974, p. 325):

$$SiO_2 \text{ (percent)} = 42.8 + 6.4\ FeO^*/MgO \qquad (1)$$

The TiO_2 content of magma shows patterns of variation more or less similar to those of FeO* as was shown in figure 12 of Miyashiro (1974). Typical TH series show enrichment in TiO_2 with a maximum during fractional crystallization, whereas CA series show monotonically decreasing TiO_2 content. Figure 1 shows the FeO* and TiO_2 contents of a large number of volcanic and related intrusive rocks, classified into TH and CA series by the above definition, from various tectonic settings (island arcs, active continental margins, stable continents, oceanic islands, and mid-oceanic ridges). For the range $FeO^*/MgO < 2.5$, points of TH and CA series rocks are mixed, whereas for higher FeO*/MgO ratios, TH series rocks tend to have higher FeO* and TiO_2 contents than CA series rocks. In both FeO* and TiO_2 diagrams, field (A) has only CA series rocks, field (B) both CA and TH series rocks, and field (C) only TH series rocks. Three fields, (A), (B), and (C), are situated in anticlockwise order with the TH-CA mixed area at the center.

Fe and Ti are elements highly concentrated in magnetite. Under a high oxygen fugacity, much magnetite would crystallize from a relatively early stage, leading to depletion of the residual magma in Fe and Ti and complementary enrichment in SiO_2; this should be the CA trend. On the other hand, under a lower oxygen fugacity, crystallization of magnetite would be delayed, and hence Fe and Ti are not so strongly removed from magma as in the CA series; this should be the TH trend (Kennedy, 1955;

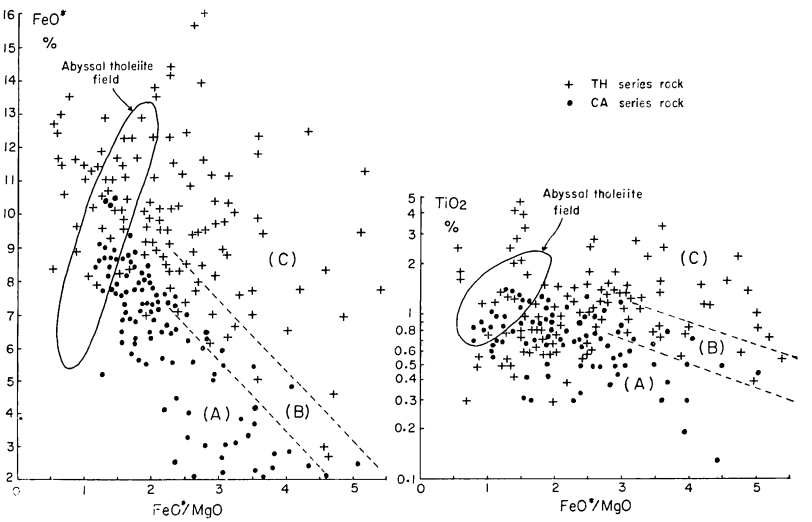

Fig. 1. FeO* and TiO_2 contents of volcanic and related intrusive rocks of the TH and CA series from island arcs, active continental margins, stable continents, and Hawaiian Islands. (The TiO_2 contents are shown on the longarithmic scale for comparison with figure 5). In this figure as well as in figures 5, 6, 8, and 9, the outline of the composition field of abyssal tholeiites is shown, but their individual analyses are not plotted.

Osborn, 1962). Later it will be shown that V behaves like Fe and Ti. This will give an additional support to this view, as V is also an element characteristically concentrated in magnetite.

TRACE ELEMENT DATA TO BE USED AND COMMENTS ON HAKONE VOLCANO AND KARROO DOLERITES

In this paper, we are interested mainly in volcanic rocks. However, some intrusions associated with volcanic rocks such as the Skaergaard, Palisades, and Karroo are also treated. Only the trace element data accompanied by major element analyses are used, because major element compositions are required for the determination of rock series and FeO*/MgO ratio. Furthermore, only the trace element data for rocks in typical tectonic settings are used, because we wish to determine the relation between the tectonic setting and trace element contents.

Island arcs.—Trace element data for TH series rocks in immature island arcs are reported for the Tongas by Bryan, Stice, and Ewart (1972). For well-developed island arcs, data are available for the Lesser Antilles (Nockolds and Allen, 1953; Baker, 1968), Hakone Volcano in Japan (Nockolds and Allen, 1956), Izu Peninsula and Asama Volcano in Japan (Taylor and White, 1966), New Zealand (Taylor and White, 1966), and Guam in the Marianas (Stark, 1963). The rocks investigated by Taylor and White (1966) belong to the CA series, and the investigated rocks from all the other areas belong to both CA and TH series.

The rocks of Hakone Volcano show diverse trends and are divided into three groups as shown in figure 2. As shown in the SiO_2 diagram of this figure, trend (1) represents the CA series, trend (2) represents a mild TH series, and trend (3) a typical TH series. Most of Kuno's hypersthenic (H) series belongs to our CA series, whereas most of his pigeonitic (P) series to our TH series. The FeO* and TiO_2 diagrams show distinctive trends of the three groups. Group (1) shows monotonically decreasing FeO* and TiO_2 with increasing FeO*/MgO; group (2) shows monotonically decreasing FeO* but nearly constant TiO_2; group (3) shows FeO* and TiO_2 curves with maxima.

Active continental margins.—Trace element data were reported for the Lassen Peak in California and the Crater Lake in Oregon (Cascade Range) by Nockolds and Allen (1953) and for the Andes in northern Chile (Siegers, Pickler, and Zeil, 1969). All these rocks belong to the CA series, except for analysis "H" of Lassen Peak.

Stable continents.—The trace element data for the typical TH series rocks of the Skaergaard intrusion (Wager and Mitchell, 1951), the Palisades diabase of New Jersey (Walker, 1969), and the related Triassic diabase near Dillsburg in Pennsylvania (Hotz, 1953) are used.

Nockolds and Allen (1956) gave data for the Karroo dolerites and the British Tertiary "tholeiites" as well. Though these rocks have been regarded as tholeiitic, examination of the major element data indicates that they show considerably diverse trends, including both TH and CA series. As this view is in contrast to the traditional one, the degree of

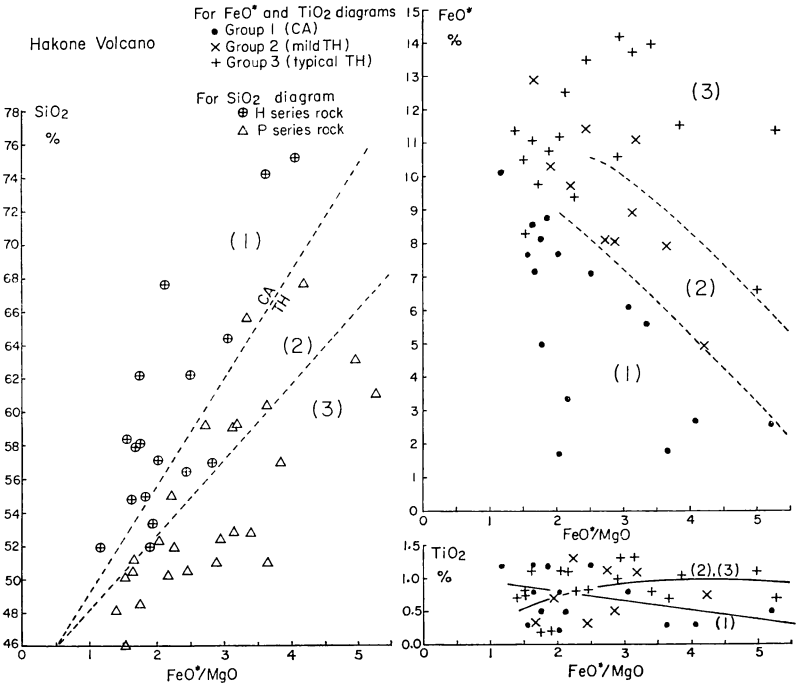

Fig. 2. Variation of SiO_2, FeO*, and TiO_2 with FeO*/MgO in volcanic rocks of Hakone volcano, Japan. The rocks are divided into three groups (1), (2), and (3). This diagram is based on tables 18 and 19 of Nockolds and Allen (1956).

diversity is shown in figure 3. (This view will be shown to be consistent with the V relation in fig. 4).

Oceanic islands.—Nockolds and Allen (1956) and Kuno and others (1957) reported trace element data for TH series rocks of Kilauea, Mauna Loa, and Koolau in Hawaiian Islands. McBirney and Williams (1969) reported data for the Galapagos Islands. These rocks appear to belong to a range of trends covering typical and mild TH series.

Mid-oceanic ridges.—Practically all the published data for fresh abyssal tholeiites from the mid-oceanic ridges are used. The authors include: Engel, Engel, and Havens (1965), Melson and Thompson (1971), Thompson, Shido, and Miyashiro (1972), Kay, Hubbard, and Gast (1970), Aumento (1968), and Campsie and others (1973). The distinction between authentic ocean-floor rocks and erratics is of vital importance. Abyssal tholeiites are defined here as tholeiitic rocks with $K_2O < 0.40$ percent from ocean floors. Rocks with $Fe_2O_3/FeO > 0.5$ and/or $H_2O > 2.0$ percent were rejected as being altered. When abyssal tholeiites are shown in diagrams in this paper, only the outlines of their composition fields are shown, and individual analyses are not plotted, because there are too many analyses to be plotted in the small fields.

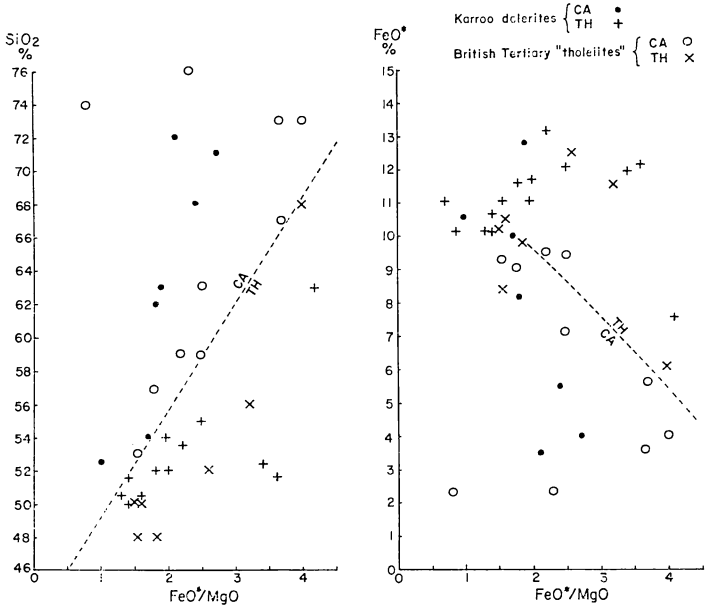

Fig. 3. TH and CA series in the Karroo dolerites and British Tertiary "tholeiites". The analyses given in tables 20, 22, 24, and 26 of Nockolds and Allen (1956) are classified into TH and CA series by the SiO_2 diagram (left) in this figure, and the FeO* contents of rocks of each series are shown in the FeO* diagram (right).

BEHAVIOR OF VANADIUM

Vanadium shows a behavior similar to FeO* (or Fe) and TiO_2 (or Ti). This is of particular interest because the behavior of FeO* is intrinsically related to the distinction between the TH and CA series.

In figure 2 we classified volcanic rocks of Hakone Volcano into three groups on the basis of behaviors of SiO_2, FeO*, and TiO_2. The V contents of rocks of these groups are shown in the upper diagram of figure 4. With increasing FeO*/MgO, series (1), which is CA, shows monotonically and rapidly decreasing V content, series (2), which is mild TH, shows slowly decreasing V content, whereas series (3), which is typical TH, shows an increasing V content at the beginning followed by a maximum and then by a decreasing V content. As all the values were determined by Nockolds and Allen (1956), there is no effect of interlaboratory differences. Thus, the behaviors of vanadium in the CA and TH series here are analogous to those of FeO* and TiO_2 shown in figures 1 and 12 of Miyashiro (1974).

The lower diagram of figure 4 shows a clear maximum in the V contents of the typical TH series diabases in the Palisades and Dillsburg sills (Walker, 1969; Hotz, 1953) as well as the rapidly decreasing V contents in the typical CA series rocks of the Lassen Peak, Crater Lake (Nockolds and Allen, 1953) and the Andes in northern Chile (Siegers, Pickler, and Zeil, 1969).

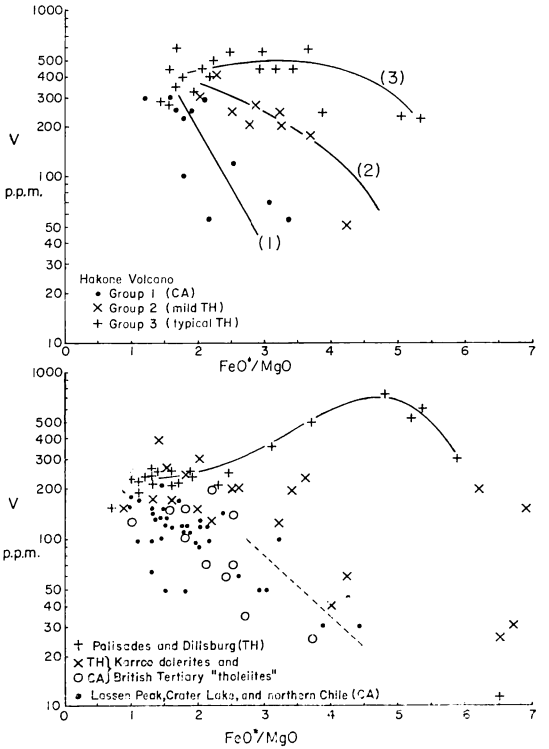

Fig. 4. Upper diagram: Vanadium contents in volcanic rocks of Hakone Volcano. The classification into three groups (1), (2), and (3) is the same as in figure 2. Lower diagram: Vanadium contents in some TH and CA series rocks.

It was stated before that the Karroo dolerite and the British Tertiary "tholeiites" include both TH and CA series (fig. 3). Hence, analyses of these rocks are classified into the two series, and their V contents are plotted in the lower diagram of figure 4. TH series rocks plot in higher positions than CA series rocks in this diagram in harmony with the above view. A boundary between the fields of the two series in this case is shown with a broken line.

Figure 5 shows the V contents of a large number of volcanic and related intrusive rocks of the TH and CA series from various tectonic settings (island arcs, active continental margins, stable continents, oceanic islands, and mid-oceanic ridges). It is observed that an area with low FeO*/MgO and high V values has both TH and CA series rocks (basalts), and with this area as a center, the diagram may be divided into three diverging fields (A), (B), and (C) in the anti-clockwise order: (A) has only CA series rock, (B) has both CA and TH series rocks mixed, and (C) has only TH series rocks. This relation is very similar to those of FeO* and TiO_2 shown in figure 1. This diagram suggests that the original basaltic magmas of all the non-alkalic series have more or less similar,

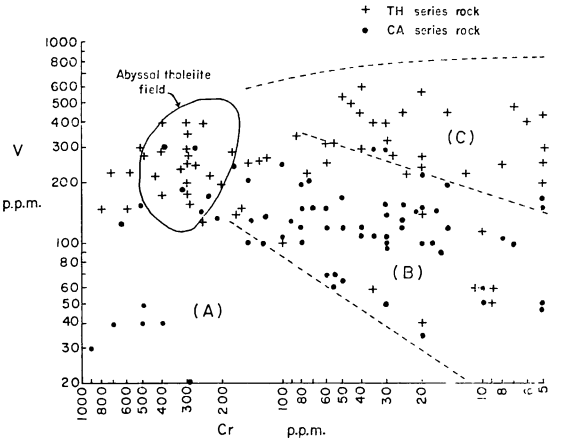

Fig. 5. Variation of vanadium content with increasing FeO*/MgO in TH and CA series rocks of various tectonic settings (island arcs, active continental margins, stable continents, oceanic islands, and mid-oceanic ridges).

high V contents (150-500 ppm), and during fractional crystallization the V content of residual magma gently increases, or remains constant, or gently decreases in TH series but rapidly decreases in CA series.

BEHAVIOR OF CHROMIUM

The Cr content of magmas of CA and TH series decreases rapidly with advancing fractional crystallization (that is with increasing FeO*/MgO). It was clearly observed, for example, in rocks from the Skaergaard intrusion, the Palisades and Dillsburg diabases, and northern Chile.

Figure 6 shows the Cr contents of TH and CA series volcanic rocks in various tectonic settings. TH and CA series rocks plot mixed in the same zone showing decreasing Cr content with increasing FeO*/MgO. The field of abyssal tholeiites is included in this zone and is near the low-FeO*/MgO end.

It is known that the Cr contents of island arc volcanics are usually much lower than those of abyssal tholeiites (for example, Jakeš and Gill, 1970). The usually high Cr contents of abyssal tholeiites, however, are related to their usually low FeO*/MgO. If we compare rocks with the same FeO*/MgO, abyssal tholeiites and island arc volcanics have similar Cr contents.

Thus, the Cr content of all the TH and CA series rocks may be regarded approximately as a function of FeO*/MgO. If we take the full straight lines in figure 6 as representing approximate Cr contents, the relation may be expressed by the following equation:

$$\log \text{Cr (ppm)} = 3.79 - 1.08\ \text{FeO*/MgO} \qquad (2)$$

Now that log Cr is approximately a linear function of FeO*/MgO, we can draw diagrams to distinguish CA and TH series analogous to

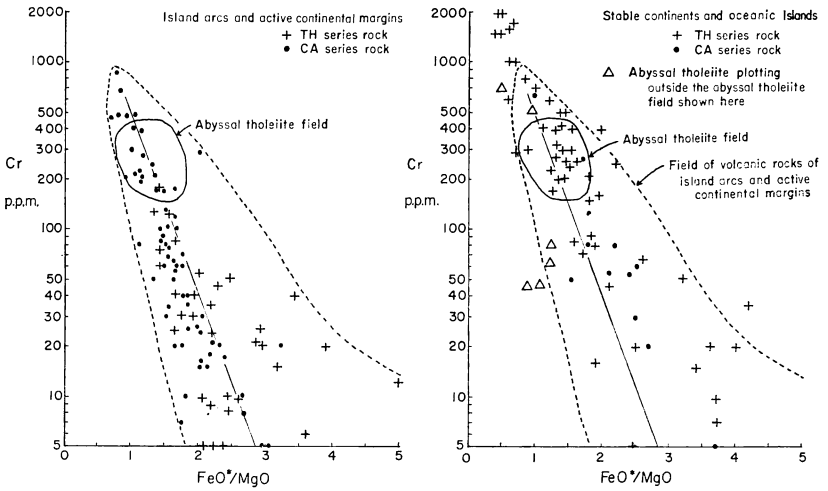

Fig. 6. Chromium contents of CA and TH series rocks in island arcs and active continental margins (left) and in stable continents and oceanic islands (right). The abyssal tholeiite field (full line) and the field of volcanic rocks of island arcs and active continental margins (broken line) are shown in both diagrams.

figure 1 of Miyashiro (1974) by using log Cr as abscissa in place of FeO*/MgO. Thus, the SiO_2 versus log Cr diagram of figure 7 is analogous to the SiO_2 versus FeO*/MgO diagram in figure 1 of Miyashiro (1974). By eliminating FeO*/MgO from (1) and (2), we have the following equation:

$$SiO_2 \text{ (percent)} = 65.3 - 5.9 \log \text{Cr (ppm)} \qquad (3)$$

This equation represents the straight broken line in figure 7, which separates TH series rocks considerably from CA series rocks.

In a similar way, a diagram which is analogous to figure 5 but uses log Cr in place of FeO*/MgO can be constructed as shown in figure 8. The boundaries between field (A) and (B) and between fields (B) and (C) have been transferred from the corresponding boundaries in figure 5 with equation (2). It is observed that these transferred boundaries fit the distribution of TH and CA series rocks fairly well. On the other hand, the abyssal tholeiite field shown in this figure has been determined by plotting real Cr and V data.

BEHAVIOR OF NICKEL

Nickel shows a behavior analogous to that of chromium. The Ni content of rocks of CA and TH series decreases rapidly with increasing FeO*/MgO. This has been observed, for example, in Hakone Volcano, the Palisades and Dillsburg diabases, and northern Chile.

Figure 9 shows that (1) the Ni contents of volcanic rocks plot in a wide zone indicating decrease of Ni with increasing FeO*/MgO; (2) rocks of stable continents and oceanic islands tend to spread in a wider field with higher upper limits for Ni contents than those of island arcs and

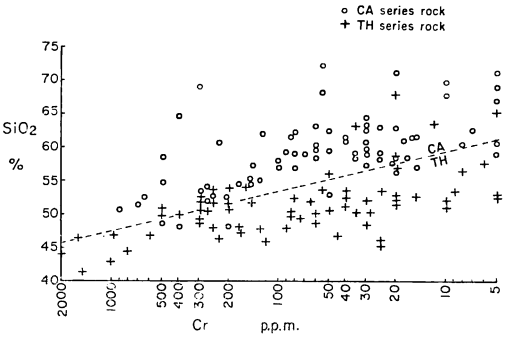

Fig. 7. SiO_2 versus log Cr diagram for CA and TH series rocks, which is analogous to figure 1A of Miyashiro (1974).

continental margins; (3) TH series rocks tend to have higher Ni contents than CA series rocks with the same FeO*/MgO; and (4) the abyssal tholeiites field is included in the zone of rocks of island arcs and active continental margins (Miyashiro and Shido, in press).

The behavior of Ni appears to resemble that of Cr in (1) and (4) but apparently differs from that of Cr in (2) and (3). However, if we carefully examine figure 6, it is observed that relations analogous to (2) and (3) are present in Cr in an incipient form. The Cr differences corresponding to (2) and (3) were so small that they were ignored in the preceding section.

If we take only volcanic rocks of island arcs and active continental margins and abyssal tholeiites, their Ni contents plot in a relatively narrow zone. If we take the straight full line shown in figure 9 as representing approximate Ni contents, the following relation holds:

$$\log \text{Ni (ppm)} = 3.22 - 0.92\ \text{FeO*/MgO} \tag{4}$$

By eliminating FeO*/MgO from (2) and (4), we have the following relation:

$$\text{Ni} = \text{Cr}^{0.85} \tag{5}$$

We can draw an SiO_2 versus log Ni diagram analogous to the SiO_2 versus log Cr diagram of figure 7. In such a diagram, TH and CA series rocks plot to a considerable extent in fields separated from each other (Miyashiro and Shido, in press). However, the degree of separation in this case is lower than that in figure 7.

Figure 9 gives a possible method of distinguishing some (not all) basalts of island arcs and continental margins from abyssal tholeiites and distinguishing some basalts of oceanic islands and stable continents from the abyssal tholeiites and island arc basalts. This may be useful in the determination of the origin of some ophiolites.

ORIGIN OF PARENTAL MAGMAS OF TH AND CA SERIES

V is an element characteristically concentrated in magnetite and to a smaller extent in ilmenite (Wager and Mitchell, 1951; Duncan and

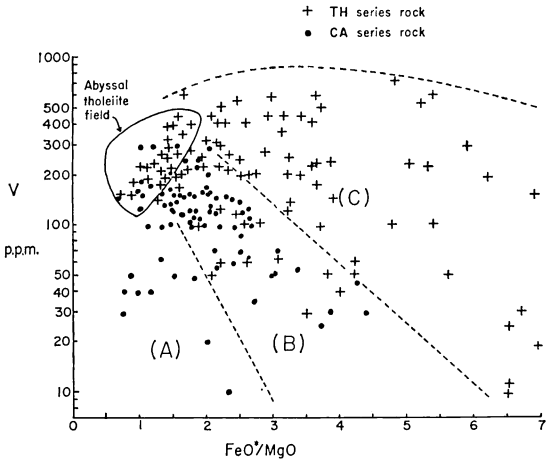

Fig. 8. Log V versus log Cr diagram for CA and TH series rocks, which is analogous to figure 5.

Taylor, 1968). It is scarce in silicates. So, the above-shown similarity in behavior of V to Fe and Ti is consistent with the view that the TH and CA series are formed by fractional crystallization of basaltic magmas, and that the difference between the two series is controlled by the crystallization stage and the amount of magnetite. Parental magmas of TH and CA series may have had different values of oxygen fugacity at the time of their generation by partial melting of the upper mantle (Miyashiro, 1974). This view is compatible with the fact that there is no substantial difference in $^{87}Sr/^{86}Sr$ ratio between many volcanic rocks of the TH and CA series.

This, however, does not preclude the possible generation of some CA series magmas by other processes, because the CA and TH series are defined by patterns of compositional variation of magmas and not by the origin of parental magmas. Many silicic rocks of the British Tertiary "tholeiites" actually belong to the CA series (fig. 3). There is strong Sr isotope evidence for the view that silicic rocks of the British Tertiary province were formed by melting of and contamination by the Precambrian basement (Moorbath and Bell, 1965). Some rocks related to the Karroo dolerites belong to the CA series (fig. 3), and here also participation of crustal rocks may well be considered as was discussed by Walker and Poldervaart (1949) in relation to the origin of granophyre. The origin of CA series rocks in island arcs and active continental margins was ascribed by some authors in the past to contamination by rocks in continental crust (Kuno, 1968, p. 146-149).

Thus, two or more genetically different groups of CA series rocks may exist. The TH series also may include genetically different groups, as suggested by the occasional occurrence of high $^{87}Sr/^{86}Sr$ ratios in some TH series volcanic rocks of orogenic belts (for example, Williams and

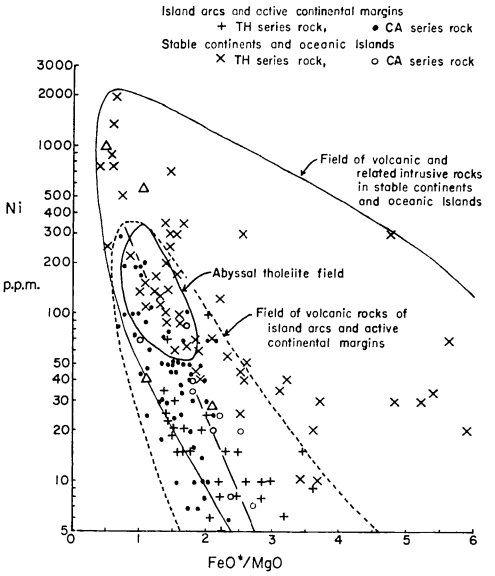

Fig. 9. Fields of nickel contents of abyssal tholeiites, volcanic, and related intrusive rocks in island arcs and active continental margins and non-orogenic regions (stable continents and oceanic islands). Rocks (except for abyssal tholeiites) are classified into TH and CA series, and their individual Ni values are plotted. A few abyssal tholeiites plot outside the abyssal tholeiite field outlined here as shown by triangles.

McBirney, 1969). If the difference between the TH and CA series is controlled by crystallization of magnetite, possible casual removal of hydrogen from a TH series magma may transfer the magma into a CA trend, as may be exemplified by a small amount of CA series rocks associated with predominant TH series ones in the Kermadec Arc (for example, fig. 6 of Miyashiro, 1974). However, no detailed discussion will be possible on such speculations until more mineralogic and chemical data of rocks belonging to well-established volcanic series become available.

Taylor and others (1969a,b) discussed the origin of andesites in orogenic belts on the basis of some trace elements including V, Cr, and Ni. Osborn (1969) criticized their view. Taylor and others emphasized that andesites were similar to high-alumina basalt in V and Ni contents. They rejected the genesis of andesites by fractional crystallization of high-alumina basalt magma and by contamination of magma by granitic rocks or deep-sea clays and ascribed the formation of andesites to a two-stage partial melting of upper mantle material. There could be some ways of reconciliation between the view of Taylor and others and ours, because our view does not preclude the possible origin of some parental magmas of the CA series by two-stage partial melting.

The presently available data, however, are too scarce for reliable discussion on such problems. For example, Taylor and others used Ni = 25 ppm for high-alumina basalt, which was based on their study of such

rocks from New Zealand. Some high-alumina basalts of the CA series from Amagi Volcano are reported to contain 50 to 60 ppm Ni (Kurasawa, 1959). We do not know what values are representative of the Ni contents of high-alumina basalts of the TH and CA series and whether other basalts need not be considered as possible parents of andesites.

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