PETROCHEMISTRY OF AN UPPER PRECAMBRIAN METASEDIMENTARY BELT IN SOUTHERN BAHIA — THE SIGNIFICANCE OF CORDIERITE

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ABSTRACT

Petrology and chemistry of an Upper Precambrian metasedimentary belt in Southern Bahia (Brazil) have been studied. The Belt consists essentially of muscovite-biotite gneisses and of sillimanite-grade rocks (kinzigites) containing lenses of acid-and sub-acid granulites, and pluton-like bodies of migmatitic-granitic rocks. Cordierite-bearing rocks are widely spread within the sillimanite-grade rocks. Cordierite appears always as the last paragenetic mineral and probably is formed through the

reaction sillimanite + garnet + quartz = cordierite. Chemical data indicate that the original material for mica gneisses and sillimanite-grade rocks consisted of a mixture of graywackes and pelitic rocks. Comparison with modern pelagic sediments reveals low contents of metals (Fe, Mn, Ni, etc.), relatively mobile in marine environments, thus suggesting possible submarine leaching. Bulk chemistry (showing low alkali and silica contents) was found to control closely cordierite

formation which is considered related to the removal of fractions of anatectic silicate liquids. The more general significance of cordierite in metasediments is discussed stressing the role of the partial H₂O pressure in the metamorphic fluids. The presence of even small amounts of H₂O-undersaturated melts strongly reduces the molar H₂O fraction and favours the stabilization of cordierite. Preservation of cordierite under more hydrous conditions depends on the further development of the P-T metamorphic regime.

INTRODUCTION

Regional geological reconnaissance and preliminary petrological and radiometric data indicate that a well-characterized metasedimentary belt of Upper Precambrian age occurs over a large area of Southern Bahia, continuing south for hundreds of kilometres essentially following the margin of the Atlan-

tic ocean. In Bahia State the belt develops in a NW-SE direction, being limited to the NE by Lower Precambrian granulite facies rocks prevalent throughout the State. The contact between Lower and Upper Precambrian rocks is not exposed. Regional studies suggest a fault contact between the two forma-

tions.

Structural and mineralogical features of the belt rocks indicate that they are similar to other metasedimentary rocks from other precambrian areas. The problems with which this paper deals may, therefore, be of wider significance.

NOTES ON GENERAL GEOLOGY

Regional geological mapping of the area covered by the Upper Precambrian sequence has been executed by workers of the DNPM-CPRM during the completion of the "Sul-Bahia Project". Belt rocks have been found in the Rio Pardo, Itapetinga, Monte Pascoal and Caravelas quadrangles, the total area extending in a NS direction between 15 and 18 lat. S and in a E-W direction between 40°30' and 42° long. and the Atlantic margin. General geology of this area has been described by Silva (1974) and is outlined in Fig. 1. The Upper Precambrian belt consists essentially of two main lithotypes covering distinct areas. They are:

i) Muscovite-biotite-oligoclase-quartz gneisses (mica gneisses) that appear to the N-E side of the belt. They are apparently at the fault contact with Lower Precambrian granulites previously studied by one of the authors (Sighinolfi, 1970, 1971).

ii) Sillimanite-grade rocks (kinzigites). They cover a large area in Southern Bahia and extend widely to the south (Grossi Sad, 1968).

No transitional contact between the two lithotypes has been found. Near the contact areas, however, repetition of mineralogical parageneses attributable to the two rock types is sometimes visible. Banded amphibolites and calc-silicate rocks are found associated with kinzigites and especially with mica gneisses. Kinzigites present felsic bands with granitoid structure like veins, lenses or layers parallel to foliation with boudinage and pinch and swell structures.

Plutonic masses of granitic-granodioritic composition are concentrated towards the SW border of the belt. Migmatitic rocks and granitic pegmatites are widely scattered within the sillimanite gneisses. Granulite-facies rocks are represented by lenses of charnockites aligned in a NW-SE direction following regional patterns. The relationships with the surrounding sillimanite-bearing rocks are not clear.

Near the NE border of the Upper Precambrian belt a structurally superimposed sequence of low-grade metamorphic sediments (phyllites, metaconglomerates, meta-arkoses) has been described and structurally defined by Pedreira *et al.* (1969) as the Rio Pardo Group.

Rb-Sr and K-Ar age data on the major rock units of the area were presented by Cordani (1973) and Cordani and Iyer (1978). Rb-Sr ages of the mica gneisses range between 1210 and 1490 my. with a maximum frequency at about 1.2 by. Sillimanite-grade rocks

are slightly younger (about one billion years). The Rio Pardo metasediments range in age between 600 and 640 my. K-Ar data over all the units of the area give invariably ages in the range of 400-450 my. Based on these data Cordani (1973) proposed a tectonic evolutionary scheme with the following main episodes:

1.3-1.6 by.: formation of the sedimentary basin, possibly correlated to the regional Espinhaço Cycle;

0.6-0.5 by.: Brazilian orogenic cycle. Epizonal metamorphism of the Rio Pardo Group. Rejuvenation of the paragneisses;

0.5-0.4 by.: End of cratonization and cooling of the Eastern Brazilian platform.

Finally, according to Leonardos and Fyfe (1974), the metasedimentary rocks of the area studied belong to a typical low-pressure type Atlantic belt widely extending southwards for several hundreds of kilometers.

DESCRIPTIVE MINERALOGY AND PETROLOGY

The mica gneisses are leucocratic, crudely foliated rocks which present very complex mineral paragenesis because of secondary reactions. Main parageneses present essentially quartz, oligoclase, non-perthitic microcline, green-brown biotite and minor amounts of muscovite and hornblende. Rutile is frequently associated with biotite. Epidote and, partially, chlorite are retrograde phases associated with the sericitization of feldspar and chloritization of biotite. Chlorite is also present as a residual mineral; it was probably involved, together with muscovite and quartz, in biotite-forming reactions as described by Bird and Fawcett (1973) and Hoscheck (1973). The frequent association of biotite and rutile suggests also a reaction of the type:

chlorite + muscovite + ilmenite = biotite + rutile + K feldspar + quartz + H₂O the last two minerals being subordinate reaction products (Ramsey, 1973).

The sillimanite-bearing gneisses are usually granoblastic and coarse-grained with porphyroblasts of cordierite and very occasionally of garnet. Constituent minerals are quartz, alkali feldspar, plagioclase, cordierite, biotite, garnet and sillimanite. The following assemblages are characteristics of the sillimanite gneisses:

- i) sillimanite-biotite- K feldspar
- ii) sillimanite-biotite-garnet-K feldspar

iii) sillimanite-biotite-cordierite

iiii) sillimanite-biotite-cordierite-garnet

Quartz and plagioclase are present in each assemblage, while graphite and magnetite are common in most. Quartz and plagioclase usually comprise more than 50 modal percent in most samples examined, although plagioclase and alkali feldspar show wide variations in their abundances depending on the mineral paragenesis. Grid-iron twinning and fine, string perthites characterize the alkali feldspar. Biotite is ubiquitous, varying from less than 1 to more than 20 per cent. It occurs as subhedral plates of variable size which are aligned with the foliation. Garnet occurs in most samples although it never exceeds 10 modal percent. It is anhedral and locally weakly elongated in the plane of foliation. Inclusions of quartz and biotite appear in some poikiloblastic grains of garnet. This, together with an apparent inverse relationship between biotite and garnet abundances, suggests that the latter, at least partially, is formed by reactions with biotite as one of reactants. Cordierite is ubiquitous within sillimanite-grade rocks. It occurs as colorless, anhedral, irregularly shaped grains which vary in size from 1 to 6 mm. They are usually poikiloblastic and contain abundant inclusions of biotite, quartz, sillimanite and rarely of garnet. Sillimanite is usually fibrolitic and intimately associated with biotite. In cordierite-bearing samples it occurs also as needles within cordierite. In a few samples andalusite crystals, apparently relictic, join sillimanite.

Also a few irregularly shaped grains of staurolite found in two samples are interpreted as relicts. "Primary" muscovite associated with quartz has not been found. Fine-grained, apparently late-stage muscovite is encountered surrounding sillimanite grains. Low temperature alteration phenomena, which probably occurred during retrogressive metamorphism, are also represented by the sericitic replacement of plagioclase, the marginal replacement of cordierite by pinite and the chloritization of ferromagnesian minerals. It cannot be excluded, however, that the formation of at least part of the white mica surrounding sillimanite grains might be caused by increased K: H ratios in the fluid phase (Eugster, 1970; Kwak, 1971). Granulite-facies rocks, present in the form of small lenses within sillimanite-grade rocks, are basically formed by andesine, perthitic K feldspar, quartz and orthopyroxene with limited amounts of biotite. No evidence of sillimanite, garnet or cordie-

rite has been found in hyperstene-bearing granulites.

CHEMISTRY

Average analytical data for 17 mica gneisses, 15 cordierite-free and 18 cordierite-bearing sillimanite-grade rocks are reported in Table 1 together with some data for amphibolite and granulite-facies rocks. The belt rocks of sedimentary origin (mica gneisses and kinzigites) show a certain range of compositional variation but most of the analyses cluster closely together around the composition of graywackes and pelitic shales (see e.g. Shaw, 1956; Pettijohn, 1963; Wedepohl, 1969; Rogers and McKay, 1972). It can thus be assumed that the Bahia belt, like other Precambrian metasedimentary belts (e.g., Ramsay, 1973; Andrae, 1974) consists of a mixture of varying proportions of non-mature and pelitic sediments. Diagrams A and B (Fig. 2) give information on the nature of the original material. These diagrams and average bulks chemistries (Table 1) indicate that there are not appreciable differences between mica gneisses and cordierite-free sillimanite grade rocks. On the contrary, cordierite-bearing kinzigites present quite distinctive chemistries; they are lower in silica and alkalis with respect to cordierite-free rocks (Table 1). Figure 2 C illustrates in more detail the control of chemical parameters (low ratios of alumina to mafic oxides, low alkali content, etc.) on the appearance of cordierite. Similar controls were found by numerous authors (Dallmeyer and Dodd, 1971; Dostal, 1975; Harriss, 1976).

Diagrams in figure 2 show that granulites have a distinctive bulk chemistry as compared to metasediments, clearly depicting an igneous trend. This might suggest that granulites originally consisted of some igneous material present within the sediments but it cannot be excluded (age data are not available on these granulites) that these rocks belong to the Lower Precambrian granulitic basement.

Additional indications about the nature of original sediments can be drawn by the analysis of interelemental relationships by the use of a linear coefficient matrix. Some significant relationships ($r = 0.70$) have been reported in Fig. 3, plotted after Chave and Mackenzie (1961) and Chayes (1971). Silica correlates negatively with most of the other main elements, as commonly observed in sedimentary series (Hirst and Kaye, 1971) and thus testifying to the presence of a detrital silica component. Also Ti probably was contained in detrital phases because of

the lack of Ti-Al relationships, typical of pelitic rocks (Wright, 1974). K and Rb are closely correlated as is common in a variety of geological environments. The lack of any correlation between Rb and Al may indicate that most of Rb was concentrated in detrital feldspar and not in the clay fraction (Stephens *et al.*, 1975). Manganese correlates closely with iron, as in igneous rocks (Schroll, 1968). This may underline the importance of igneous minerals or debris within the starting materials concentrating mafic elements.

There has been much discussion recently on the contribution of submarine volcanism to the accumulation rates of some elements (Fe, Mn, Cu, V, Ni, etc.) in oceans. Some studies (e.g. Bostrom and Peterson, 1969; Bostrom *et al.*, 1973) on the composition of marine sediments where exhalative activity at the present time is intense (e.g. the East Pacific Rise) show that there is a gradual increase in some elements like Fe and Mn, while the concentration of detrital elements like Ti falls to negligible values. On the assumption that elements like Al and Ti are totally of continental derivation, while Mn and Fe may be in part correlated to submarine igneous activity, element ratios like Fe/Ti or Al/(Al+Fe+Mn) (Bostrom, 1970; Bostrom *et al.*, 1972) and Fe+Mn/Ti (Strakhov, 1976) have been used to detect whether it is, in part, of submarine exhalative origin. Calculation of these ratios for the Bahia metasediments does not reveal any enrichment of "exhalative" elements. On the contrary, these elements are depleted relative to the "detrital" elements, when compared with modern oceanic sediments. This presumably reflects the influence of different sedimentation environments, even if submarine leaching of mobile elements, as proposed by Glasby (1975), cannot be excluded.

OUTLINE OF METAMORPHISM

The interpretation of the metamorphic evolution which the belt rocks have undergone and the inference of T,P,P_{H2O} conditions during the various steps of metamorphism are hampered on one hand by the superposition of retrogressive phenomena, and on the other by the lack of compositional data on coexisting mineral phases. As the effects of retrograde metamorphism are particularly intense in mica gneisses, the following discussion will concern only

sillimanite-grade rocks. A prerequisite to the general interpretation of metamorphism is the analysis of equilibria between mineral parageneses. There are many criteria to assess equilibrium conditions. The most frequently considered base on the form of grain boundaries, on the phase rule and on the composition of coexisting minerals. On account of the lack of the latter information and of the limited applicability of phase-rule criteria (Dallmeyer and Dodd, 1971), the analysis of phase equilibria is here mainly made by considering reciprocal phase contacts (note at this point criticisms by various authors, e.g. Loomis, 1976). Grain boundaries suggest that most of the major phases of kinzigites (biotite, sillimanite, K feldspar and most of garnet) represent high-temperature assemblages formed during a high-grade metamorphic episode. Cordierite is the last mineral formed during the same episode. Staurolite and andalusite represent residual reactant phases and are considered left-overs of reactions (see e.g. Yardley, 1977) which give as products the high-temperature assemblages. As regards staurolite, its rarity in the analyzed samples is probably related to its extremely limited P-T stability field (Richardson, 1968; Thompson A.B., 1976) rather than to compositional factors (Hoschek, 1969; Winkler, 1974).

The coexistence of andalusite and sillimanite in a few samples might connote univariant equilibrium. However, the relict character of andalusite strongly suggests that it is persisting metastably within the stability field of sillimanite (Albee and Chodos, 1969). Approximate P-T estimates for the high-temperature assemblages of kinzigites may be derived from experimentally and thermodynamically determined phase equilibria. The rocks are restricted to the sillimanite field, since this is the only stable Al_2SiO_5 polymorph in our area (alumosilicate relations on Fig. 4 after Holdaway, 1971). Further restriction of the P-T conditions is given by the observation that muscovite and quartz are incompatible. This suggests, according to experimental data (Evans, 1965; Hess, 1971), that, assuming intermediate (5-6 Kb) total pressure values, the temperatures exceeded 960° to 720°C. These conditions are largely above the granite minimum for P_{H2O} = P_{tot} (Luth *et al.*, 1964) and on account of the bulk "granitic" composition of the rocks involved extensive anatexis had to occur, assuming the fluid pressure is determined by water pressure. The following observations seem to indicate that water undersaturated conditions

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have prevailed during the high-grade metamorphic episode:

i) the general aspect of the rocks suggests that anatexis occurred on a limited scale;

ii) graphite is still widespread and, according to Eugster and Skippen (1967), the mole fraction of water in a C-H-O fluid in equilibrium with graphite at 700°C or at higher temperatures is considerably less than the unity.

Reliable information on total pressure conditions can be extracted from numerous experimental data on cordierite stability at various pressures (Hirschberg and Winkler, 1968; Currie, 1971; Hensen and Green, 1970, 1971, 1973). The following schematized reactions leading to cordierite formation can account for the observed parageneses in the Bahia kinzigites:

Obviously cordierite might form, during the same metamorphic event, by two or more reactions; being these reactions usually discontinuous reactants may persist together with reaction products. Reaction *a* might account for the observed garnet-free cordierite-bearing parageneses, as may also reaction *c* (in the case of complete garnet breakdown). Hoffer (1976) has recently found that reaction *a* occurs only in a very narrow P-T interval, more specifically at very low P (2.5-3 Kb). This seems to imply that this reaction is not important in regional metamorphism. Nevertheless, reaction *a* has been observed also in areas of regional metamorphism (e.g. Schreyer, 1966). Reaction *b* may explain the few occurrences of cordierite-bearing rocks having garnet in the form of poikiloblastic grains including biotite and sillimanite. However, there is no clear evidence of mutual compatibility or equilibrium between garnet and cordierite. Most of the observed cordierite-bearing parageneses were probably formed by reaction *c* which has been used, as is well known, by many researchers to calibrate the composition of coexisting garnet and cordierite as a function of temperature and pressure. Our lack of data on garnet and cordierite composition do not enable us to make more detailed deductions on the P-T metamorphic regime. The possible significance of cordierite, because of its widespread occurrence in many metasedimentary rocks, will be discussed in the next paragraph.

The assemblage quartz-sillimanite-garnet-cordierite is common both in regionally high-grade metasediments (Read, 1952; Reinhardt, 1968; Hutcheon *et al.*, 1974) and in metasediments in contact aureoles around granites (Lal and Moorhouse, 1969; Gonzales-Bonorino, 1971; Dostal, 1975). In areas of regional metamorphism, cordierite-bearing sillimanite-grade rocks are normally intimately associated with their non-cordierite counterpart. The causes of selective occurrence of cordierite is the main problem that these rocks pose, and the complexity of this problem is evident from the commonly reported unsystematic patterns of development of cordierite. Hypotheses currently proposed (see Lal and Moorhouse, 1969) which may be pertinent to the genesis of most cordierite-bearing metasediments are:

- 1) Variations in the P-T metamorphic setting, total pressure being obviously the most important factor. Phenomena which may introduce such variations are essentially: a) rapid uplift of crustal segments; b) overprint of thermal events.
- 2) Adequate bulk-rock composition leading to the formation of cordierite-bearing parageneses, the P-T metamorphic conditions being less important for the selective occurrence of cordierite. Numerous chemical parameters have been found to control the appearance of cordierite. The most important seem to be: high Mg/Mg+Fe ratios (Hensen and Green, 1971), low Al/Mg+Fe (Dallmeyer and Dodd, 1971), low lime content (Wynne-Edwards and Hay, 1963; Kays and Medaris, 1976), low alkali and silica content (Dostal, 1975; Harriss, 1976). Some of these chemical controls may be well explained by considerations on equilibria of cordierite-forming reactions with sillimanite and quartz as reactants are known to widen the stability field of cordierite (Hensen and Green, 1971). Low alkali contents and in general more mafic bulk compositions of cordierite-bearing rocks, with respect to their cordierite-free equivalents, were frequently explained as the result of anatectic processes with consequent loss of fractions of granitic silicate melts (Dostal, 1975; Harriss, 1976). Thus cordierite in metasediments would sometimes indicate a "restite" character of the rock. This interpretation, which may obviously apply to the cordierite gneisses of southern Bahia, is consistent with most of the well-documented chemical features of these

rocks. For example, the selective appearance of cordierite in lime-poor rocks accounts for the well-known control of plagioclase composition on the anatexis rate of granitic material. These considerations support the hypothesis that the cordierite-bearing kinzigites here studied formed under the influence of anatectic phenomena. On the other hand, anatexis in these rocks is undoubtedly documented by the numerous lit-par-lit leucosomes of granitic composition with conformable contacts.

We want, however, to stress the role of water pressure in some cordierite-forming reactions under various fluid phase compositions. It has been found (Schreyer and Seifert, 1969; Newton, 1970; Newton *et al.*, 1974) that Mg-cordierite in dry systems is stable over wider P-T fields than in wet systems. Kays and Medaris (1976) in investigating reactions with sillimanite and biotite as reactants and garnet and for $P_{H_2O} = P_{tot}$. However, a decrease in the X_{H_2O} mole fraction from 1.0 to 0.5 stabilizes cordierite and garnet (Fig. 3). The supposed large differences in H_2O -content of Fe- and Mg-cordierite end members (see the thermodynamic arguments by Wood, 1973, and counter arguments by Weisbrod, 1973) may also be an indication of the control of water pressure on cordierite-bearing assemblages. The most important consequence of this control is that cordierite composition and water pressure are closely intercorrelated variables. At a given T, P, P_{H_2O} set, rock composition (e.g. a given Mg-Fe ratio) may represent a critical parameter for the appearance of cordierite. Otherwise, for any P-T value and rock composition cordierite forms under a set of metamorphic conditions in which P_{H_2O} is critical. It has been argued (Wood, 1973) that conflicting P-T data arising from the use of the cordierite-garnet geobarometers and geothermometers (Currie, 1971, 1974; Hensen, 1972; Hensen and Green, 1970, 1971, 1973; P.H. Thompson, 1976) may only reflect the effect of variable water pressures.

In metamorphism, conditions of $P_{H_2O} = P_{tot}$ might be attained essentially by:

- i) dilution of the pore fluids by other fluids that reduce the activity of water,
- ii) selective incorporation of water in a liquid silicate phase. The first process has been proposed by Lal and Shukla (1975) and Hoefs and Touret (1975), among others. Hoefs and Touret (1975) suggest that the very low H_2O content of fluid inclusions in deep-seated rocks (Touret, 1970, 1971) is correlated to fluid exchange reactions with CO_2 of mantle provenance. According to current opinions (e.g. Reverdatto *et al.*,

1974), however, anatexis is the most efficient process which may affect fluid phase composition during high-grade metamorphism. Many authors (Brown and Fyfe, 1972; Fyfe, 1973; Wyllie, 1977) have underlined that, because almost all the water available at depth is related to the breakdown of hydrated phases, silicate liquids produced by anatexis must be strong water-undersaturated. If this is true, any water from residual pore fluid fractions or liberated by progressive dehydration reactions tends to be immediately captured by the melt, even if it is present in very limited amounts. This phenomena might have at least two important effects:

i) to initiate other dehydration reactions occurring for $PH_2O = P_{tot}$ under different P-T conditions. An example of this chain reaction could be represented by the progressive breakdown of muscovite, biotite and hornblende;

ii) to decrease strongly the H_2O molar percent in the fluid phase, thus favouring the onset of cordierite-forming reactions. This will be a consequence of the greater solubility of H_2O compared to that of CO_2 and of other fluid components in silicate liquids at metamorphic temperatures (Wyllie and Tuttle, 1959).

Thus the formation of cordierite for any given rock composition would frequently coincide with stages of incipient anatexis. The commonly found "restite" character of cordierite bearing rocks in comparison to their cordierite-free counterparts suggests that the production of anatectic melt is frequently accompanied by remotion of melt fractions. This, however, does not necessarily occur and liquids may recrystallize *in loco* giving "granitic" mineral parageneses. If the liquid-solid system remains sufficiently closed to water during recrystallization of liquids it should be expected an inverse reaction between liquids becoming progressively H_2O -saturated and relatively anhydrous cordierite generated under low PH_2O conditions. Such a reaction would presumably lead to break-down of cordierite and this is not usually observed, cordierite persisting together with newly formed hydrated phases (biotite) in granitic leucosomes.

The persistence of cordierite under varying PH_2O conditions may be imputed to kinetic factors inherent to metamorphic reactions but, probably, may be related to the much debated rate of diffusion of water within high-grade cordierites. It is well known

that high-grade cordierite may contain high H_2O contents independently on their bulk composition and recently Lepezin and Melnosky (1977) showed that the persistence of hydrous cordierite under conditions of high-grade metamorphism is possible only if cooling of the rocks after metamorphic climax occurs at a nearly constant P_{tot} .

Under these conditions, if PH_2O increases, early formed anhydrous cordierite may re-equilibrate to hydrous cordierite without undergo destruction. In this case final stages of regional metamorphism will be essentially related to variations in thermal regimes, for example as consequence of reduced addition, after the metamorphic climax, of heat from deep thermal sources.

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Tabel. 1 Average composition of the main rock types

	musco. -- biot. gneiss	amphibolite	cordierite-free kinzigites	cord. - bearing kinzigites	granulites
Nº	17	2	15	18	5
SiO ₂	69.28	46.42	69.18	64.65	58.16
TiO ₂	0.63	0.92	0.39	0.79	0.89
Al ₂ O ₃	15.05	17.87	16.19	16.58	18.74
Fe tot. +	3.68	7.94	2.41	6.98	7.30
MnO	0.06	0.20	0.04	0.14	0.15
MgO	0.50	8.95	0.65	2.80	2.38
CaO	1.76	13.59	1.65	1.04	6.51
Na ₂ O	3.47	1.67	3.54	1.84	2.30
K ₂ O	4.40	0.21	4.29	2.72	1.88
P ₂ O ₅	0.21	0.05	0.18	0.17	0.51
Cr ppm	26	312	51	236	161
Ni "	26	140	13.1	43.3	7.4
Rb "	138.2	1.	196.6	111.3	65.8
Sr "	195	415	246	123	272
Ba "	1067	210	899	622	552
Zr "	788	83	508	411	506

* = number of samples; + = as Fe₂O₃