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CL-RICH MINERALS IN ARCHEAN GRANULITE FACIES IRONSTONES FROM THE BEARTOOTH MOUNTAINS, MONTANA, USA: IMPLICATIONS FOR FLUIDS INVOLVED IN GRANULITE METAMORPHISM.

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INTRODUCTION. Although Cl-rich minerals have been recognized to develop in a number of petrologic environments ranging from submarine hydrothermal vents [1] to late mafic pegmatitic dikes and Pt-rich horizons in layered magmatic intrusives [2,3], they are most commonly found in granulite facies rocks. It is probably the earlier work on the high grade terrains of South India that most clearly demonstrated this [e.g. 4,5,6,7,8]. Nonetheless, most of the investigations on Cl-rich minerals from granulites dealt primarily with their chemical characterization but have not considered their petrologic implications. Clearly, before fully understanding granulite facies metamorphism the role of Cl in granulites must be explored.

There are three factors that influence the incorporation of Cl into hydrous silicates and phosphates: fluid composition, temperature and crystallochemical constraints. It has been experimentally confirmed that as chlorinity and acidity in an aqueous fluid increase and/or as temperature increases, the Cl concentrations in the coexisting micas also increase [9,10]. Experimental studies have also established that the compositional parameters (especially increasing ferrous Fe contents) that enable the hydroxyl site to approach ideal hexagonal symmetry in hydrous silicates strongly favor Cl incorporation [11].

With these factors in mind, the hydrous minerals of the granulite facies ironstones from the Beartooth Mountains, Montana are considered in this investigation.

BACKGROUND. The eastern portion of the Beartooth Mountains, composed predominantly of 2800 Ma granitic to tonalitic granitoids, gneisses and migmatites, contain enclaves of various supracrustal lithologies that range in size from a few cm to a few km [12]. The supracrustal lithologies, including pelitic schists, felsic gneisses, amphibolites, mafic gneisses, quartzites and ironstones, typically display mineral assemblages that are indicative of granulite facies metamorphism [13]. Application of a series of geothermobarometers indicate that the peak metamorphism took place at pressures of 5-6 kbar and temperatures of 750-800 C. Some of the lithologies are partially-to-completely reset by a subsequent amphibolite facies metamorphism that is attributed to the major magmatic event at 2800 Ma. The Rb-Sr isotopic systematics on the supracrustal lithologies produce a poorly constrained isochron of approximately 3400 Ma that has been interpreted as the time of the granulite facies metamorphism [12].

MINERALOGY AND PETROLOGY OF THE IRONSTONES. The iron-rich metasedimentary rock of the eastern Beartooth Mountains is a typical quartz-magnetite banded iron formation with bulk compositions ranging from 47 to 61 weight % SiO_2 , 30 to 40 % FeO

and 1 to 3 % Al_2O_3 . The bulk compositions and REE abundances indicate an origin of the ferruginous sediment on a continental shelf rather than a eugeoclinal depositional environment [12].

The mineral assemblages of the ironstones are, in general, similar to other granulite facies ironstones [14] with the most common assemblage being quartz + magnetite + ferrohypersthene + almandine + clinopyroxene. In addition, there are trace or minor amounts of a blue-green amphibole and dark brown biotite that are found as both inclusions and matrix phases. As such, they are interpreted as having been involved in the prograde metamorphism of the ironstones. Very minor amounts of cummingtonite after ferrohypersthene and actinolite after clinopyroxene localized along later fractures indicate the subsequent amphibolite facies metamorphism did not significantly affect the ironstones.

Based on calculations of coexisting mineral compositions it is estimated that the fluid phase has an $X(H_2O) < 0.3$ and is relatively oxidizing (near the NNO buffer). "Primary" fluid inclusions are apparently CO_2 -rich suggesting CO_2 is the dominant component in the fluid phase.

Both the biotites and amphiboles found in these ironstones have some of the highest Cl levels that have been documented to date (with the biotites and amphiboles in the matrix containing more Cl than the biotite and amphibole inclusions in garnet and ferrohypersthene). The biotites not only contain up to 2.9 wt % Cl (22 % of the OH site), but also have substantial amounts of Ba (up to 10.5 wt%) and Ti (up to 6.9 wt %) (see Table 1). The amphiboles contain up to 2.8 wt % Cl (40 % of the total OH site) and range in composition from a ferroan pargasite to a Cl-rich potassium hastingsite (see Table 1). In these minerals there is a general positive correlation among Cl levels and Fe, Ba and K (in amphiboles). These trends are in accordance with the expected crystallochemical controls of Cl incorporation.

Not all of the Cl variations, however, are purely controlled by the crystallochemical constraints. Based on the Cl contents of the biotites, the relative $f(H_2O)/f(HCl)$ ratios [10] range from 0.02 to 1 suggesting that some of the biotite variability must result from changes in the local fluid compositions. These calculations indicate that the amount of HCl in the fluid must be substantial and are probably only attained in an aqueous fluid. Furthermore, the increasing Cl-enrichment of the matrix biotite and amphibole relative to the included biotite and amphibole suggest an increasing chlorinity with grade. Nonetheless, based on fluid calculations and fluid inclusion work it is known that the dominant fluid is a CO_2 -rich fluid. Such a fluid will not support the high levels of Cl necessary to produce the elevated Cl contents in the silicates [cf. 15]. Consequently, we must entertain the possibility of an immiscible highly saline and acidic aqueous fluid coexisting with the dominant CO_2 fluid typically found in granulites.

IMPLICATIONS. Evidence for CO_2 -brine immiscibility have been found in some medium grade metamorphic rocks [e.g. 15,16]. However, this current study indicates that this immiscible behavior of fluids may extend into granulite facies conditions.

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The presence of such a high grade immiscible fluid may have significant affects on the behavior of granulite fluids. The aqueous brines may be preferentially absorbed on the surfaces of minerals relative to the nonpolar CO₂ fluid phase resulting in differential movement of the unmixed brine and CO₂ fluids [17]. Due to its capacity to form complexes, Cl-rich aqueous brines have been implicated in the movement of elements such as Pt group elements, Pb and rare earth elements [2,18].

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TABLE 1. Representative Cl-rich biotite and amphibole analyses

Sample name	QC82-45	QC82-45	QC81-113	QC82-45	QC82-45	QC82-45
Analysis pt	BIOTITE 1	BIOTITE 2	BIOTITE 3	AMPHIB 1	AMPHIB 2	AMPHIB 3
Remark	MATRIX	OPX INCL	OPX INCL	MATRIX	GAR INCL	MATRIX
SiO ₂	28.79	30.58	33.28	37.11	37.97	39.70
Al ₂ O ₃	14.12	13.88	15.05	11.38	10.86	11.78
TiO ₂	6.29	5.84	1.02	1.40	1.84	0.12
Cr ₂ O ₃	0.00	0.00	0.02	0.04	0.04	0.01
FeO	29.08	29.37	27.14	27.28	25.75	24.96
MnO	0.11	0.10	0.13	0.17	0.17	0.26
MgO	2.29	3.96	6.66	3.83	4.96	5.70
CaO	0.02	0.01	0.03	11.50	11.62	11.52
Na ₂ O	0.00	0.05	0.08	0.65	1.06	0.61
K ₂ O	5.47	6.93	8.18	3.20	2.39	2.97
BaO	9.65	7.25	2.90	0.55	0.53	0.19
Cl	2.74	2.26	2.41	2.61	1.82	1.34
F	0.10	0.07	0.07	0.08	0.25	0.21
SO ₃	0.00	0.17	0.00	0.03	0.04	0.00
Total	98.66	100.47	96.97	99.81	99.30	99.37
O=F,Cl	0.66	0.54	0.57	0.62	0.52	0.39
TOTAL	98.00	99.93	96.40	99.19	98.79	98.98