COMPOSITIONAL AND VOLUME CHANGES ACCOM-PANYING PROGRESSIVE SERPENTINIZATION OF DUNITES FROM THE WEBSTER-ADDIE ULTRAMAFIC BODY, NORTH CAROLINA

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Abstract

Chemical and modal analyses of 22 partially serpentinized dunites from the Webster-Addie ultramafic body in North Carolina suggest losses of Mg and Fe, additions of Si and Na, and an increase in volume during progressive serpentinization of the dunites.

INTRODUCTION

Although many petrologic studies of serpentinized ultramafic rocks are available (Thayer, 1966; Hostetler, *et al.*, 1966; Barnes, *et al.*, 1967; Page, 1966, 1967; Himmelberg and Coleman, 1968), existing data are not sufficient to determine if significant compositional and volume changes accompany progressive serpentinization. A major factor contributing to the inability to resolve this problem is the lack of quantitative geochemical and mineralogical data from a given ultramafic body relating the degree of serpentinization of a specific rock type to rock composition. Such data are presented herein for a suite of partially serpentinized dunite samples from the Webster-Addie ultramafic body in North Carolina.

The Webster-Addie ultramafic body, one of the many alpine ultramafic bodies in the Appalachian belt (Larrabee, 1966), is located in the northwestern part of Jackson County, North Carolina near the town of Sylva. It is elliptical in shape (about 10 km long and 5 km wide), dips outward, and varies in outcrop width from a few meters to about 600 m. A complete description of the petrology and geochemistry of the body is in preparation (Madison and Condie, in prep.) The body is composed chiefly of dunite, with lesser amounts of peridotite and pyroxenite (websterite). Most of the rocks are partially serpentinized and/or steatitized. In order to limit the effects of primary mineralogical variability and of steatitization, only partially serpentinized dunites were sampled for this investigation.

Twenty-two partially serpentinized dunites were analyzed for Si, Al, Fe, Mg, Ca, and Ni by nondestructive X-ray fluorescence and for Na by nondestructive neutron activation. The details of the analytical methods are described in Volborth (1963), Condie (1967a; 1967b), and Stueber and Goles (1967). X-ray fluorescence calibration curves for Si, Fe, Mg, and Ca were constructed from average values of two or more wet chemical analyses for each of four or five dunites from the ultramafic body representing varying degrees of serpentinization. Standard diabase W-1 was used as a standard for the Ni determinations and Na solutions were used as standards for the Na analyses (by neutron activation). The estimated analytical error for all elements is ≤ 3 percent.

Modal analyses were made with a mechanical point counter and sufficient area was counted on each thin section to insure an error $\lesssim 3$ percent. Olivine compositions were estimated by the X-ray diffraction method proposed by Yoder and Sahama (1957) with corresponding errors about 5 percent.

Rock densities were determined to an accuracy of greater than one percent with an Ohaus triple beam balance using 50 g portions of each sample. The results show a good correlation between sample density and modal serpentine. Such a correlation attests both to the accuracy of mineral identifications and to the representation of the hand specimens by their corresponding thin sections.

Results and Discussion

Chemical and modal analyses and densities of the partially serpentinized dunite samples are given in Table 1 in order of increasing serpentine content.¹ The samples included in this investigation have serpentine contents ranging from about 6 to 45 percent. The serpentine occurs both between olivine grains and as crack fillings within olivine grains. Large fractured grains of optically continuous olivine are often cross-cut by serpentine veinlets which compose up to 50 percent by volume of the original grains. X-ray diffraction studies of serpentine separates using the method proposed by Whittaker and Zussman (1956) suggest that the serpentine is chiefly, if not entirely, chrysotile. Olivine, in general, decreases in abundance as serpentine increases. The variation in olivine composition shown in Table 1 (Fo₈₄ to Fo₉₅) probably results chiefly from analytical error in the X-ray diffraction analyses. The dunites, as a whole, have a mean olivine composition of Fo90. Minor amounts of ubiquitous. euhedral chromite are present in all of the samples; continuous bands of euhedral chromite grains in some parts of the dunite suggest a primary

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¹ To obtain a copy of Table 1, order NAPS Document #00478 from ASIS National Auxiliary Publications Service, c/o CCM Information Sciences, Inc., 22 West 34th Street, New York, New York 10001; remitting \$1.00 for microfiche or \$3.00 for photocopies payable to ASIS-NAPS.

origin for this mineral. Studies under reflected light indicate that the chromite is extremely fresh and virtually unaffected by serpentinization. Traces of magnetite ($\leq 0.3\%$) of apparently secondary origin occur irregularly distributed in the serpentine veinlets. Varying but minor amounts of enstatite (En₈₈), diopside (Di₉₈ Hd₂), talc, chlorite, and tremolite occur in some of the samples. Neither the enstatite nor the diopside is appreciably affected by serpentinization and bastite pseudomorphs were not found in any of the samples. Optical and X-ray diffraction studies did not reveal the presence of brucite or of carbonates in the dunites. It is also important to note the absence of a clear-cut relationship between the geographic distribution of the samples and the degree of serpentinization. Serpentinization appears to vary in intensity in a random manner throughout the ultramafic body.

The geochemical data indicate that FeO and MgO decrease and that SiO_2 and Na increase with the amount of serpentine present in the rock. These relationships are illustrated in Figure 1 where the concentrations of these elements are plotted as functions of serpentine content. The data have been approximated with straight lines by linear regression analysis in the figure. A similar analysis of the CaO, Al_2O_3 , and Ni data failed to reveal any systematic relationships between the concentrations of these elements and the degree of serpentinization. The lack of a correlation between CaO and serpentine content differs from the results of Page (1966) which suggest that calcium is lost during serpentinization.

The partially serpentinized dunite samples have rather constant MgO/ (MgO+FeO) ratios (0.83-0.85, Table 1) regardless of degree of serpentinization. A similar observation has been reported for partially serpentinized ultramafic rocks from the Red Mountain-Del Puerto ultramafic body in California (Himmelberg and Coleman, 1968). If the Webster-Addie dunites were of approximately the same original composition prior to serpentinization, this composition being determined by extrapolating the MgO, FeO and SiO, curves in Figure 1 to zero percent serpentine (i.e., about Fogo), it appears that MgO and FeO have been removed and that SiO, has been added during serpentinization (as suggested by the curves in Fig. 1). The fact that the average olivine composition of the dunites (Fogo) is equal to this extrapolated initial dunite composition supports this idea if the average olivine composition records the composition of the dunites prior to serpentinization. If this model is correct, Fe and Mg must have been lost in a manner which approximately preserved the initial MgO/MgO+FeO ratio of 0.84. A suggested reaction which permits both SiO, addition and MgO and FeO loss is as follows:

olivine	added		serpentine	lost in solution
$5Mg_2SiO_4$	$+ \widetilde{SiO_2}$	+ 6H ₂ O -	$\rightarrow 3Mg_3Si_2O_5(OH)_4 +$	MgO
(704 gm, 220 cc)	(60 gm)	(108 gm)	(831 gm, 330 cc)	(40 gm)



FIG. 1. Variation in FeO, MgO, SiO₂, and Na in the Webster-Addie dunites as a function of serpentine content. m= slope; b= intercept at zero percent serpentine; r-correlation coefficient. All correlation coefficients are significant at the 95 percent confidence level.

If a given mass of dunite were completely serpentinized by this reaction, a volume increase of about 50 percent would accompany the serpentinization. However, because the Webster-Addie dunites attain a maximum of about 50 percent serpentinization, the corresponding overall volume increase in the ultramafic body would be less than this value. The lack of an obvious increase in the amount of magnetite with degree of serpentinization (as well as the absence of iron-rich serpentine) suggests that most of the iron in the olivine was lost in solution during serpentinization.

Thayer (1966) suggests that the presence of apparently undisturbed primary mineral banding in serpentinized ultramafic rocks is evidence for constant-volume serpentinization. Such primary mineral bands (chiefly of chromite) occur in the Webster-Addie ultramafic body and do not appear to be disrupted by progressive serpentinization. It appears possible that such bands can be preserved during "increasing-volume" serpentinization. For the chromite bands in the Webster-Addie body, it would require that the chromite grains move away from each other (keeping their euhedral shapes) as the band width increases during an overall volume increase. It should be possible to resolve this problem by tracing primary bands along strike as a function of increasing serpentinization. Until evaluated in such a manner, undisturbed primary mineral banding should not be used to support constant-volume serpentinization in ultramafic bodies.

The SiO₂ that appears to have been added to the dunites during serpentinization may have been derived from silica-rich gneisses and schists surrounding the ultramafic body. Existing data, however, are not sufficient to uniquely determine the source of the silica. Hostetler, *et al.* (1966) discuss the problem of adding silica during serpentinization and suggest that even small amounts of silica addition require large and perhaps unreasonable amounts of water. However, in view of the fact that little is known about (1) silica diffusion rates in serpentinizing waters; (2) the time span accompanying serpentinization; or (3) the amount of water available over that time span; their suggestion is open to question. The data presented above suggest that up to about 3 percent of silica was added during the serpentinization of the Webster-Addie dunites.

The data in Table 1 and Figure 1 also suggest that sodium was introduced into the Webster-Addie dunites during serpentinization. Although some sodium appears to have been introduced by late, granitic pegmatites which cross-cut the ultramafic body (Madison and Condie, in preparation), the correlation of Na-content with percent serpentine shown in Figure 1 suggests that some sodium (and probably other mobile trace elements) was introduced during serpentinization. Samples included in this study were not collected from parts of the body suspected of being contaminated with sodium by pegmatite fluids. Although the source of the sodium introduced during serpentinization remains unknown, upper crustal rocks which are enriched in alkali elements are logical candidates. These data reemphasize the fact that it is of critical importance to know in which phases trace elements are concentrated before making conclusions based on trace element distribution regarding the source of ultramafic rocks.

The absence of any Mg-rich (and to a lesser degree, Fe-rich) mineral assemblages (carbonates and/or silicates) around the border of the Webster-Addie ultramafic body suggests that magnesium and iron were either carried away in solution (perhaps along fractures) and/or that they "dribbled out" into adjacent county rocks in such small amounts as to go undetected (a mechanism suggested by Hostetler, *et al.*, 1966).

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