THE EFFECT OF URALITIZATION UPON THE CHEMICAL COMPOSITION OF THE SUDBURY NORITE[1]

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ABSTRACT

Chemical and mineral analyses of fresh and altered norites from Creighton township, and chemical analyses of the constituent ferromagnesian minerals, hypersthene, diallage, and uralite, show that little chemical change except hydration has taken place during the uralitization of the pyroxenes. The results also prove that a Rosiwal analysis of one thin section is not, in general, representative of the mineral composition of a medium-grained rock.

The Sudbury norite as the world's largest and most important nickel intrusive, has been the object of much chemical and mineralogical investigation, a large proportion of it carried out to provide evidence in the classical controversy over the theory of origin of the valuable ore deposits. Although most investigators recognized the severity of the widespread alteration in this rock, few apparently tried to obtain uniformly fresh or altered samples for analysis. This paper gives the results of an attempt to determine whether the process causing the commonest mineral change, that of the pyroxenes to fibrous amphibole—the "uralitization"—has materially affected the chemical composition of the rock and if so, in what way and to what extent. The solution of this problem may lead to a fuller understanding of the mechanism of uralitization in basic rocks in general, and may in particular provide a basis for evaluatng the worth of previous analyses of Sudbury norite specimens, and consequently of the theories of origin based upon these analyses. In addition, it may be possible to establish how carefully sampling must be done on this and similar intrusives to be truly representative.

The general nature of uralitization. The term "uralite" was first applied by Rose (1812) to a mineral with the crystalline form of a pyroxene and the structure of an amphibole. Dana (1892) states that the change begins on the outer surface of the pyroxene transforming it into an aggregation of slender amphibole prisms, parallel in position to each other and the parent pyroxene. When complete, the entire crystal is changed into a bundle of fibers. The most prominent changes in composition are those in lime and magnesia. Clarke (1924, p. 600) points out that the change is

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complicated in many cases by the simultaneous formation of epidote or zoisite. A separation of magnetite may occur and calcite may be formed due to the removal of lime. Generally augite alters to hornblende or actinolite, and diopside to tremolite.

The causes of uralitization have received only occasional attention. Duparc & Hornung (1904) suggested that, after the pyroxene had crystallized in the magma, it was acted on by the residual magma and the transformation occurred into patches of amphibole. In a later paper, Duparc (1908) said that the phenomenon of uralitization is confined chiefly to feldspathic rocks and he attributed it to a slightly later local injection of feldspathic magma. He cites cases in the Asiatic Urals where uralitization is caused by the injection of leucocratic dikes into pyroxenites. The common occurrence of uralitization as a deuteric alteration effect has been noted by Colony (1923). In the end stages of crystallization some of the quartz and the alkalies are concentrated in liquid form of low viscosity and great penetrating power. At this stage olivine and magnesia-rich pyroxenes are susceptible to alteration resulting in serpentinization or uralitization, the latter showing a powerful penetrating power by invading the feldspar in every direction. He also notes that these phenomena may alter the chemical and mineral composition of the rock. Schwartz (1939) stated that uralitization is a frequent product of hydrothermal alteration, but that it may occur during metamorphism of various types.

Previous analyses of the Sudbury norite and deductions from them. The first important investigation was carried out by Walker (1897) who analysed five samples from across the intrusive in the neighbourhood of the Blezard mine, and from the results concluded that there is a regular increase in acidity towards the centre of the intrusive. No other complete series of analyses was made until Knight (1923) analyzed seventeen samples from the intrusive in MacLennan township. From these he concluded that there was no gradual increase in acidity; in fact, "the most basic part of the eruptive is not along the outside, but from 1,500 to 2,100 feet from the outside edge." Phemister (1925) attacked the problem in a different manner by doing mineral analyses on samples from eleven traverses across the intrusive. He concluded that the norite of the south range is most basic from one-half to three-quarters of its total width of outcrop from the basal contact. Above this, it becomes more acid by the development of a mesostasis of quartz and alkali feldspar and beneath it by the presence of quartz, which is a late crystallization. Coleman, Moore, & Walker (1929), dissatisfied with the above and other conclusions, chemically analyzed samples from across four complete sections of the intrusive: the Capreol and Levack sections on the north range, and the Cameron Mine and Creighton on the south. Their conclusion was "that the eruptive sheet is a unit formed by the segregation of a magma into a more basic portion, norite, passing without a break upwards into a more acid phase, micropegmatite."

A point which all the above investigators appear to have overlooked is that the deuteric alteration, particularly the uralitization, may have considerably altered the bulk composition of the rock. Suppose, for example, that one exposure of fresh norite and another in which all the pyroxene and part of the feldspar have been altered to amphibole are found twenty feet apart, then which exposure should be sampled, or does it make any difference? If the samples are chosen at random, they will be in different stages of alteration, and may not give a true picture of the composition gradient. It may be necessary to obtain samples which are at least fairly uniformly altered.

Some of the discrepancies which *may* be due to this late alteration are found in the previously published analyses. For example, an analysis of a sample from forty-seven yards northwest of the Creighton mine gives only 0.69% MgO (Knight, 1917); this can hardly be typical norite. Another analysis of Knight's of a sample from 1,083 yards northwest of Creighton shows 1.56% MgO, but analyses of two samples straddling this one (Coleman *et al.*, 1929) show 5.51 and 6.62% MgO respectively.

Mineral analyses, too, may not be representative unless special precautions are taken to make them so, for the change in mode can be great without appreciably affecting the chemical composition. The method itself may introduce large errors as Larsen & Miller (1935) have emphasized: "Certain it is that many rocks, particularly coarse-grained rocks, are not well mixed on the scale represented by a thin section." This statement certainly applies to the norite in which the ferromagnesians have a strong tendency to cluster. Larsen & Miller found that Rosiwal analyses of thin sections from the same hand specimen varied by from 2 to 10% for the major constituents, and they warn that "the danger of basing far-reaching conclusions on a few Rosiwal determinations of specimens from widely scattered outcrops is evident." Phemister's work, however, cannot be criticized in this regard because all his results are shown by graphs, and there is no account of the number of samples or thin sections that he used.

EXPERIMENTAL RESULTS

Samples. The norite samples used in the present study were collected on Concessions 1, 2 and 3, Lot 2, Creighton township, about the centre of the norite zone. They were taken from below the zone of weathering and thus could not have been subjected to surface alteration.

General petrography. Unaltered Sudbury norite is composed essentially of plagioclase, hypersthene, diallage, and quartz or granophyre. The order of crystallization of these minerals is clearly shown by the textural relationships. Hypersthene was generally the first mineral to crystallize although in some sections it is later than plagioclase, indicating that the two minerals crystallized in part contemporaneously. The irregular shape of diallage and its habit of enclosing euhedral crystals of hypersthene and feldspar show that the diallage is later. Quartz, or a granophyric intergrowth of quartz and soda feldspar, or rarely microcline, was the last important mineral to crystallize, as shown by its habit of filling in between the larger minerals in characteristic triangular or irregular shapes, and by its corrosion of the adjacent feldspar.

Plagioclase occurs as euhedral laths of a distinctive brown color. It is sodic labradorite. The brown color of the feldspar is due to minute inclusions of ilmenite, hematite, or magnetite as indicated by an analysis reported by Coleman *et al.* (1929) showing it to contain 0.23% TiO_2 and 1.10% FeO. In places these inclusions can be seen in the plagioclase as minute black needles in small patches, forming a nearly right-angled grid structure. Hypersthene occurs in euhedral stubby prisms which are commonly clustered together, and has the refractive indices $\alpha=1.692$, and $\gamma=1.699$ (both ± 0.002). Diallage is present as large irregular grains, commonly as simple twins, and has the indices of refraction $\alpha=1.689$, $\gamma=1.710$ (± 0.002). Other primary minerals are hornblende, magnetite, apatite, and yellow sulphides. Primary hornblende is of minor occurrence, and is distinguished from secondary amphibole by its blue-green color and non-fibrous nature. The magnetite and apatite show evidences of both early and late crystallization, and they also show highly variable amounts in different sections, but these special problems were not investigated during the present study.

Alteration. The nature of the uralitic alteration is observed more easily in the hypersthene than in the diallage which has a lamellar structure controlling the amount and distribution of the fibrous material. In hypersthene the alteration always begins at the outer border and works progressively inwards by veining until only isolated remnants of the fresh pyroxene remain; in diallage, because of the structural control, most of the mineral may be altered and yet have partly fresh borders. Both hypersthene and diallage may alter directly to uralite or may pass through a bastitic phase with the separation of small grains of magnetite. Serpentine, the identification of which was confirmed by an x-ray powder photograph, is gray to light green in color and has a massive or aggregate structure. The uralite is pale gray to green in color with indices of refraction $\beta=1.631$, $\gamma'=1.643$ (± 0.002). As alteration intensifies, the uralite recrystallizes around the outer edges to a darker green, more pleochroic amphibole, and it may become mixed with many small biotite plates. With extreme alteration, this mixture or the uralite alone spreads out through the rock replacing the feldspar along grain boundaries and twinning planes or in irregular patches.

Alteration of the plagioclase is by no means as common or intense as that of the pyroxenes, but even in the freshest norite an incipient type of alteration occurs at the centre of the large feldspar grains. Rarely this alteration is intense and the large feldspar crystal loses its brown color and becomes packed with small crystals which resemble zoisite. With severe alteration, the feldspar may break down completely, lose its brown color, and the rock becomes a mixture of clear albite, epidote, and amphibole.

TABLE 1. CHEMICAL ANALYSES OF FRESH AND ALTERED NORITE

Constituents	Fresh	Altered
SiO_2	53.30	54.42
Al_2O_3	17.60	19.65
Fe_2O_3	2.88	1.87
FeO	7.13	6.07
CaO	9.05	8.74
MgO	6.06	5.15
Na_2O	2.58	2.80
K_2O	1.34	1.53
H_2O+	0.22	0.30
Total	100.16	100.53
S. G.	2.87	2.88

Analyses by T. A. Oliver.

The results in table 1 show that there is little chemical difference between fresh and altered norite; the differences are so small that they may be within the range of experimental error. Silica and alumina appear to be added in the altered sample, whereas there is an apparent decrease of all other constituents except the alkalies and water. These differences are slight, however, and may not be significant. The water content of both samples is surprisingly low, but redeterminations gave similar results.

Separation and analysis of hypersthene, diallage, and uralite. The differences in composition of the fresh and altered rock are small, but possibly some of the differences are real. To determine whether or not this is so, pure hypersthene, diallage, and uralite were separated from crushed rock and analysed. Two samples about fifteen feet apart, one fresh and the other altered, were chosen for the separation. Pure uralite was obtained with ease by separating the minus 35-plus 60 fraction in Clerici's solution. The first concentrate of uralite was purified by two further separations. By counting grains in index oil, the concentrate was found to be 97% pure. The separation of hypersthene and diallage was complicated by the fact that the specific gravity of diallage, about 3.28, is intermediate between that of hypersthene and uralite, about 3.37 and 3.04 respectively. Therefore, hypersthene partially altered to uralite has about the same specific gravity as the diallage, and these come down together in the separation. By repeated separations, however, about 1.5 grams of 98% pure hypersthene was recovered from the mixture, and about half a gram of 95% pure diallage was laboriously picked by hand under the binocular microscope.

Table 2. Chemical Analyses of Hypersthene, Diallage, and Uralite

Constituents	1	2	3	4
SiO_2	52.11	51.61	52.34	54.3
Al_2O_3	1.22	4.17	5.01	
Fe_2O_3	1.65		2.26	18.4
FeO	19.96	11.48	13.04	
CaO	1.63	16.09	6.98	8.4
MgO	22.16	15.49	15.06	18.1
K_2O			0.80	0.8
H_2O+			0.71	
Total	98.73	98.84	96.20	100.0

1. Analysis of hypersthene, close to $Mg_2FeSi_3O_9$.
2. Analysis of diallage, close to $Ca_2(Mg, Fe)_4Si_6O_{18}$.
3. Average of two analyses of uralite.
4. Average of uralite recalculated to 100% by taking into account addition of silica +alumina and water (including the deficit).

Analyses by T. A. Oliver.

The results of the chemical analyses of these three minerals are shown in table 2, along with the closely approximated chemical formulae of hypersthene and diallage. Only the main portion of these two minerals was completed, owing to the small amounts available. The analysis of uralite shows it to be roughly intermediate in composition between hypersthene and diallage. The first analysis of uralite showed a deficit of about 4%. This was considered to be an analytical error but a second analysis verified the first. The nature of this analytical deficit in amphibole has been discussed by Jakob (1937), who found that it is generally absent from asbestiform (one-dimensional) varieties but never from compact (three-dimensional) varieties. The deficit was once thought to be due to an excess of "molecular" oxygen but Jakob has shown and others have confirmed that it is actually caused by water given off above 1000° C. This water is more or less reduced to hydrogen by divalent titanium.

Calculations based on the analyses. It is now possible, by making simple calculations involving the compositions of hypersthene and diallage, and the ratio of hypersthene to diallage, to determine whether the uralitization was accompanied by any change in chemical composition. The composition of uralite theoretically calculated from the composition and proportions of hypersthene and diallage should agree with the composition of uralite found by chemical analysis if no changes in chemical composition accompany the uralitization. The ratio of hypersthene to diallage in the rock was determined by Rosiwal analyses of 13 thin sections and by several grain counts to be about 1.7 to 1, which means that 0.63 parts hypersthene plus 0.37 parts diallage form one part uralite.

Several assumptions and adjustments have to be made before making the calculations. If it is assumed that none of the constituents change except silica+alumina and water, then the other constituents must be adjusted to 100% before a true comparison can be made. The assumption that there is an actual gain in silica+alumina agrees with the results of the rock analyses, in which there was a total gain of about 3% in these constituents in the altered rock. In the uralite there is a gain of about 3.1% in silica+alumina assuming a hypersthene: diallage ratio of 1.7:1, and a gain of 4.5% in water assuming the whole deficit to be water, making a total gain of 7.6%. This amount is divided proportionally among total iron, lime, and magnesia in the uralite should be equivalent to those in the two pyroxenes when the ratio of these two minerals in the norite is taken into account if the above assumptions are valid. Using the above hypersthene:diallage ratio, uralite theoretically derived from hypersthene and diallage should contain 18.0% total iron whereas analyzed uralite adjusted to 100% contains 18.4% ferrous iron. Theoretically, the uralite derived solely from the hypersthene and diallage should contain 6.9% lime and 19.6% magnesia; actually it contains 8.4% lime and 18.1% magnesia.

If the above calculations are absolutely correct, it means that in the uralite formed from the pyroxenes by alteration, 0.4% total iron and 1.5% lime have been added, and 1.5% magnesia has been lost. However, when consideration is given the numerous sources of possible error, the separation of pure minerals, the chemical analyses themselves, and the determination of the hypersthene to diallage ratio, the differences are probably within the limits of error, and the results appear to indicate no marked change in composition with the process of uralitization.

Nature of the uralite. The secondary amphibole in the norite may have a constant composition or two distinct amphiboles may have been formed from the two pyroxenes. If there are two, then the hypersthene and diallage merely underwent hydration in the deuteric alteration, but if there is only one of constant composition then there must have been an interchange of calcium, magnesium, and iron. The optical properties do not afford much evidence. The uralite formed from hypersthene does appear different from that formed from diallage in slightly altered specimens, that derived from diallage having higher interference colors and being finer-grained than that derived from hypersthene. However, the lamellar structure of diallage may control the size of the alteration product. Further evidence suggesting two amphiboles is that the hypersthene seems to be much more easily altered than the diallage; some sections show much of the hypersthene severely altered whereas much of the diallage is still fresh. In severely altered norite, all of the uralite has the same appearance, suggesting a constant composition.

As mentioned earlier, hypersthene occurs in euhedral prisms whereas diallage has an irregular habit. In the early stages of alteration, the secondary amphibole retains these distinctive shapes. It was therefore possible to polish a section of rock and scrape out, with a diamond point, enough of the two types for *x*-ray powder photographs. Again the results were inconclusive; the data from both patterns are in substantial agreement with each other and with those given in the A.S.T.M. Card Index (1945) for actinolite (II–1908). Thus, no conclusion is possible concerning the comparative chemical compositions of the two uralites, the one derived from hypersthene, the other from diallage.

Conclusions

The conclusions based on the above work are as follows:

(1) The results of chemical analyses of fresh and altered norite show that, in the samples investigated, no important changes in the chemical composition of the rock occur as a result of the process causing uralitization. This fact was confirmed by chemically analysing hypersthene, diallage, and uralite separately; these results show that water and possibly a small amount of silica and alumina are added in the formation of uralite whereas the differences in the other constituents are within the limits of experimental error. However, no samples of norite were analysed in which the feldspar had been largely replaced by the uralite or late quartz, and such material may be radically altered in composition. Furthermore, there may have been a change in bulk composition due to the process of uralitization in other parts of the intrusive.

(2) A Rosiwal analysis of one thin section will not in general give reliable values for the mineral composition of a medium or coarse-grained rock.

(3) The process of uralitization in the Sudbury norite appears to be carried out by a residual liquid, possibly carrying small amounts of silica and alumina. In the first stages of the alteration, simple hydration of the pyroxene takes place, resulting in serpentinization or uralitization of the hypersthene and leaving the diallage relatively unaltered. With increasing alteration an interchange of calcium, magnesium, and iron between the two pyroxenes may occur, resulting in the formation of an amphibole of constant composition.

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