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VARIOLES IN ARCHEAN BASALTS: PRODUCTS OF SPHERULITIC CRYSTALLIZATION*

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Abstract

Petrography and geochemistry indicate that the globular structures (varioles) of numerous Archean basalts of the Abitibi Greenstone Belt result from spherulitic crystallization. Samples, taken across the chilled margin of little altered or deformed basalt, show a regular progression of plagioclase morphology with degree of undercooling. The texture changes from small, fibroradial spherulites near the quenched margin to coarser, more open types with increasing distance from the contact and, finally, to plagioclase dendrites. The present felsic composition of the varioles merely reflects the albitization of the former Ca-rich plagioclase. The distribution of rare earths between the varioles and their mesostases is incompatible with the hypothesis that these rocks represent quenched immiscible liquids. The depletion of Eu in some of the varioles and slight enrichment of the remaining *REEs* in others, with respect to their matrices, are probably due to alteration superimposed on *REE* concentrations that were presumably similar in both cases. This distribution reflects the reduced atomic mobility of the *REE* in the undercooled liquid and the fact that, during growth, spherulites entrap melt between the fibres.

Keywords: spherulite, variole, liquid immiscibility, rare earths, Archean, basalt, Abitibi.

Sommaire

Une étude pétrographique et géochimique détaillée démontre que les textures globulaires, que l'on appelle "varioles", courantes dans les basaltes archéens de la ceinture volcano-sédimentaire de l'Abitibi, résultent d'une cristallisation sphérolitique. Des suites d'échantillons, pris près des bordures figées des coulées basaltiques peu altérées ou déformées, illustrent une progression régulière de la morphologie du plagioclase en fonction du degré de trempe. La texture change, quand on s'éloigne de la bordure figée, de sphérolites fibroradiés très près du contact, à des sphérolites plus ouverts et, finalement, à des dendrites loin du contact. La composition felsique actuelle des varioles n'est qu'une conséquence de l'albitisation du plagioclase calcique primaire. La répartition des terres rares entre les varioles et leur mésostase est incompatible avec l'hypothèse que ces roches représentent des liquides immiscibles. Le léger enrichissement de certaines varioles en terres rares ainsi que l'appauvrissement en Eu, toujours par rapport à leur mésostase, sont probablement des effets d'une altération superposée à une distribution probablement semblable dans les deux phases. Cette distribution reflète une mobilité inefficace de ces éléments dans le liquide refroidi et le piégeage de la fraction liquide entre les fibres lors de leur croissance.

(Traduit par la Rédaction)

Mots-clés: sphérolite, variole, immiscibilité liquide, terres rares, Archéen, basalte, Abitibi.

Introduction

Gélinas *et al.* (1976) postulated that the round felsic masses (varioles) found in the Blake River Group basalts of the Rouyn – Noranda area of Quebec were formed as the result of liquid immiscibility. The ensuing discussions (Cawthorn 1977, Hughes 1977, Philpotts 1977) and a subsequent reply by Gélinas *et al.* (1977) proved the liquid immiscibility hypothesis to be controversial for these rocks (variolites) and left the question of their origin largely unresolved. However, Gélinas *et al.* (1976) did provide evidence that varioles formed directly from a liquid as a result of undercooling, and that their present-day composition is felsic relative to the adjacent matrix. These authors did not test the predicted liquid–liquid partitioning of trace elements formulated by Hess (1971). Interest in the origin of varioles remains strong because they are proximal to many major Archean gold deposits (*e.g.*, at the Dome mine, Timmins, Ontario). Recently, variolites have been used as evidence to support the concept of Archean bimodal volcanism (Thurston *et al.* 1985).

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The goal of the present study was to sample variolitic rocks in areas least affected by metamorphism and alteration and to report on the textural features and the distribution of trace elements, principally the rare earths. In parts of the Abitibi belt, the variolitic texture is unusually well preserved and ideal to test the liquid-immiscibility model of Gélinas *et al.* (1976). The textures reported here are best interpreted to be due to spherulitic crystallization directly from the melt. The geochemical data reflect crystal–liquid disequilibrium partitioning during rapid growth, and subsequent mobility of elements under conditions of relatively mild alteration. We conclude that the varioles were produced by rapid crystallization and underwent later albitization, which results in their present-day felsic composition.

Nomenclature

In our experience, the term *variole* has been inconsistently used; it covers a variety of different meanings, both observational and genetic. Phillips (1973, after Lasaulx 1875) defined varioles as "consisting of radiating fibres of feldspar occurring in fine-grained basaltic rocks, particularly in the tachylitic margins". Lofgren (1974, after Johannsen 1939) defined varioles as "spherical bodies appearing on the weathered surfaces or marginal portions of certain diabases. They are composed of various secondary minerals and are intimately intergrown with the rock. Often they are spherulites". Lofgren (1974) added that the term *variole* "has been inappropriately extended to include spherulites of all kinds in all basic rocks". According to the AGI Glossary (Bates & Jackson 1980), a variole is "a pea-sized spherule, usually composed of radiating crystals of plagioclase or pyroxene" generally found in mafic igneous rocks, and a variolite is an aphanitic or fine-grained igneous rock containing varioles. The glossary defines a spherulite as a "rounded or spherical mass of acicular crystals, commonly feldspar, radiating from a common point". In contrast, MacKenzie *et al.* (1982) considered varioles as fan-like arrangements, commonly branching, composed of plagioclase. They stated that varioles differ from spherulites in that they are not discrete identifiable spherical bodies. In this paper we retain the usage of Lofgren (1974, after Keith & Padden 1963) for spherulites: they are radially symmetrical arrays of crystalline fibres having the same fibre axis, each fibre having a crystallographic axis in a slightly different orientation from its parent. As Lofgren (1974) pointed out, they need not be spherical, as they do form conical arrays, sections through which yield fan-like shapes. They are further divided into fine and coarse depending on fibre diameter, and open if there is visible foreign material between fibres, and closed if not. In this paper, the term *variole* will be used in the most liberal sense to refer to megascopic globular masses. A *variolite* is a rock containing varioles.

Geology

Samples for the study were collected from numerous sites within the western part of the Archean Abitibi greenstone belt between Rouyn–Noranda, Quebec, and Timmins, Ontario (Fig. 1). In this area the supracrustal rocks are preserved in a large east-trending synclinorium (MERQ-OGS 1983). The volcanic rocks result from three major cycles of volcanism that began with komatiitic volcanism and culminated with calc-alkaline and alkaline volcanism (Jensen & Langford 1985). The third cycle is the best preserved and accounts for most of the rocks exposed in the synclinorium, whereas those of the first two cycles (the Lower Supergroup) are intruded by granitic rocks, poorly preserved, and restricted in their occurrence to the periphery of the synclinorium. Only rocks of the youngest (third cycle) were sampled in this study.

Volcanic rocks of the third cycle consist of four distinct groups. The Stoughton–Roquemaure and Larder Lake Groups, at the base, are composed of flows of komatiitic and tholeiitic basalt; the proportion of the latter increases toward the top of the sequence. Above both komatiitic successions are rocks of the Kinojevis Group, composed of magnesium-rich and iron-rich tholeiitic basalt flows. This group has a pronounced upward iron-enrichment trend and contains volumetrically minor dacite and rhyolite toward the top of the sequence. Calc-alkaline basalt, andesite, dacite and rhyolite of the Blake River Group discordantly overlie the Kinojevis Group. Finally, alkaline to subalkaline trachytic volcanic rocks of the Timiskaming Group unconformably overlie both the Blake River and Kinojevis Groups.

The volcanic rocks have been regionally metamorphosed in the prehnite–pumpellyite facies (Jolly 1978). Locally the rocks have been metamorphosed to the greenschist facies and higher grades of contact metamorphism near late granitic plutons and rhyolitic centres of volcanism.

Two major east–west-trending systems of faults, the Destor–Porcupine and the Kirkland Lake – Larder Lake – Cadillac fault zones (breaks), transect the north and south limbs of the synclinorium, respectively. Close to the fault zones, the volcanic rocks have subparallel schistosities; elsewhere, penetrative deformation is weak or nonexistent.

Variolites were collected from flows of tholeiitic basalt located in the Stoughton–Roquemaure, Larder Lake, Kinojevis, and basal Blake River Groups. Sample sites include areas close to the major fault-

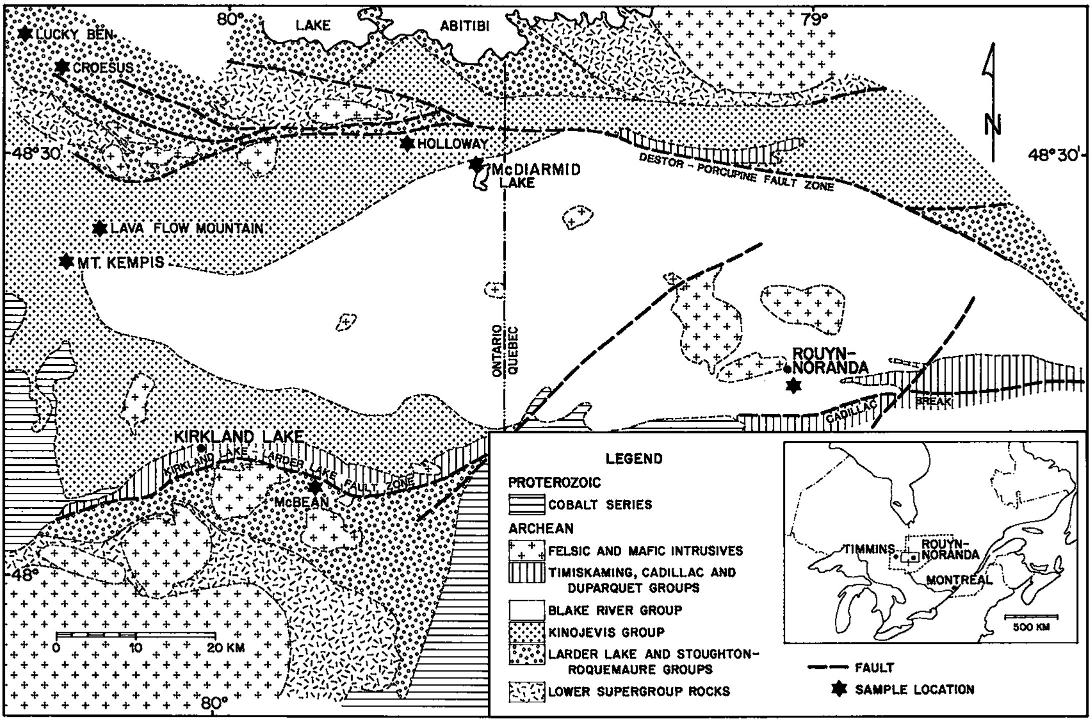

FIG. 1. Geological map of the central Abitibi belt, showing the locations discussed in text.

zones as well as undeformed areas, and areas of subgreenschist metamorphism as well as those of greenschist and higher-grade metamorphism (Fig. 1).

APPROACH

To test the hypothesis that the globular textures were produced by a process of liquid immiscibility, we sampled numerous well-preserved variolitic basalts. Hess (1971) showed on theoretical grounds that incompatible elements such as the *REE* should be partitioned strongly into the mafic phase of a pair of immiscible liquids, relative to the felsic phase. The felsic liquid is more polymerized than the mafic, that is, the structure of the felsic liquid consists of a three-dimensional network of linked silicon–oxygen and aluminum–oxygen tetrahedra. The linkages in this network are provided by the bridging oxygen atoms, each bound to two atoms of silicon (or other tetrahedrally co-ordinated cation). A mafic liquid, having a lower Si/O ratio, contains fewer of these networks and is relatively depolymerized with respect to a felsic liquid. As a consequence, with two-liquid equilibrium, the mafic liquid will contain more free oxygen and more nonbridging, singly attached oxygen atoms than the felsic liquid. Therefore, highly charged cations such as the *REE*, which cannot substitute for silicon in tetrahedra, will tend to concentrate in the more mafic phase. Here they can co-ordinate with oxygen in order to form polyhedra. Watson (1976) confirmed the predictions of Hess (1971) by using trace-element-doped charges in the system K_2O–Al_2O_3–FeO–SiO_2. He found that the *REE* tend to concentrate in the mafic fraction relative to the felsic fraction by a factor of about four and that the partitioning increases with field strength (charge/radius).

Ryerson & Hess (1978) showed that the *REE* preferentially partition into a ferropyroxenitic liquid relative to a granitic liquid by a factor of ten and that the liquid–liquid distribution coefficients of the *REE* studied (La, Sm, Dy, Yb) are all equal within experimental error. Clinopyroxene in equilibrium with the granitic liquid and with the ferropyroxenitic liquid shows partition coefficients for the *LREE* of approximately 0.7 and 0.07, respectively, and for the *HREE* of approximately 12.0 and 0.2, respectively. Ryerson & Hess (1978) argued that because 1) the slopes of the two clinopyroxene spectra are parallel, and 2) the liquid–liquid partition coefficients of the *REE* are equal, the observed enrichment in *HREE* is due to selectivity by the mineral and not the two liquids. In other words, cations of high field-strength become less soluble in polymerized melts relative to depolymerized melts, causing minerals in the polymerized liquid to incorporate them more successfully. Ryerson & Hess (1978) also suggested that divalent Eu would not partition into mafic liquids to the same degree as the trivalent *REE*; this yields a mechanism to produce or modify Eu anomalies during liquid–liquid equilibria.

Thurston & Fryer (1983) interpreted *REE* spectra for two variole - matrix pairs from the Uchi-Confederation greenstone belt (Ontario) to be indicative of liquid immiscibility. Considering analytical uncertainty, there is no real difference between the varioles and the adjoining matrix. However, these authors considered the varioles to be the product of immiscibility and argued that the slight enrichment of the *REE* (10%) in the more mafic phase is due to reduced liquid–liquid partitioning because the bulk composition of some of their rocks is andesitic.

In contrast, Vogel & Wilband (1978), in their studies of natural systems having textures thought to represent immiscible liquids, have largely corroborated the experimental results. They showed an enrichment of *LREE* from five to ten times in the mafic phase relative to the granitic pegmatite phase of the composite dykes studied. However, the degree of enrichment decreased with decreasing field-strength. Clearly, if the variolitic rocks in this study represent quenched immiscible liquids, the varioles should show a comparable depletion of their *REE* content relative to their mafic matrix; this is the opposite of that expected from rocks generated by ordinary crystal–liquid equilibrium.

METHODS

Samples containing globules were sawed and handpicked prior to grinding in an agate mortar. The analyses for the *REE*, U and Th were performed at the Institut de Génie Nucléaire, Ecole Polytechnique, Montréal, using the general technique of Gordon *et al.* (1968). No unusual γ-ray interferences were encountered in the course of this study, and the overall error of the analytical data, based upon replicated analyses of U.S.G.S. standard rocks, is estimated to be within 5 to 10%. The *REE* data were chondrite-normalized using the data of Anders & Ebihara (1982). The analyses of Rb, Sr, Y and Zr were performed on a Philips X-ray-fluorescence spectrometer at the Université du Québec à Chicoutimi and rely on the general technique of Sumartojo & Paris (1980), which uses the Compton scatter of Mo as a measure of the mass-absorption coefficient of the sample. The error in the Rb and Sr data, based upon replicate analyses of U.S.G.S. standard rocks, is estimated to be less than 5%. Similarly, that of the Zr and Y is estimated to be less than 10%. The X-ray-

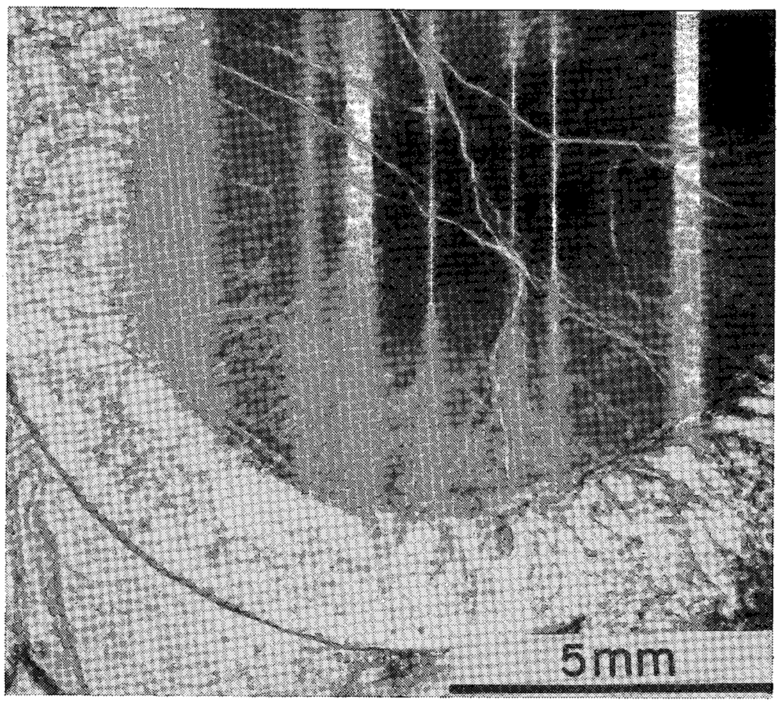

FIG. 2. Sample from McDiarmid Lake hyaloclastite adjacent to flow contact, showing a fine, fibroradial spherulite and chlorite-rich alteration rim. Plane-polarized light.

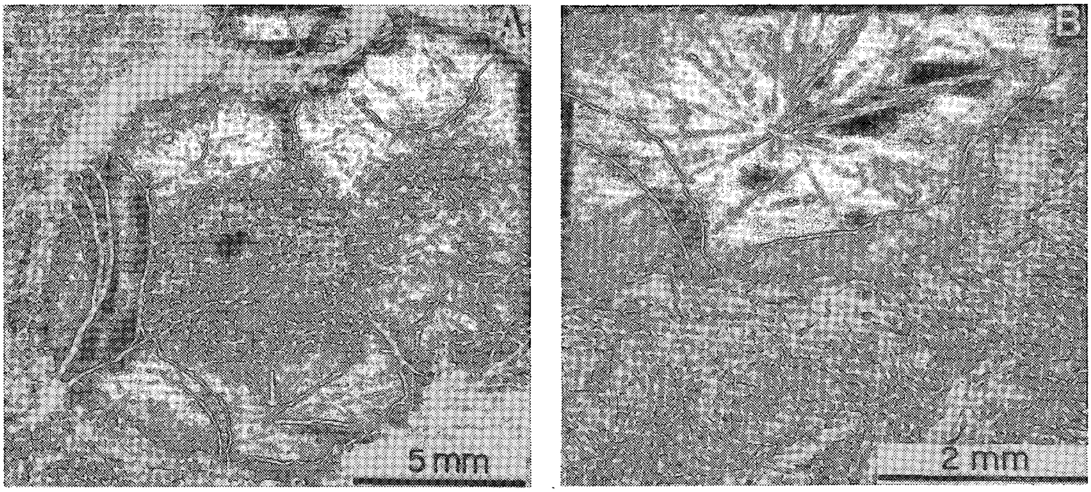

FIG. 3. A. Coarse fibroradial spherulite from within flow; the central leucocratic area is dominated by fibroradial plagioclase spherulites and clusters of opaque iron oxides. B. The periphery is composed of prominent bladed augite coarse spherulites with demonstrable nuclei; the intervening area is composed of plagioclase spherulites and iron oxides. Note the spherulitic nature of the matrix. Plane-polarized light.

diffraction studies were carried out at the University of Ottawa using a Philips diffractometer and a quartz internal standard.

PETROGRAPHY

The basalts occurring near the top of the Kinojevis Group at McDiarmid Lake (Fig. 1) are part of a volcanic unit with a strike length of greater than 60 km (Dimroth *et al.* 1973, Jensen 1978); the unit is characterized by its variolites. In the vicinity of McDiarmid Lake, the rocks are in the prehnite–pumpellyite metamorphic facies and virtually undeformed. Alteration, here interpreted as pre-Kenoran, is expressed locally by the development of chlorite, albite, epidote, and very minor carbonate. In this area, the variolitic unit is about 950 m thick and consists of interlayered variolitic and nonvariolitic flows. On the northwestern side of McDiarmid Lake, variolitic flows 0.5 to 1.5 m thick separated by thin beds of hyaloclastite up to 10 cm thick were studied and sampled. An adjacent nonvariolitic massive flow greater than 10 m thick was also sampled. At this locality the textural preservation of the varioles was found to be much better than their more altered counterparts south of Rouyn–Noranda and elsewhere. Based upon their mineralogy, morphological characteristics and mode of occurrence, the globular textures that occur in the thin flows at McDiarmid Lake can be divided into two broad groups, fine and coarse fibroradial. The fine type (Fig. 2) occurs within hyaloclastite near the flow boundary, in autobrecciated altered glass or within the flow close to its boundaries. This type consists of a single array of plagioclase fibres radiating from a common point. The coarser type occurs deeper within the flows and consists of an inner zone of fine fibroradial and branching plagioclase surrounded by an outer zone of coarse plagioclase and coarse spherulites of clinopyroxene. Both types commonly attain sizes of a approximately 1 cm in diameter.

The fine fibroradial globules (Fig. 2) are spherical and composed of numerous radiating fibres of low albite. They are concentrically zoned, iron oxide stained, and have increasing amounts of chlorite toward their extremities. In hyaloclastite, a globule's contact with the host generally is sharp, and demarcated by an abrupt change in texture and a thin selvage of chlorite + quartz. The matrix to such globules, where they occur within the flow, is composed of fine-grained wispy branches of plagioclase and augite in part replaced by masses of incipiently polygonized quartz, turbid albite and chlorite. Close to their borders, some globules have a very indistinct polygonization that is secondary to the dominant fibroradial texture. Clearly, these globules are fine, closed spherulites because of their spherical fibroradial habit, small size of fibre and lack of visible foreign material between fibres.

The coarser type of globule (Fig. 3A) contains individual domains of coarse radial pyroxene intergrown with branching plagioclase and pyroxene surrounding fibroradial plagioclase cores. These globules are roughly spherical in shape but have an irregularly indented surface in comparison to the fine fibroradial spherulites described above. Apparently, fibroradial plagioclase grew first and controlled the overall spherical form, whereas later growth of

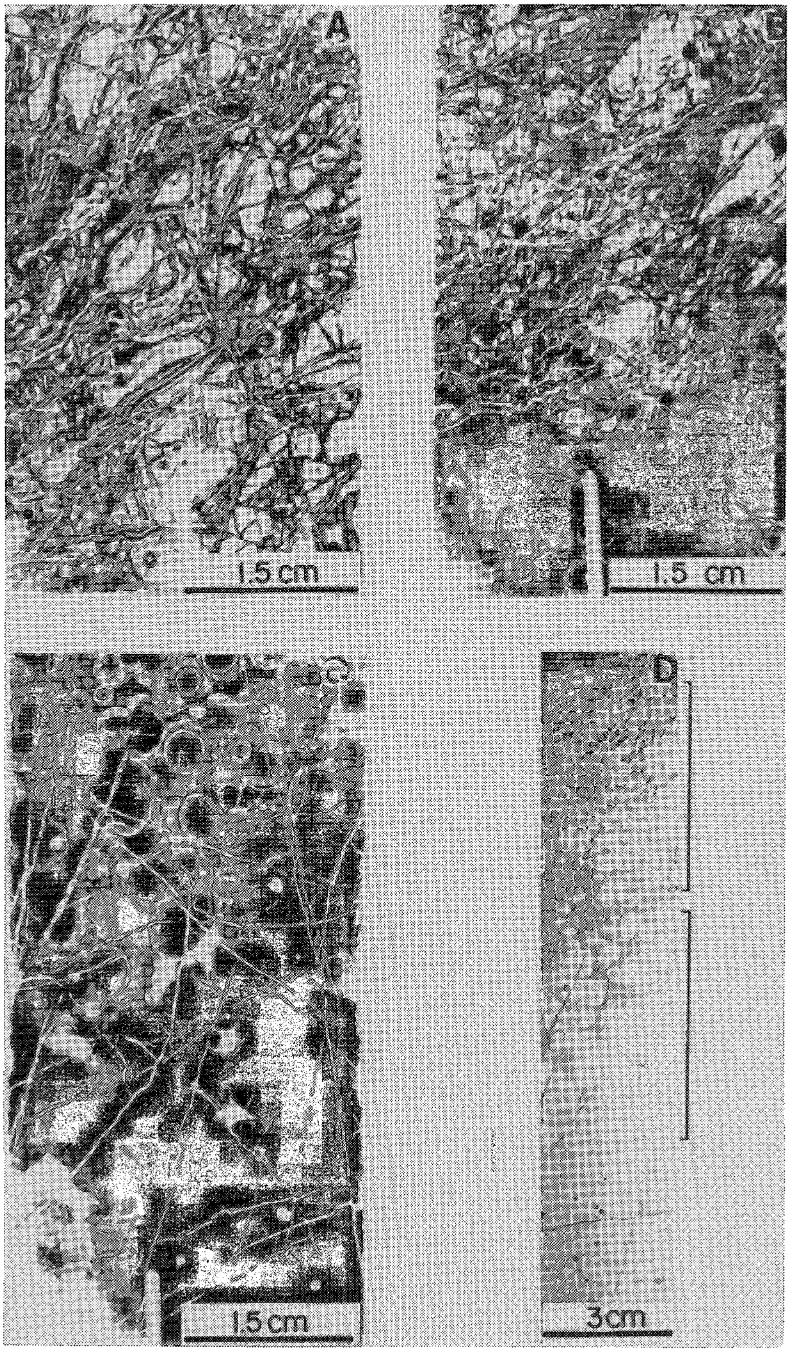

FIG. 4. Photographs of whole thin-sections of core, and photograph of core from Holloway Township, showing transition from hyaloclastite to *in situ* breccia to holocrystalline rock. The brackets in D illustrate the original position of B and C in the core, B being above C. Section A was contiguous with the original core, and was cut from directly above B. The saw marks indicate the bottom direction. Sections A and B show the transition from hyaloclastite to *in situ* breccia. Note the absence of globules at the top of A, and their progressive appearance and increase in size with depth. C. Variolite composed almost completely of coalesced fibroradial plagioclase spherulites. Figure 4D also shows the progressive increase in the amount of globules with depth; the small black dots are amygdules.

coarse pyroxene spherulites and coarse plagioclase spherulites produced the surface irregularities. Figure 3B illustrates coarse, radial pyroxene spherulites in which the individual pyroxene fibres nucleated on a pre-existing crystal and grew in both the globule and the matrix. Note that the matrix texture is very similar to that of the spherulites, the former being dominated by branching plagioclase and pyroxene that are approximately half the size of the latter. In places the globules appear more melanocratic than their hosts despite the fact that they are more felsic in composition. This anomaly is due to the presence of iron oxide. Like the fine fibroradial type, the coarse spherulites are partly chloritized close to their extremities and are surrounded by a thin selvage of chlorite + quartz + epidote at the contact with the matrix. The matrix to these is composed of relatively coarse branches of plagioclase and augite containing minor amounts of epidote and some patches of polygonized albite.

Samples from the nearby large flow contain no varioles but are composed of a fine-grained mass of subequant plagioclase and indistinct dendritic plagioclase, and ill-formed laths and fan spherulites of clinopyroxene with secondary epidote and calcite. These rocks have a porphyritic radiate texture (Lofgren *et al.* 1975).

A 40-cm-long core obtained from the lower part of the Kinojevis Group in Holloway Township, Ontario (Fig. 1) provided an excellent opportunity to study the distribution of varioles as a function of cooling (Fig. 4). The core transects hyaloclastite (23 cm of which are not shown in Fig. 4D), through *in situ* autobrecciated altered glass to globule-bearing basalt (Figs. 4A to C). Globules first appear as 1–2 mm spheres in the autobrecciated zone within 5 cm of the basalt and increase in size to approximately 2–3 mm over this interval (Figs. 4B, C). They are composed of spherical fibroradial plagioclase spherulites and, where appropriately cut, have nuclei that appear to be plagioclase microlites. Some of the nearby domains of translucent, almost isotropic, altered glass contain plagioclase microlites. A small minority of the globules have irregular outlines, where two or more have grown into one another. In no case were the well-preserved fragments of hyaloclastite, which are up to five times the size of the globules, found to have spherulites. This absence, combined with the fact that the spherulites first appear associated with the altered glass only near the flow contact in the autobrecciated portion, argues against their formation prior to extrusion.

Within the crystallized portion of the core below the former glass, the spherulites have coalesced and form approximately 95% of the rock. In general the following progression of plagioclase morphologies is found with distance from the cooling contact: fine, closed spherulites (Fig. 5A) in the autobrecciated altered glass and near its contact with the holocrystalline rock, followed by progressively coarser, more open spherulites with branching (plumose) terminations (Figs. 5C,D,E) and finally dendritic plagioclase (Fig. 5F). The spherulites have commonly nucleated on plagioclase microlites (Figs. 5B,D,E). The rock contains numerous amygdules; it is evident that the gas phase exsolved at about the time that the globules formed because in many instances apophyses of spherulitic lava project back into the vesicles, and in at least one case, a spherulite nucleated on a vesicle. This has been documented by Phillips (1973) and is the result of localized cooling due to the energy required to form a bubble.

Samples were also taken of the variolitic Kinojevis basalts that crop out at the summit of Mt. Kempis in Cooke Township, Ontario. The globules occur in pillow basalt and tend to be confined to within 15 cm of the periphery in large (1.5 × 0.5 m) pillows or distributed throughout small (0.5 × 0.5 m) pillows. Petrographic analyses of thin sections taken from the border into the core of a whole pillow retrieved from this locality again show plagioclase in fine, closed, spherical fibroradial spherulites, some with a nucleus, grading into more open and coarser spherulites and finally into plagioclase dendrites with distance from the selvage. The samples from Mt. Kempis were not analyzed because we wished to avoid the effects of alteration caused by a nearby granodiorite stock; the core described earlier was not analyzed because there was not enough matrix to sample.

At the remaining localities (Fig. 1), we were unable to relate the variolites to pillow or flow boundaries owing to the small size of the outcrops or severe deformation and alteration. However, we sampled these localities to detect textures indicative of their origin.

Samples taken from basalts just south of Rouyn–Noranda contain globular-textured material from what Gélinas *et al.* (1976) described as a series of thin flows within the Blake River Group. The flows in this area are strikingly similar to those of the Kinojevis Group in Ontario. The rock is composed of a fine-grained mass of albite, chlorite and quartz cut by a network of small (1 mm) veins containing the same minerals. The globules, although highly altered and deformed into shapes with elliptical cross-sections, do retain relict spherical fibroradial spherulites.

Globules sampled from the Kinojevis Group of Lava Flow Mountain, Cook Township, Ontario, range in diameter from about 1 to 3 cm, are circular in section, and have rather convoluted outlines. As noted by Jackson (1980), the varioles tend to increase in size with distance toward the centre of individual pillows. Unfortunately, the samples collected have a rather strong fabric, and primary textures are

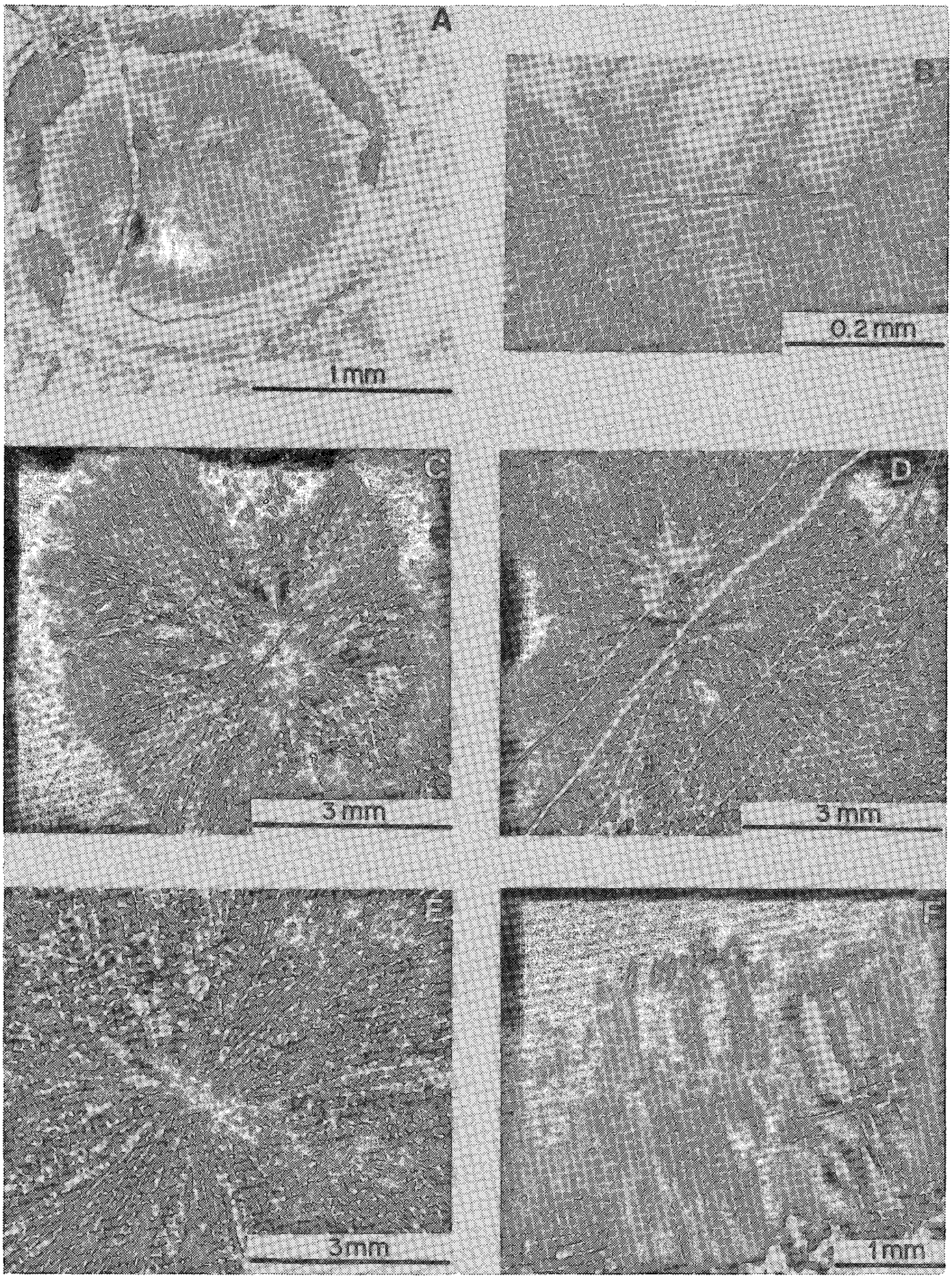

FIG. 5. Photomicrographs of textures in core from Holloway Township. A. Single fine, fibroradial spherulite, in autobrecciated altered glass located 1.5 cm from the bottom of slide shown in Figure 4B; plane-polarized light. B. Axiolitic growth of plagioclase fibres on nucleus of A; plane-polarized light. C. Within variolite, coarser spherulite illustrating development of plumose (branching) texture with growth; crossed nicols. D. Same view showing nucleus and fan-shaped inner portion of the spherulite; plane-polarized light. E. Deeper within variolite; coarse, fibroradial plagioclase spherulite with plagioclase nucleus; crossed nicols F. Plagioclase dendrites; crossed nicols.

difficult to discern. The globules are leucocratic, composed of felted plagioclase and polygonized quartz with aligned laths of chloritized augite, all of which are cut by stringers of quartz and carbonate. The matrix is composed of chlorite, augite and carbonatized plagioclase. Relict branching plagioclase and pyroxene needles can still be observed in the globules.

In Gauthier Township, Ontario, similar globules occur in the Larder Lake Group about 400 m south of Inco's McBean open-pit gold mine, 15 km east of Kirkland Lake. Local shearing has obscured the contact relationships, and the rocks have been strongly affected by the intrusion of lamprophyric and syenitic magmas. We have been unable to detect flow contacts or pillows and, therefore, have been unable to relate the varioles to contacts. The varioles are prominent in that they have coalesced to masses on the order of tens of centimetres across. These weather white, in strong contrast to their grey matrix. In one globule, chain and "wagon-wheel" augite (Lofgren 1980) is well preserved; elsewhere the varioles are similar to those of the Lava Flow Mountain area and consist of hornblende after augite, plagioclase and calcite.

Near the Croesus gold mine, Munro Township, Ontario, globular textures occur in highly carbonatized basalt close to the contacts of what appear to be thin flows. Primary textures are all but totally obliterated, with the exception of what appear to be several spherical fibroradial spherulites associated with a highly altered hyaloclastite matrix.

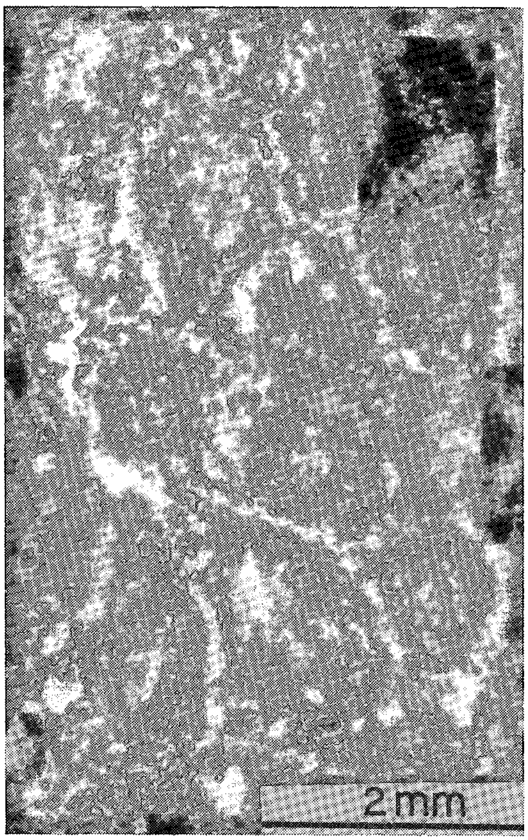

FIG. 6. Photomicrograph of orb texture interpreted to be due to devitrification, Lucky Ben prospect. Plane-polarized light.

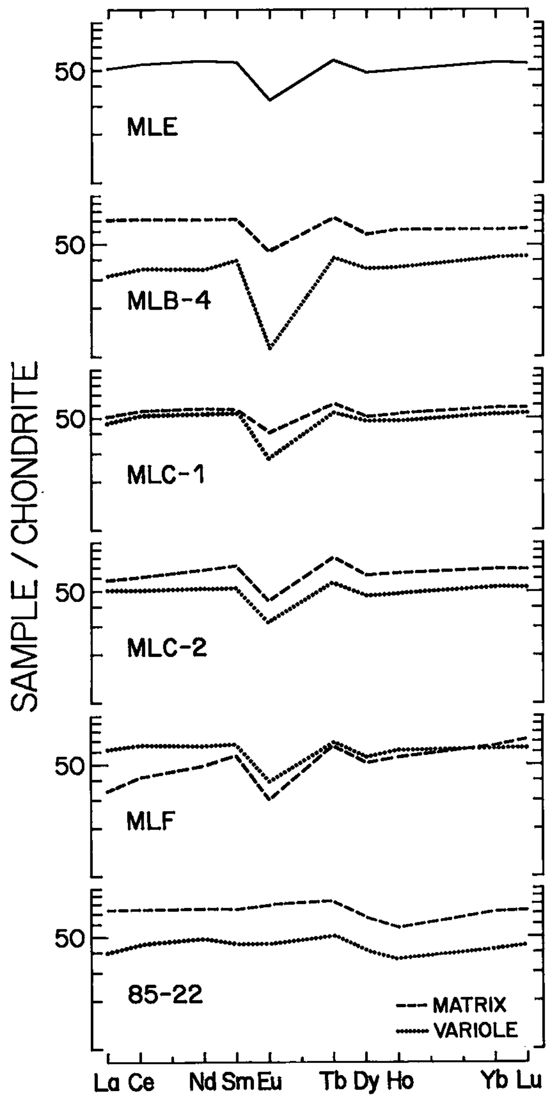

FIG. 7. The normalized *REE* patterns. MLC-1, variolite from transition of hyaloclastite to holocrystalline rock, McDiarmid Lake. MLC-2, variolite from deeper in same flow. MLB-4, variolite from transition of hyaloclastite to holocrystalline rock of a different flow. MLE, sample from the massive flow, McDiarmid Lake. MLF, variolite from Rouyn - Noranda. 85-22, variolite from Lava Flow Mountain.

At the Lucky Ben prospect, Munro Township, Ontario, both large (0.5 cm) and small (0.1 mm) globules are found in a highly carbonatized, chloritized and silicified metabasalt. For the most part, the globules have no recognizable internal textures, owing to the alteration; however, some of the large ones have relict, coarse plagioclase spherulites with a central nucleus. Other large globules are a random arrangement of spherulitic plagioclase intergrown with and partly obscured by secondary quartz and carbonate. The small globules (Fig. 6) appear to be identical to the type described by Lofgren (1971) as orb textures, produced during experimental hydrothermal devitrification of rhyolite glass at

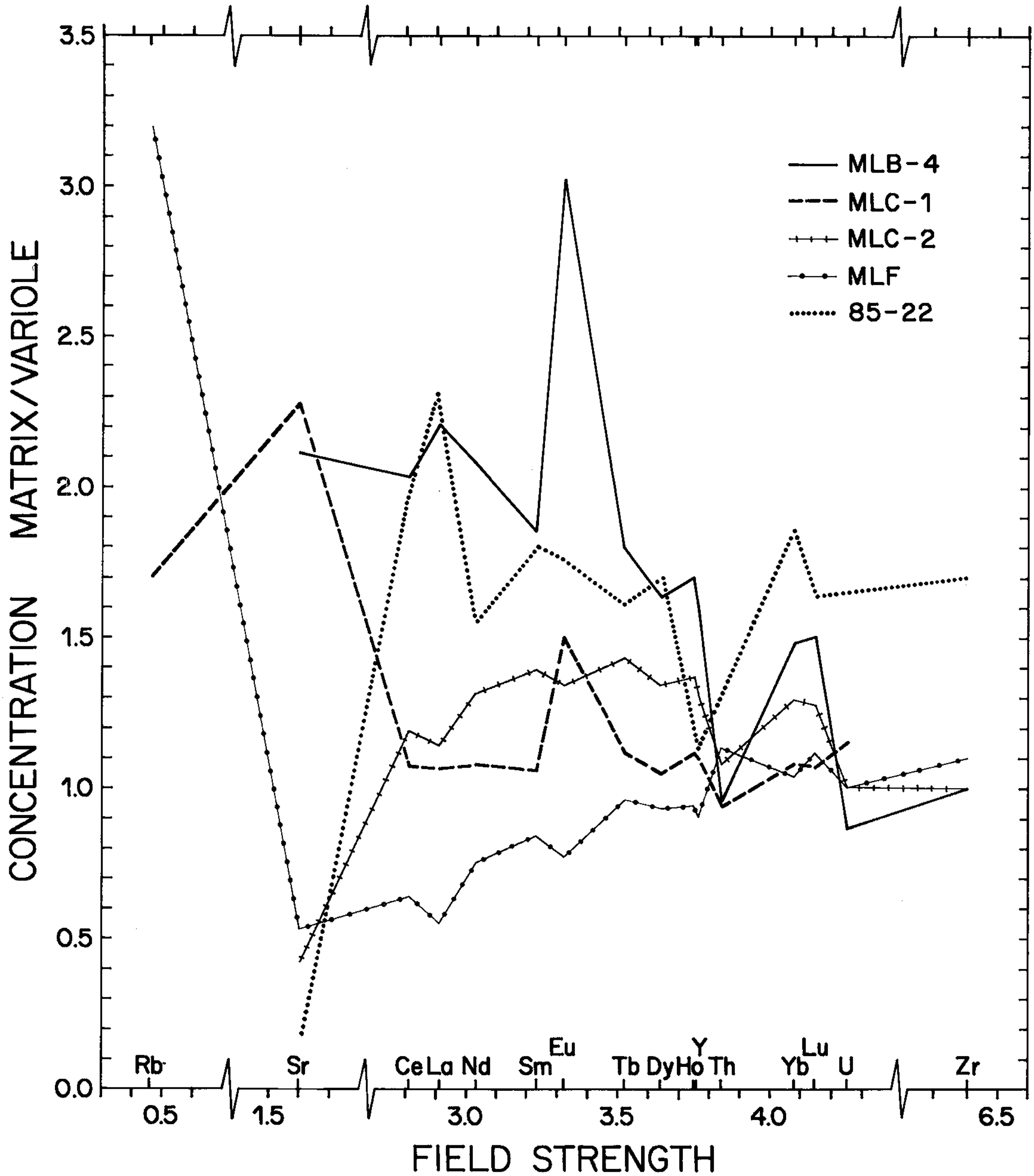

FIG. 8. Matrix/variole ratio *REE,* Rb, Sr, Y, Zr, U and Th *versus* field strength.

elevated temperatures in the presence of an alkaline aqueous fluid. We interpret the large globules to represent spherulitic growth from the liquid and the smaller ones as devitrification spherulites.

Geochemistry

Figure 7 and Table 1 present the chondrite-normalized *REE* spectra and trace- and major-element data for the three variole - matrix pairs (MLB-4, MLC-1, 2) and the massive flow from McDiarmid Lake (MLE), and one pair each from the vicinity of Rouyn-Noranda (MLF) and Lava Flow Mountain (85-22). Note that all the *REE* spectra are rather flat, with some evidence of *LREE* depletion, suggesting earlier minor partial melting in their source region. The samples show a rather prominent negative Eu anomaly, except for the Lava Flow Mountain samples (85-22). With the exception of the Rouyn-Noranda data (MLF), there is a slight depletion of all *REE* in the varioles compared to their matrices. Figure 8 shows data for the *REE*, Sr, Rb, Y, Zr, U and Th plotted in the form of the matrix/variole concentration ratio *versus* field strength. Note that in two samples (MLB-4 and MLC-1), Eu is significantly more abundant in the matrix than in the variole.

In terms of the major elements (expressed as oxides), the varioles are enriched in SiO_2 and Na_2O (approximately 12 and 3%, respectively), and depleted in Fe_2O_3 (total), CaO and MgO [approximately 6, 2 (excluding MLC-2) and 3%, respectively] in comparison to their matrix.

Discussion

In light of the above observations, the hypothesis that the globules represent a quenched immiscible liquid requires re-examination. We are in agreement with Gélinas *et al.* (1976) to say that the globular textures of the metabasalts have developed directly from a melt and that they are not a product of devitrification. However, as nucleation is a random process, we find it highly improbable that crystallization of an immiscible droplet would result in a spherical array of radial fibres emanating from the droplet's centre. One would expect a more random distribution and orientation of crystals during the rapid cooling of two liquids. Indeed, the chondrules of chondritic meteorites, which have a spherical shape, are generally throught to result from the undercooling of liquid droplets. These globules clearly have randomly arranged or noncentred spherulitic textures (Blander *et al.* 1976). Also, the common observation

TABLE 1. GEOCHEMICAL DATA ON VARIOLE-BEARING ARCHEAN METABASALTIC ROCKS

	MLC1M	MLC1V	MLF1M	MLF1V	MLC2M	MLC2V	MLB4M	MLB4V	MLE	8522M	8522V
SiO_2	53.22	62.22	45.95	58.89	44.74	60.27	44.83	56.87	54.36	34.24	73.23
TiO_2	.98	1.08	1.47	1.20	1.21	1.07	1.12	1.07	1.10	2.28	1.30
Al_2O_3	14.46	14.31	15.19	14.22	14.06	15.52	16.36	13.50	14.03	12.48	10.17
Fe_2O_3	11.42	8.07	17.42	9.95	17.76	8.07	15.14	10.10	11.46	31.70	4.65
CaO	8.31	4.25	6.60	5.69	3.89	5.59	4.75	3.02	5.01	7.60	4.37
MgO	5.55	4.20	7.45	3.54	10.22	4.04	9.28	6.32	5.67	5.30	.91
Na_2O	1.04	4.07	2.70	4.95	.55	4.47	1.78	3.26	2.56	.46	4.22
K_2O	.02	.02	.63	.32	.07	.03	.02	.03	.03	.09	.07
MnO	.13	.09	.15	.07	.22	.09	.17	.11	.12	.49	.08
P_2O_5	.43	.24	.34	.30	.20	.29	.25	.16	.26	.98	.57
H_2OT	3.62	2.15	2.57	1.27	6.10	1.96	5.91	4.15	4.78	-	-
La	12.1	11.3	8.1	14.6	13.8	12.1	16.5	7.5	11.8	17.3	9.2
Ce	34.0	31.6	26.1	41.1	37.4	31.4	44.5	21.9	34.0	45.0	28.0
Nd	25.9	24.0	22.3	29.7	31.4	23.6	32.6	15.7	26.2	34.0	22.0
Sm	8.3	7.8	8.4	10.0	10.7	7.7	10.9	5.9	8.4	11.2	6.8
Eu	2.3	1.6	1.7	2.2	2.5	1.8	2.6	.6	1.9	4.4	2.5
Tb	2.1	1.9	2.3	2.4	2.8	2.0	2.6	1.5	2.1	2.9	1.8
Dy	12.3	11.7	12.6	13.6	15.4	11.5	14.2	8.7	12.1	16.0	10.0
Ho	2.9	2.6	3.1	3.3	3.6	2.6	3.4	2.0	2.8	3.2	2.0
Yb	9.1	8.4	10.3	9.9	10.9	8.5	9.9	6.7	9.0	11.3	6.7
Lu	1.4	1.3	1.7	1.6	1.7	1.3	1.6	1.1	1.4	1.8	1.1
U	.22	.19	.26	.26	.27	.27	.15	.17	.26	-	-
Th	.89	.95	1.37	1.21	1.03	.95	.82	.85	1.04	-	-
Rb	1.2	.7	16.0	5.00	-	-	-	-	-	-	-
Sr	225	100	112	210	52	123	95	45	100	15	115
Y	84	78	84	95	106	79	101	63	84	63	56
Zr	211	219	271	237	225	222	201	200	223	353	212

The major oxides are reported in weight percent, whereas the trace elements are in ppm. Suffixes M and V denote matrix and variole separates, respectively. MLC1 and MLB4 are both from near the contacts of thin flows at McDiarmid Lake. MLC2 samples are from the middle portion of a thin flow. MLF samples are from what appears to be the contact of a thin flow south of Rouyn-Noranda. The samples 85-22 are from a pillow margin from Lava Flow Mountain. Sample MLE is from the thick non-variolitic flow at McDiarmid Lake.

of nuclei composed of plagioclase microlites in the centre of the spherulites argues against the two-liquid hypothesis.

Philpotts (1979, 1982) showed that liquid immiscibility does occur and is common in quartz-normative basalts. However, he described globules within glass that are on the order of micrometres in diameter. We found no evidence of these small-scale globules in polished section. This could be due to alteration, even though relatively mild in our McDiarmid and Holloway samples.

The slopes and abundances of the *REE* data in the varioles and matrix are not at all similar to either those predicted from theory or observed in experimental and natural silicate systems showing immiscibility. As described above, the *REE* should be enriched by a factor of at least four in the matrix relative to the varioles, and the partitioning should increase with field strength if they are the products of two-liquid systems. On the contrary, the patterns tend to converge at the *HREE* end. The maximum difference between the variole–matrix pair (except for Eu), is 2.2 in the case of La in sample MLB–4, a rock containing fine fibroradial plagioclase spherulites immediately adjacent to hyaloclastite, and 2.3 for the sample from Lava Flow Mountain (Fig. 8). MLF–1, a sample from Rouyn-Noranda, shows a maximum difference of 0.6 at La; however, the matrix is depleted relative to the globules. Both MLC–1 and MLC–2, from the contact and middle portion of a thin flow, respectively, show relatively uniform *REE* patterns (Fig. 8), with the matrix being only slightly (1.3 ×) enriched. Eu is depleted in the globule of sample MLC–1 and strikingly so in MLB–4 relative to their respective matrix, whereas the middle-flow sample (MLC–2) has no Eu depletion. Eu may be divalent and, as such, substitutes for Ca in silicates such as plagioclase. If divalent in a two-liquid system, the ion would have a lower field-strength than the remaining *REE* and hence partition less strongly into the mafic phase. This is exactly opposite to what is observed in the spectra. The similar overall *REE* abundances of the varioles and their matrix and the textural relationships documented above require that some alternative hypothesis to liquid immiscibility be postulated.

Philpotts (1977) argued that the presence of central nuclei and many other textural features of the Archean variolites are inconsistent with an origin by liquid immiscibility. He found the textures to be more reminiscent of those expected through the formation of spherulites during devitrification. Similarly, Dimroth & Lichtblau (1979) interpreted the varioles to be large spherulites.

Lofgren (1974, 1980) reproduced spherulitic textures in basaltic systems, and demonstrated that a variety of textures can result, depending on the degree of superheating and supercooling of a melt. He showed that spherulites can grow directly from a melt where there is a high degree of supersaturation, which occurs when nucleation is suppressed until the phase is well below its liquidus temperature (*i.e.*, undercooled). Melts that cool rapidly from a temperature above the liquidus essentially bypass equilibrium crystallization at the liquidus because of the large incubation-time required for nucleation. Spherulites grow quickly once nuclei have formed and represent the case where crystal growth is rapid relative to the rate of diffusion in the melt. Lofgren (1974) showed that there is a progression of textures to be expected, depending on the degree of undercooling, from fine, close fibroradial, to coarse, open spherulites with larger fibres, to dendrites, to elongate skeletal crystals and, finally, to tabular crystals (thin perpendicular to 010) in the plagioclase system between compositions of An_{20} and An_{70}. In contrast, for compositions more sodic than An_{15}, the skeletal and dendritic crystals do not form, and the habit varies directly from spherulitic to tabular depending on the degree of undercooling. Lofgren (1980) also found that pyroxene shows a similar progression from spherulitic, to dendritic, to skeletal, to elongate and, finally, to equant forms with decreasing amounts of undercooling.

The petrographic data presented above for the three cases in which we were able to relate the development of the varioles to cooling surfaces are entirely consistent with the observations of Lofgren (1974). Where the degree of undercooling was highest, in what was formerly glass, there are no spherulites. Spherulites are associated with the devitrified glass only close to flow boundaries. They are fine, spherical fibroradial spherulites that commonly appear to have nucleated on small, preexisting crystals. Deeper within the flows, these give way to coarser, more open, branching (plumose) plagioclase spherulites or spherulites with a fibroradial core surrounded by branching plagioclase and radiating blades of pyroxene and, finally, to plagioclase dendrites. Natland (1978) documented a similar progression of textures related to the degree of undercooling from cored basalts of the Mid-Atlantic Ridge. He described varioles as mats of extremely fine-grained plagioclase needles and skeletal olivine crystals that "appear to be centres of nucleation which formed at slightly lesser undercoolings than the extreme edges of the cooling units".

We interpret the varioles to be spherulites formed from melts erupted from shallow reservoirs of superheated magma, thereby eliminating nearly all nuclei. Rapid cooling after eruption reduced the rate of formation of nuclei, as diffusion slowed owing to increased viscosity. The commonly observed nuclei in the centre of some spherulites indicate that the nucleation was heterogeneous, probably on preexisting feldspar microphenocrysts. Undercooling stabilized spherulite formation and inhibited diffusion necessary to form new nuclei. Exsolution of a gas phase may have helped to form the spherulites by raising liquidus temperatures and viscosity locally.

Roedder (1979) has suggested that nuclei within melts might have caused to the formation of both immiscible globules and, subsequently, spherulites. However, this is incompatible with the observed relationship between the textures and the degree of undercooling and the fact that the associated hyaloclastite contains no globules. Also, the fact that a plagioclase of composition more sodic than An_{15} (in the system $An–Ab–H_2O$: Lofgren 1974) does not form dendrites may indicate that the globules were not initially felsic in composition.

Hughes (1977) concluded that the major-element data of Gélinas *et al.* (1976) are a reflection of alteration processes and not liquid immiscibility. The rocks were albitized, probably on the sea floor or shortly after their formation during burial metamorphism, because for the most part they lack a penetrative metamorphic fabric. They contain albite and epidote, have a spilitic composition (Table 1) and, judging by the vast amount of tholeiitic lavas in the area (Jensen & Langford 1985), were probably originally quartz-normative tholeiites. The globules were originally rich in calcic plagioclase and were metasomatically altered to their present-day low albite during the spilitization. Their albite-rich nature explains their enrichment in Si, Na, and depletion in Ca; a comparison of their composition by Gélinas *et al.* (1976) to a low-K rhyolitic liquid is thus fortuitous.

The relative depletion of the plagioclase-rich varioles in Eu and their slight enrichment in the other *REE* are interpreted as being due, in part, to alteration. Fowler & Doig (1983) have shown that Eu is preferentially mobilized with respect to the other *REE* during albitization; Hellman & Henderson (1977) concluded that spilites have increased *REE* contents relative to their unaltered precursors. Also, Michard *et al.* (1983) reported that the hydrothermal waters sampled from vents of the East Pacific Rise are highly enriched in the *LREE* and have large positive Eu anomalies (Eu/Eu* = 10). However, without a detailed knowledge of the assemblages of unaltered minerals, it is difficult to discern the extent of *REE* mobility because, as shown by Humphris *et al.* (1978), the extent of *REE* loss from any particular basalt depends to a large extent on its mineralogy and, therefore, crystallization history. The sample of the massive flow described from McDiarmid Lake has a *REE* concentration almost identical to that in the globules and their matrix. This rock probably did not suffer extensive *REE* loss as it is massive, *i.e.*, not permeable, and it contains significant secondary epidote, which can accommodate the *REE* in its structure.

As the *REE* are relatively large trivalent cations, they were probably unable to diffuse away from the rapidly growing spherulites in the undercooled liquid during crystallization. Therefore, their distribution reflects liquid–crystal disequilibrium partitioning. This feature, and the fact that some melt is trapped between fibres during spherulite formation, explain the similar *REE* concentrations within the spherulites and their matrix. The *REE* data for the variolites most likely represent values close to the original liquid, modified only by subsequent alteration.

The samples obtained from Lava Flow Mountain and near the McBean mine differ from those discussed above. They contain equant grains, dendrites and spherulites of augite. We were neither able to relate the textural development to cooling surfaces nor could we convincingly demonstrate that undercooling was the cause of the globules. However, the distribution of the *REE* in the one relatively fresh sample (85–22) is clearly incompatible with the two-liquid hypothesis. Lacking more conclusive evidence, we interpret variolites there to represent extrusion and subsequent undercooling when clinopyroxene was on the liquidus.

Conclusions

The data demonstrate that varioles within Archean tholeiitic basalts are spherulites produced by crystallization in response to undercooling of the liquid during cooling. Varioles are not found in the associated abundant, large, and relatively unaltered fragments of hyaloclastite. They are only found in autobrecciated glass or hyaloclastite within a few centimetres of flow contacts. However, within a flow they show a consistent morphological progression from fine, closed plagioclase spherulites to coarse, open spherulites, to plagioclase dendrites, from the contacts into the flows. They are analogues of komatiites inasmuch as they owe their textural development to supersaturation brought about by supercooling. Their textures and the distribution of high-field-strength elements are inconsistent with their development through immiscibility. The present-day felsic composition of the varioles results from the albitization of what was once Ca-rich plagioclase. Therefore, models that rely upon the presence of varioles as indicators of liquid immiscibility in order to produce the perceived great abundance of bimodal Archean volcanic suites (*e.g.* Thurston *et al.* 1985) should be re-examined, as data from this part of the Abitibi Belt do not support that concept.

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