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RELATIONSHIPS OF COTICULE GEOCHEMISTRY TO STRATIGRAPHY IN THE PERRY MOUNTAIN AND MEGUNTICOOK FORMATIONS, NEW ENGLAND APPALACHIANS

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Abstract

Coticules (garnet-rich quartzites) are chemically distinctive lithologies of controversial origin. They generally occur in pelitic schists, amphibolites, cherts, and quartzites as thin layers or discontinuous lenses in Paleozoic formations in the New England Appalachians and other orogenic belts. Geochemical data were collected to characterize coticules and their host rocks from the Middle Silurian Perry Mountain Formation of southern New Hampshire (near Rochester) and western Maine (near Rangeley), and the Cambrian Megunticook Formation of coastal Maine (Camden and Calais). Relative to their host rocks, the coticules are enriched in Fe, Mn, and P, and depleted in Ti and alkalis. Electron-microprobe analyses of garnet show higher spessartine and lower almandine components relative to garnet in the host. Coticules are light-*REE*-enriched and heavy-*REE*-depleted. They have concentrations between 10 and 100 times those of chondrites, generally lack a Ce anomaly, and have a negative Eu anomaly. Major-element compositions and *REE* profiles argue strongly against a hydrothermal origin for these rocks, and do not obviously support a hydrogenous (diagenetic) origin such as Mn–Fe formation. Rare-earth-element patterns differ as much within a local area as for widely separated samples within a single formation. All three samples from the Megunticook Formation are nearly identical, supporting stratigraphic correlation. But they are also like some of the Perry Mountain Formation samples, so that coticule compositions for a formation are not unique.

Keywords: coticules, bulk composition, rare-earth elements, Perry Mountain Formation, Megunticook Formation, Maine, New Hampshire.

Sommaire

Les coticules (quartzites grenatifères) sont des roches ayant une composition distincte et une origine controversée. On les trouve en général dans des séquences de schistes pélitiques, d'amphibolites, de cherts, et de quartzites, en minces couches ou en lentilles discontinues dans les formations paléozoïques des Appalaches de la Nouvelle-Angleterre et ailleurs dans des ceintures orogéniques. Des données géochimiques ont été prélevées afin de caractériser les coticules et leurs roches-hôtes de la Formation de Perry Mountain (âge silurien moyen) du sud du New Hampshire, près de Rochester, et de l'ouest du Maine, près de Rangeley, ainsi que la Formation de Megunticook (cambrien), de la partie côtière du Maine, près de Camden et de Calais. Par rapport à leurs roches-hôtes, les coticules sont enrichies en Fe, Mn, et P, et appauvries en Ti et en alcalins. Les analyses du grenat à la microsonde électronique montrent une teneur plus élevée en spessartine et plus faible en almandin que le grenat de la roche-hôte. Les coticules sont enrichies en terres rares légères et appauvries en terres rares lourdes. Leurs concentrations en terres rares varient entre 10 et 100 fois les teneurs chondritiques. En général, les coticules sont sans anomalie en Ce, mais elles montrent une anomalie négative en Eu. Les compositions en termes des éléments majeurs et les profils des terres rares montrent clairement qu'une origine hydrothermale ne peut être retenue, de même qu'une origine "hydrogénée" (diagénétique), comme c'eest le cas pour une formation de Mn–Fe. Les profils en terres rares d'échantillons d'une seule formation diffèrent autant au sein d'une région restreinte que sur une plus grande échelle. Les trois échantillons de la Formation de Megunticook sont presqu'identiques, ce qui pourrait encourager une application pour fins de corrélation stratigraphique. Mais ils ressemblent aussi aux échantillons de la Formation de Perry Mountain, ce qui démontre que la composition des coticules n'est pas distincte dans une formation particulière.

(Traduit par la Rédaction)

Mots-clés: coticules, composition globale, terres rares, corrélation, Formation de Perry Mountain, Formation de Megunticook, Maine, New Hampshire.

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Introduction

Coticules, or garnet-rich quartzites, are distinctive lithologies of controversial origin. The term *coticule* is primarily a field term used to describe fine-grained garnet-rich quartzites. Such rocks are typically richer in manganese than average pelite or shale, and thus commonly contain spessartine. They occur in sequences of pelitic schist, amphibolite, chert, and quartzite as thin layers or lenses in formations of Cambrian to Devonian age throughout the New England Appalachians and into Newfoundland. Individual coticule-bearing formations are continuous or nearly so along strike for distances up to 350 km.

Historically, coticule geochemistry has not received significant attention in the literature. Early papers primarily focused on detailed field and petrographic descriptions, with few data on mineral compositions and essentially none on whole rocks. Whereas most authors would agree that manganese is derived from a sedimentary precursor, a variety of protoliths have been proposed: Mn-rich sandy layers (Clifford 1960), impure manganiferous chert layers (Renard 1878, Emerson 1898, Schiller & Taylor 1965, Eusden *et al.* 1984, Thompson 1985), Mn-carbonate concretions (Woodland 1939, Bennett 1989, Schreyer *et al.* 1992), and manganiferous sediment with volcanogenic affiliations (Kramm 1976, Lamens *et al.* 1986, Krosse & Schreyer 1993).

In this paper, I present new major- and trace-element geochemical data for coticules from the middle Silurian Perry Mountain Formation of southern New Hampshire and western Maine and the Cambrian Megunticook Formation of coastal Maine. Results of electron-microprobe analyses of coticule and host-rock garnet also are presented. The goals of this paper are to evaluate (1) major- and trace-element compositions and rare-earth element (*REE*) patterns for these coticules and their possible use in protolith determination, and (2) the possibility of using these data as tools for stratigraphic correlation of coticules over short and long distances.

Background

Although bulk geochemical analyses of coticules have received considerable attention in the literature (Kramm 1976, Docka 1985, Lamens *et al.* 1986, Bennett 1987, Wonder 1987, Spry & Wonder 1989, Lottermoser 1989, Spry 1990, Schreyer *et al.* 1992, Krosse & Schreyer 1993), there is a paucity of published data on their *REE* concentrations. Based on fairly scant major and *REE* geochemical data at the time, Docka (1985) concluded that the Mn–Fe metasedimentary units studied in New England were derived from a mixture of hydrogenous sediment (*e.g.,* ocean floor Mn–Fe nodules and pavements) and deep-sea pelagic sediment (*e.g.,* open-ocean sediments free of a coarse clastic fraction).

Spry (1990) compared *REE* patterns of coticules associated with metamorphosed massive sulfide deposits from Georgia (Wonder *et al.* 1988) and the Willyama complex of Australia (Lottermoser 1989) with those of hydrothermal, hydrogenous, and mixed-provenance manganiferous sediments. He also considered those coticules from New England that are not associated with base-metal mineralization (Docka 1985). In his study, Spry demonstrated that coticules have very distinctive chondrite-normalized *REE* patterns. He concluded that coticules were derived from a mixture of hydrothermal sediment (e.g., such as at present-day ocean-floor spreading centers) with pelagic sediment. In a more recent investigation, Spry *et al*. (2000) suggested that the contribution of detrital material to coticules spatially and temporally associated with hydrothermal ore deposits lies between 30 and 70%.

Coticules are typically light-*REE* enriched, heavy-*REE* depleted, and show a negative Eu anomaly. Rare-earth-element concentrations of coticules are typically 10 to 100 times those of chondrites. With the exception of a slight positive Ce anomaly in coticules from New England investigated by Docka (1985), most coticules show an absence of a Ce anomaly. A positive Ce anomaly is typically taken as evidence of a hydrogenous origin; purely hydrothermal sediments are characterized by a negative Ce anomaly (Bonatti 1975). Marchig *et al*. (1982) also noted that chondrite-normalized *REE* patterns of hydrogenous and hydrothermal sediments exhibit distinct trends and that these differences can serve as geochemical discriminants between the two.

The data suggest that most coticules considered by Spry (1990) have *REE* concentrations that lie within the region for hydrothermal sediments (typically between 20 and 200 times those of chondrites), but well below the region for hydrogenous sediments (typically between 100 and 1000 times those of chondrites). Spry (1990) also noted that the compositions of coticules overlap those of pelagic sediment.

It is clear that more *REE* analyses of coticules are necessary to improve any understanding of sedimentary precursors for these distinctive lithologies. Furthermore, *REE* patterns of individual coticules may be used in making long-range geochemical correlations between rocks considered as stratigraphic equivalents on the basis of lithological similarities.

General Geological Setting

The New England samples of coticules investigated here are from the Merrimack synclinorium in southern New Hampshire and western Maine and from within the St. Croix belt in Maine (Fig. 1). The Merrimack synclinorium is a large-scale structure associated with the Devonian Acadian orogeny, related to the final closure of the Iapetus Ocean in a continent–continent collision. The Merrimack synclinorium contains Silurian and Devonian sediments shed southeastward from the Bronson Hill anticlinorium. The resulting sedimentary rocks and pre-Silurian basement units were intensely deformed and metamorphosed during the Acadian orogeny (~410–360 Ma). The St. Croix belt is a geologically diverse belt of Silurian–Devonian shallow marine clastic and minor volcanic rocks built on older metavolcanic and metasedimentary rocks (Osberg & Berry 1991).

Coticule-Bearing Formations Studied

Perry Mountain Formation

Coticule-bearing samples of the middle Silurian Perry Mountain Formation were collected from the southeastern limb of the Merrimack synclinorium near Northwood Narrows and Center Strafford, New Hampshire (Rochester area; distal to source region) and the northwestern limb of the synclinorium near Coos Canyon, Maine (Rangeley area; proximal to source region). The stratigraphic sequence includes the Silurian Rangeley, Perry Mountain, Smalls Falls, and Madrid formations and the Lower Devonian Carrabassett Formation (Maine) or Littleton Formation (New Hampshire) (Hatch *et al*. 1983, Eusden & Lyons 1993). This well-known stratigraphic sequence is characterized by thinly to thickly interbedded quartzites and pelites with

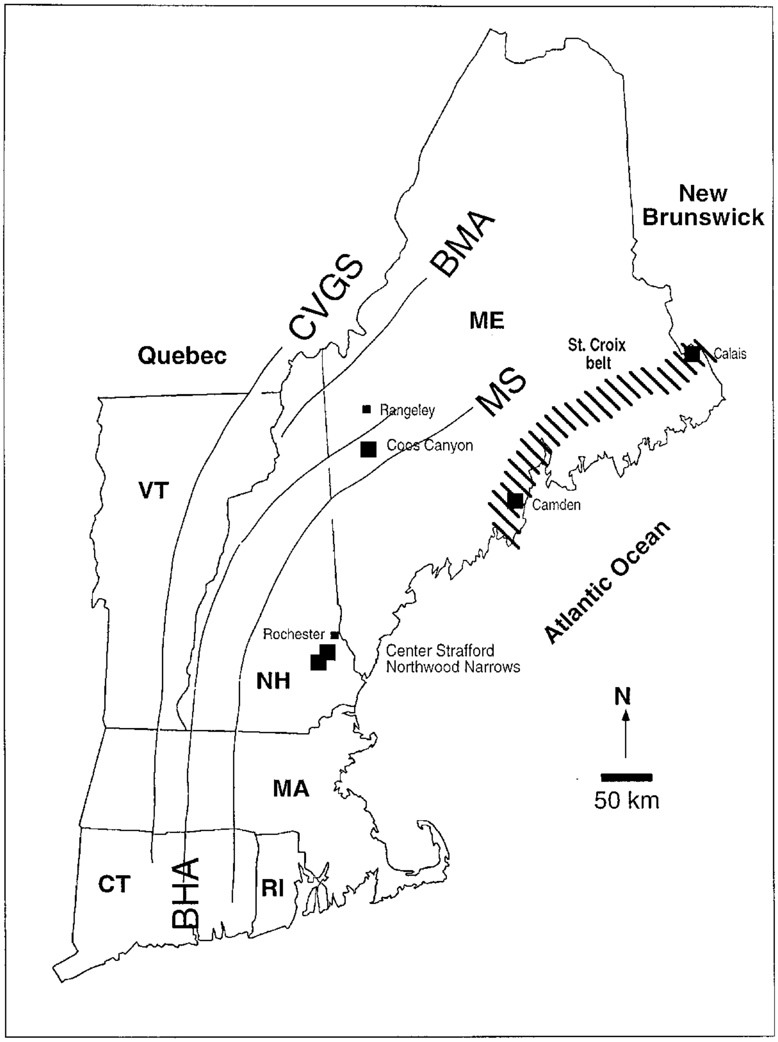

FIG. 1. Selected tectonic features in New England and sample localities. Symbols: MS Merrimack synclinorium, BHA Bronson Hill anticlinorium, BMA Boundary Mountain anticlinorium, CVGS Connecticut Valley – Gaspé synclinorium. Patterned area: St. Croix belt.

local metaconglomerates, as well as interbedded calcareous biotite granofels and quartzite and the distinctive metamorphosed black shale of the Smalls Falls Formation, which was deposited in anoxic conditions (Guidotti 2000).

The upper Perry Mountain Formation near Northwood Narrows (Stop 2 of Eusden 1988) and Center Strafford (Stop 7A of Eusden *et al.* 1984), New Hampshire consist of well-bedded gray garnet–staurolite schist and coticule-bearing quartzite. The two localities are approximately 6 kilometers apart along strike. The Perry Mountain Formation in the Coos Canyon area of Maine consists of coarse garnet–staurolite schists and coticule-bearing fine-grained, micaceous quartzite (Guidotti 2000).

Three distinctive types of coticules were observed within the Perry Mountain Formation localities (Fig. 2): (1) pink-colored lenses or pods (10–20 cm long), which are difficult to collect, (2) laterally extensive but discontinuous horizons of pink-colored coticules of variable thickness (2–20 cm), and (3) dark-colored (black) coticules found as thin (to 2 cm), laterally extensive, and commonly complexly folded horizons. The laterally extensive horizons of coticule are parallel to bedding. Samples collected from the Northwood Narrows locality include both folded type-3 horizons and some type-1 coticules; at Center Strafford and Coos Canyon, the coticules are of type 2 and variable in thickness.

Metamorphic grade of Perry Mountain Formation samples

The Perry Mountain Formation in the southern New Hampshire localities and the western Maine locality are polymetamorphic. Three events have been recognized by Eusden *et al.* (1984) in New Hampshire, including an early low-pressure facies-series event (M1) followed by contact metamorphism near igneous bodies (M2), and a later retrogressive (M3) event. The samples from near Center Strafford may exhibit contact metamorphic effects (M2) from intrusion of a nearby diorite. Metamorphism attained the amphibolite facies (sillimanite–muscovite grade) (Eusden 1988).

Guidotti (1970, 2000) and Guidotti *et al.* (1991) have suggested a polymetamorphic history for the western Maine locality as well. A low-grade (M1) metamorphic event was followed by a second event (M2) that brought the rocks up to staurolite – andalusite – biotite grade. Guidotti *et al.* (1991) and Guidotti (2000) have shown that a later staurolite-grade metamorphism (M3) was superimposed on the staurolite – andalusite – biotite grade metamorphism (M2), resulting in the formation of coarse muscovite after andalusite as well as minor chlorite.

Megunticook Formation

Coticule-bearing samples of the Cambrian Megunticook Formation were collected from three localities over 200 kilometers apart along strike (Fig. 1). In central coastal Maine, samples were collected from the Camden Hills area (one site in Camden and one in Lincolnville Center) from the St. Croix lithotectonic belt (Berry & Osberg 1989), a fault-bounded block of metamorphosed Precambrian to Ordovician stratified rock. The Megunticook Formation in these localities typically consists of gray quartz – muscovite – biotite – andalusite – garnet schist and quartz – plagioclase – biotite granulite (Osberg & Berry 1991). It also locally contains abundant layers (generally <5 cm) of pink to orange coticule (Fig. 3a). The Megunticook Formation is overlain by the Cambrian–Ordovician Penobscot Formation, the base of which is marked by pillow basalts dated as latest Cambrian (Osberg & Berry 1991). The Penobscot Formation is a rusty weathered, black to gray quartz – muscovite – phlogopite – sulfide schist (Osberg & Berry 1991) formed from sediments deposited in an anoxic environment. Samples from Lincolnville Center (CM–4) are from the top of the Megunticook Formation, just below the Penobscot contact (H.N. Berry, pers. commun., 1995).

Rocks considered to be stratigraphic equivalents to the Megunticook Formation in the Camden Hills area are exposed along the St. Croix River in Calais, Maine (Ludman 1978). These rocks, of limited exposure, were originally assigned to the Cambrian–Ordovician Cookson Formation, but are now considered to correlate with the Cambrian Megunticook Formation (Allan Ludman, pers. commun., 1995) as defined by Berry & Osberg (1989). The host rocks to the coticules here consist of dark, argillaceous quartzite with small pink garnetiferous pods and lenses scattered throughout (Fig. 3b).

Metamorphic grade of the Megunticook Formation samples

The rocks in the central coastal Maine area (Camden Hills) are polymetamorphic, with two prograde events and local retrogression to white mica + chlorite (Berry 1986). Pseudomorph textures are evident. The low-pressure metamorphism that affected these rocks attained andalusite–sillimanite grade. Samples from Camden (CM–3) are from near the southeastern edge of the Silurian Youngtown granite, which has associated muscovite–tourmaline pegmatites.

The rocks in Calais, Maine are found within a complex epizonal contact aureole involving at least four episodes of pluton emplacement including a gabbro and three granites. The argillaceous host-rocks contain cordierite that formed as a result of contact metamorphism, but the local regional grade is greenschist facies (chlorite zone) (Ludman 1978). At any rate, the rocks in Calais were metamorphosed at lower grades than those in Camden Hills.

FIG. 2. Distinctive types of coticules observed within the Perry Mountain Formation. (a) Lenses or pods (Northwood Narrows). (b) Discontinuous layers (Center Strafford). (c) Laterally extensive, folded horizons (Northwood Narrows).

SAMPLE DESCRIPTIONS

Perry Mountain Formation

Northwood Narrows: Four samples of dark, laterally extensive coticules (Type 3; 1–1.5 cm thick) and three samples of pink, lenticular pods (Type 1) from Northwood Narrows, New Hampshire were collected from Stop 2 of Eusden (1988). Both types are hosted by a foliated garnet (5%) – biotite (partly altered to chlorite) quartzite with decussate laths of muscovite. Tourmaline, biotite, and opaque phases are commonly concentrated along the contacts between the coticule and host quartzite, both of which have little, if any, other tourmaline. Only the laterally extensive coticules were thin-sectioned. They are characterized by anhedral grains of garnet (60–65%; to 0.5 mm) set in a matrix dominated by quartz with subordinate amounts of biotite, apatite

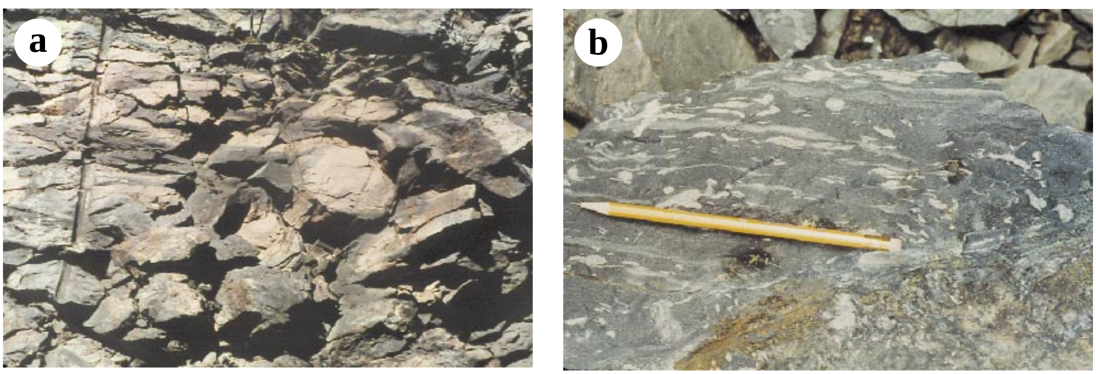

FIG. 3. Coticules observed within the Megunticook Formation, St. Croix belt. (a) Thinly laminated coticule (Camden). (b) Lenses or pods (Calais).

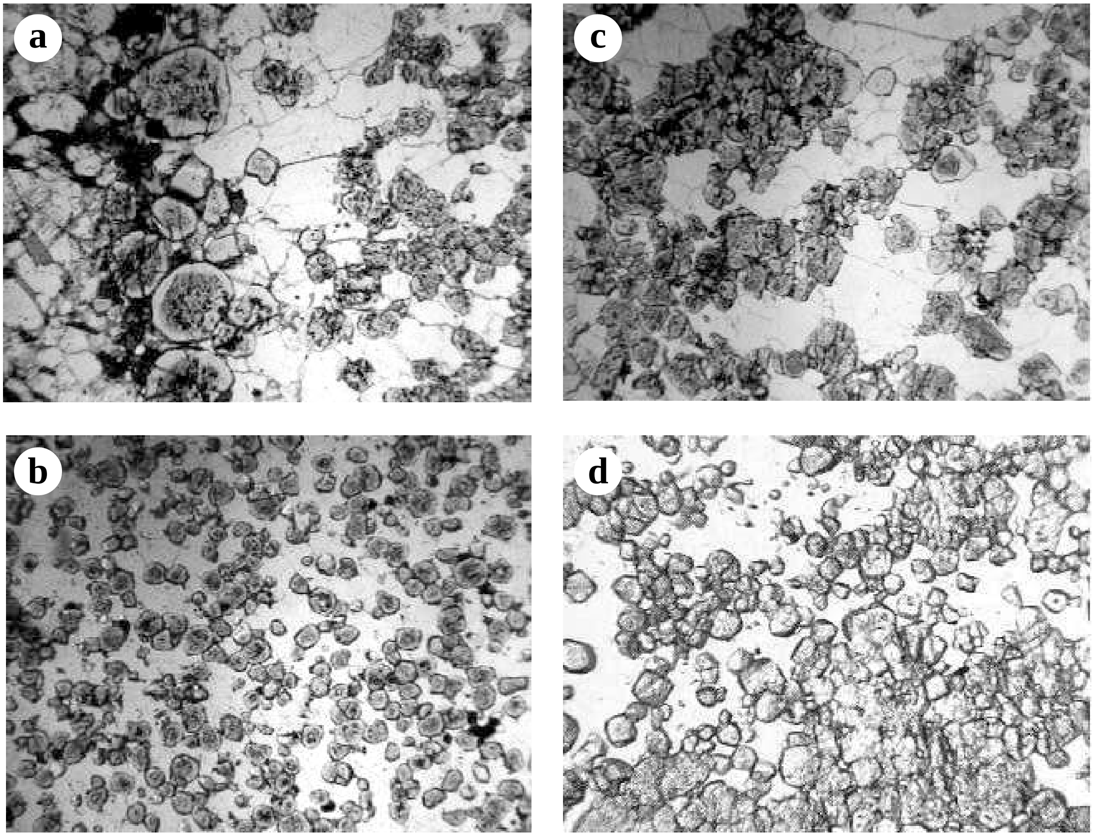

FIG. 4. Photomicrographs of Perry Mountain Formation samples (field of view for a–c: ~5 mm; d: ~1 mm). (a) Dark-colored coticule (NN–2/c). (b) Thick pink coticule. Garnet rims are inclusion-free (CS–7/c). (c) Intermediate-thickness pink coticule with finer-grained garnet (CS–6/c). (d) Pink coticule (CC–1/c). Symbols: NN Northwood Narrows, CS Center Strafford, CC Coos Canyon.

and ilmenite (Fig. 4a). The individual grains of garnet have many opaque inclusions throughout. Some coarser grains of garnet found near (to 1 mm) the borders of the coticule are characterized by an inclusion-rich core and an inclusion-free rim.

Center Strafford: The laterally extensive pink-colored coticules of Type 2 from near Center Strafford, New Hampshire (Stop 7A of Eusden *et al.* 1988) are characterized by anhedral grains of garnet (45–60%; to 0.5 mm) in a quartz-rich matrix with microscopically visible grains of tourmaline, apatite, rutile and opaque phases (ilmenite?). Most grains of garnet contain randomly oriented opaque inclusions throughout, although some have a distinct inclusion-free rim (Fig. 4b). The coticule adjacent to the host-rock contact is characterized by anhedral grains of garnet set in a matrix of quartz and subordinate amounts of altered plagioclase. Rutile is particularly abundant at the contact. The host rock is predominantly quartzite with little garnet, biotite, chlorite, muscovite, tourmaline, and rutile. Intermediate-thickness and thin pink (Type 2) coticules consist of finer-grained anhedral garnet (with few randomly oriented inclusions of opaque phases) set in a matrix of quartz, biotite, chlorite, muscovite, tourmaline, apatite, and opaque phases (Fig. 4c).

Coos Canyon: Thin-section analysis of a sample collected from a rather continuous type-2 coticule in a host of micaceous quartzite shows isolated anhedral grains of garnet (55–60%; to 0.5 mm) and numerous coalesced garnet grains in a quartz-rich matrix with apatite, rutile and opaque phases (Fig. 4d). The coarser isolated grains of garnet have an inclusion-free rim, whereas the coalesced aggregates of garnet grains are essentially inclusion-free. The host rock is a well-foliated biotite – garnet quartzite with plagioclase and opaque phases.

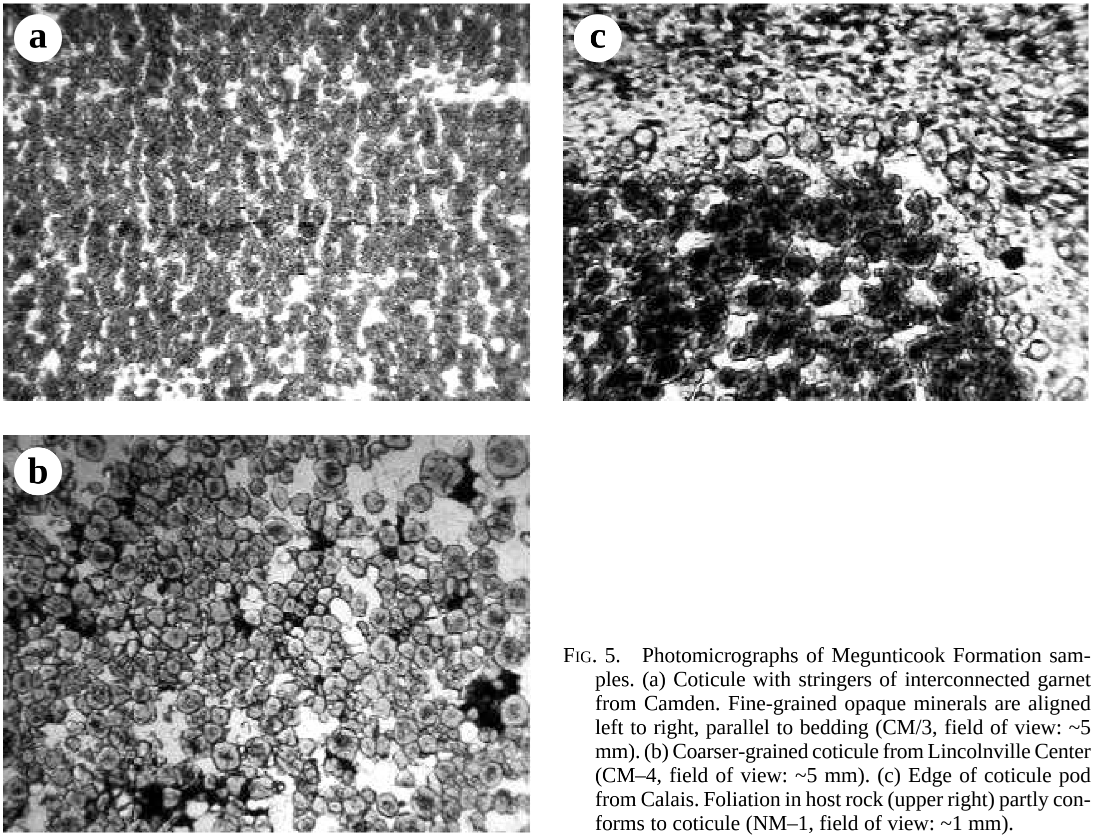

FIG. 5. Photomicrographs of Megunticook Formation samples. (a) Coticule with stringers of interconnected garnet from Camden. Fine-grained opaque minerals are aligned left to right, parallel to bedding (CM/3, field of view: ~5 mm). (b) Coarser-grained coticule from Lincolnville Center (CM–4, field of view: ~5 mm). (c) Edge of coticule pod from Calais. Foliation in host rock (upper right) partly conforms to coticule (NM–1, field of view: ~1 mm).

Megunticook Formation

Camden: Only very coticule-rich samples were collected from one locality near Camden, Maine; the host rock was not collected. Sample CM–3 is distinctly layered both in outcrop (Fig. 3a) and on a thin-section scale (Fig. 5a), with very garnet-rich layers alternating with fine-grained quartzite layers. The quartzite layers contain biotite and chlorite and extremely thin zones enriched in an opaque phase (opaque phase within garnet). The garnet-rich layers, which have interstitial quartz, apatite, and biotite (altered to a Fe-dominant chlorite), are characterized by "stringers" of interconnecting garnet grains (60–65% garnet) (Fig. 5a). These stringers are aligned perpendicular to the layering in the rock. Individual grains of garnet are cluttered with inclusions that are too fine to identify with a petrographic microscope, but their alignment parallels the layering.

Lincolnville Center: The sample from Lincolnville Center (CM–4) is characterized by more distorted layering and coarser grain-size than samples from Camden. Individual layers are difficult to trace in thin section (Fig. 5b). Grains of garnet (to 0.5 mm) are scattered throughout the rock, with none of the interconnectivity described above. These grains have an orange-colored, inclusion-rich core and an inclusion-free rim. The matrix consists of interstitial quartz, plagioclase, biotite, apatite, and opaque phases.

Calais: The dominant rock-type is a hard, dense, fine-grained black argillaceous quartzite comprised of biotite, garnet, and quartz ± cordierite. The red-brown biotite defines a foliation that is cross-cut by biotite-free poikilitic porphyroblasts (to 0.5 mm) of slightly altered cordierite (?) and isolated euhedral grains of garnet to 0.5 mm across. The foliation partially wraps around or is disrupted by scattered pods of coticule to 2 cm across (Fig. 5c). The coticule consists of fine-grained equigranular garnet (~80%), interstitial quartz, opaque phases, apatite, and traces of biotite and muscovite. The garnet grains have an isotropic core rich in opaque inclusions; their relatively inclusion-free rim is slightly anisotropic.

GARNET COMPOSITIONS

Garnet was analyzed with a Cameca MBX three-spectrometer electron microprobe housed at the GeoAnalytical Laboratory at Washington State University in Pullman, Washington. Operating conditions for analyses included an accelerating potential voltage of 15 keV, and a sample current of 15 nA. A beam diameter of 1 to 5 μm was used for garnet. Table 1 shows representative compositions of the garnet core from selected coticules and from some of their host rocks. Garnet grains from Coos Canyon and the Camden, Maine area were not analyzed.

Garnet in coticule from the Perry Mountain Formation (NN–2/c, CS–3/c, CS–6/c, and CS–7/c) is dominated by almandine (Alm) and spessartine (Sps) components. The spessartine content is highest (33.4%) in thick (8 cm), pink coticules (CS–7/c) and lowest (18.4%) in the thin (2 cm), pink coticules (CS–3/c). Pink coticules of intermediate thickness (3–4 cm) (CS–6/c) have intermediate Mn contents (26.1% Sps). Garnet in thin (2 cm), black coticule (NN–2/c) also have high Mn contents (27.9% Sps), but not as high as in the thick, pink coticules.

In each case, garnet in coticule from the Perry Mountain Formation is more manganiferous than that in the host rock. Figure 6 illustrates the variation in garnet compositions across individual layers of coticule. Each data point represents the core composition of an individual grain of garnet within the coticule layer. With the exception of NN–2/c (thin, black coticule), most individual grains of garnet have similar compositions across the width of the coticule. In all cases, the Sps content of host-rock garnet decreases away from the coticule–host contact. Individual grains of garnet are homogeneous at the scale of resolution of the electron microprobe.

TABLE 1. REPRESENTATIVE COMPOSITIONS OF GARNET CORE IN COTICULES (c) AND HOST ROCKS (h)

Sample No.	NN-2/h	NN-2/c	CS-3/h	CS-3/c	CS-6/c	CS-7/h	CS-7/c	NM-1/h*	NM-1/c*
Cot. Thickness	-	1.2 cm	-	1.5 cm	6.3 cm	-	8 cm	-	-
SiO_2 wt.%	36.89	36.92	36.60	36.71	36.53	37.27	37.06	36.54	36.48
Al_2O_3	20.92	20.49	20.39	20.61	20.59	20.47	20.46	20.41	20.31
FeO	28.61	27.16	36.25	31.70	27.54	27.00	23.73	22.72	23.02
MnO	11.23	12.46	3.56	8.18	11.63	11.95	14.94	16.56	15.76
MgO	2.34	2.52	2.24	2.33	2.66	2.62	2.64	1.92	1.61
CaO	0.82	0.78	0.81	0.60	0.81	0.91	1.39	1.26	2.15
Total	100.81	100.33	99.85	100.13	99.76	100.22	100.22	99.41	99.33
Si *apfu*	2.916	2.918	2.983	2.901	2.887	2.945	2.928	2.888	2.883
Al	1.949	1.909	1.898	1.920	1.917	1.906	1.906	1.901	1.891
Fe	1.890	1.795	2.395	2.095	1.820	1.784	1.568	1.501	1.521
Mn	0.752	0.834	0.238	0.547	0.778	0.800	1.000	1.108	1.055
Mg	0.276	0.297	0.264	0.274	0.313	0.309	0.311	0.226	0.190
Ca	0.069	0.066	0.068	0.051	0.068	0.077	0.118	0.107	0.182
X_{Fe}	0.873	0.858	0.901	0.884	0.853	0.852	0.834	0.869	0.889
Alm	0.633	0.600	0.808	0.706	0.611	0.601	0.523	0.510	0.516
Prp	0.092	0.099	0.089	0.092	0.105	0.104	0.104	0.077	0.064
Sps	0.252	0.279	0.080	0.184	0.261	0.269	0.334	0.377	0.358
Grs	0.023	0.022	0.023	0.017	0.023	0.026	0.039	0.036	0.062

Formulae based on 12 atoms of oxygen. * Average of 4 analyses. Symbols: NN Northwood Narrows, New Hampshire; CS Center Strafford, New Hampshire; NM northern Maine (Calais). Analyzed at The GeoAnalytical Laboratory at Washington State University, Pullman, Washington.

The compositional variations across individual layers of coticule are approximately symmetrical with respect to bedding and may very well represent a compositional gradient imposed on the layers by early diagenesis and sedimentation. Diffusion during metamorphism may have modified the compositional gradient, but the extent of modification is unclear. Even coticules of significant thickness metamorphosed under granulite-facies conditions may retain compositional gradients of sedimentary origin (Thomson, 1992).

Of those grains analyzed, the most manganiferous is found in the samples of host rock and coticule from the Megunticook Formation in Calais, Maine (NM–1). Here, garnet in the host rock has a higher Mn content (37.7% Sps) than in the coticule (35.7%), in contrast to the observations made in the Perry Mountain Formation. Garnet in coticule from Calais, Maine also has a higher Grs content than does garnet of the Perry Mountain Formation (Table 1).

Whole-Rock Geochemistry of Coticules and Their Hosts

Major-, trace-, and rare-earth element analyses were done on selected samples of coticule and their host rocks. Samples of coticule were carefully separated from their adjacent host-rocks by trim saw. Small chips were collected from each coticule and the host-rock immediately adjacent to it. Chips were ground on silicon carbide 240 grit to remove saw marks, and prepared for XRF and ICP–MS analysis following the specific procedures outlined by each laboratory responsible for the analyses. Concentrations of major elements (Si, Ti, Al, Fe, Mn, Mg, Ca, Na, K, P) and additional trace elements (Zr, Ga, Zn, Ni, Cr) were established at the Ronald B. Gilmore X-Ray Fluorescence Laboratory at the University of Massachusetts, Amherst, by X-ray fluorescence (XRF) spectrometry. Samples were analyzed for trace elements including the *REE* at the GeoAnalytical Laboratory facilities at Washington State University, Pullman, by inductively coupled plasma – mass spectrometry (ICP–MS). Representative results of the analyses of coticules and their host rocks from this study are given in Tables 2–5.

Figure 7 illustrates the ratio of the concentration of each oxide in coticules from the Perry Mountain Formation to the concentration in schistose host-rocks. Sample CS–1 was excluded from the comparison because it is quartzite. Relative to the interbedded schists, the Perry Mountain coticules have comparable SiO_2 and Al_2O_3. MgO and CaO are either slightly enriched or slighly depleted relative to schists. As expected, the Perry Mountain Formation coticules are depleted in TiO_2, Na_2O, and K_2O, and enriched in Fe_2O_3, MnO, and P_2O_5, relative to the schistose host-rocks. Coticules in

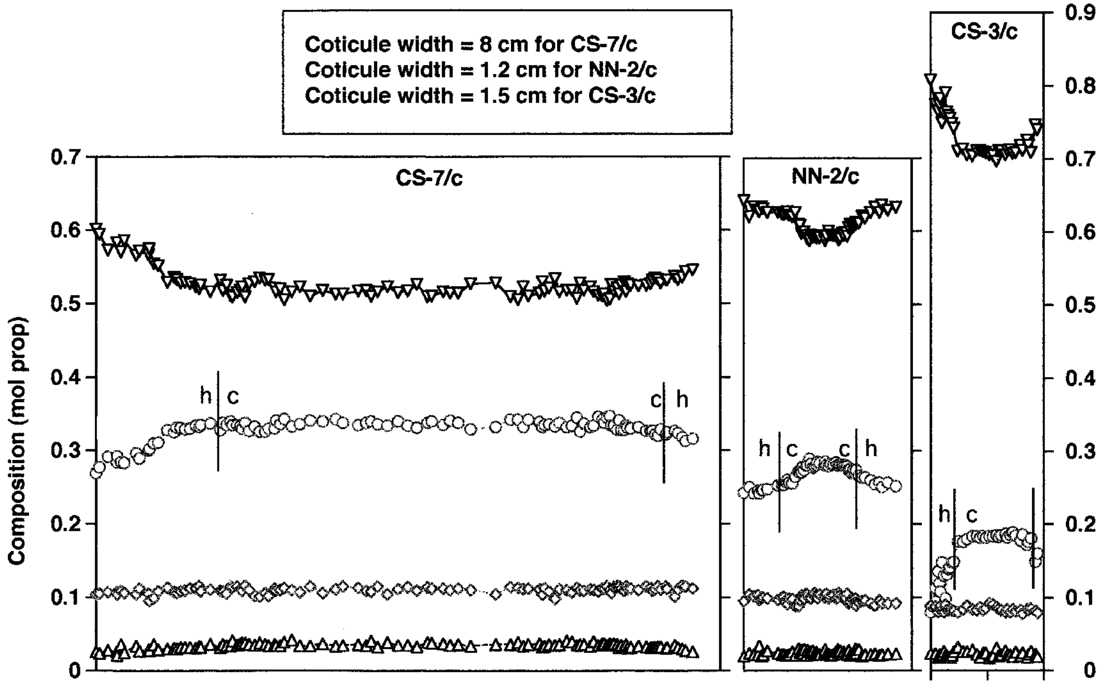

FIG. 6. Compositions of individual garnet cores across three coticules from the Perry Mountain Formation, ranging in width from 1.2 to 8 cm. From top to bottom, the profiles pertain to the almandine, spessartine, pyrope and grossular components. Symbols: c: coticule, h: host.

the Megunticook Formation are more difficult to deal with owing to the difficulty in separating coticule from host rock in samples collected from the Camden area. These samples are characterized by thinly laminated and intercalated coticule–host horizons. However, coticules from Calais, Maine (NM–1/c) are enriched in Fe_2O_3, MnO, and CaO relative to the host rock (NM–1/h).

The differences in major-element composition are reflected by differences in modal mineralogy of the samples. Coticules are enriched in almandine–spessartine garnet and quartz and depleted in micaceous minerals, which would account for high iron and manganese and low alkalis. High phosphorus concentrations are a reflection of significant amounts of apatite in coticules.

The *REE* profiles of the Perry Mountain (Fig. 8a) and Megunticook Formation (Fig. 8b) coticules relative to chondrites closely resemble those of coticules investigated by Spry (1990). In general, they are light-*REE* enriched, with concentrations between 10 and 100 times those of chondrites, and lack a Ce anomaly. Most have a negative Eu anomaly. But a closer examination of Figure 8a reveals that the coticules from the Perry Mountain Formation can be subdivided into five groups depending on their *REE* profiles. Furthermore, the groups correspond with the coticule types recognized in the field and outlined above. Type-2 coticules (pink, laterally extensive coticules of variable thickness) can be subdivided into thin (to 2 cm; CS–1/c, CS–3/c, CS–4/c and CC–1/c, CC–2b/c), intermediate (4–6 cm; CS–2/c, CS–6/c), and thick (to 8 cm; CS–7/c) coticule. Note that the *REE* patterns for thin pink coticules from Coos Canyon, Maine differ from those from Center Strafford, New Hampshire. Rare-earth-element patterns of the intermediate-thickness pink coticules from Center Strafford have higher light *REE* abundances than thin pink coticules from the same location. Both are characterized by a negative Eu anomaly. The thickest pink coticule (CS–7/c) from this locality has even lower light *REE* abundances than the thin pink coticules in this region. Thin pink samples from Coos Canyon are characterized by light *REE* abundances between those of the thickest pink coticule and the intermediate-thickness coticules in New Hampshire and a less-pronounced negative Eu anomaly. Finally, type-3 coticules (dark-colored, laterally extensive coticules to 2 cm; NN–2/c and NN–3/c) are light-*REE*-enriched but lack any Eu anomaly.

Coticules from the Megunticook Formation are light-*REE*-enriched and have a negative Eu anomaly. They differ from the *REE* patterns of coticules from the Perry Mountain Formation in that they are characterized by a very slight positive Ce anomaly.

TABLE 2. COMPOSITION OF COTICULES (c) AND HOST ROCKS (h), NEW ENGLAND APPALACHIANS

Sample No.	NN-2/c	NN-3/c	CS-1/c	CS-2/c	CS-3/c	CS-4/c	CS-6/c	CS-7/c	CC-1/c	CC-2b/c	CM-3a/c	CM-4b/c	NM-1/c
SiO_2 wt.%	60.85	59.23	61.75	61.12	69.14	70.45	60.41	57.48	57.95	62.23	57.02	59.22	51.01
TiO_2	0.37	0.22	0.09	0.59	0.14	0.19	0.55	0.09	0.48	0.67	0.42	0.60	0.69
Al_2O_3	13.63	13.75	12.34	11.89	10.36	10.27	12.24	13.72	13.43	12.18	13.97	13.78	16.99
Fe_2O_3	15.89	16.71	18.02	17.13	15.68	15.20	17.10	16.64	19.79	17.52	9.89	11.74	15.57
MnO	5.88	6.89	5.44	5.89	3.10	2.24	6.27	9.00	3.09	2.73	14.68	10.40	9.37
MgO	1.62	1.74	1.41	1.50	1.04	1.06	1.61	1.82	2.46	2.09	2.58	2.47	2.18
CaO	1.08	1.26	0.87	1.43	0.66	0.53	1.36	1.23	1.16	1.11	1.33	0.79	1.98
Na_2O	0.99	0.63	0.33	0.14	0.23	0.21	0.22	0.33	0.27	0.75	0.19	0.59	0.29
K_2O	0.30	0.17	0.11	0.14	0.28	0.57	0.14	0.05	1.18	0.77	0.26	0.48	1.16
P_2O_5	0.37	0.52	0.30	0.75	0.28	0.20	0.68	0.35	0.24	0.25	0.20	0.27	0.30
Total	100.98	101.12	100.66	100.58	100.91	100.92	100.58	100.71	100.05	100.30	100.54	100.34	99.54

Sample No.	NN-2/h	NN-3/h	CS-1/h	CS-2/h	CS-3/h	CS-4/h	CS-6/h	CS-7/h	CC-1/h	CC-2b/h	CM-3b/m	CM-4a/m	NM-1/h
SiO_2 wt.%	59.00	64.38	89.91	n.a.	n.a.	63.13	n.a.	76.36	68.95	59.07	51.78	58.20	56.64
TiO_2	1.03	1.04	0.56	n.a.	n.a.	0.91	n.a.	1.02	0.96	1.04	0.44	0.56	0.96
Al_2O_3	20.34	17.50	3.83	n.a.	n.a.	17.34	n.a.	11.32	14.65	21.20	14.29	13.84	19.52
Fe_2O_3	7.67	6.07	2.94	n.a.	n.a.	11.03	n.a.	4.37	6.70	9.04	11.99	13.08	12.34
MnO	1.03	0.66	0.34	n.a.	n.a.	0.75	n.a.	1.19	0.19	0.23	13.28	10.03	1.33
MgO	1.97	1.85	0.64	n.a.	n.a.	2.14	n.a.	1.08	2.18	2.55	3.01	2.23	2.09
CaO	1.43	1.42	0.18	n.a.	n.a.	0.25	n.a.	0.79	0.95	0.88	3.00	0.72	1.03
Na_2O	4.56	4.36	0.04	n.a.	n.a.	0.44	n.a.	1.96	2.98	3.21	0.03	0.63	0.80
K_2O	2.67	2.12	0.79	n.a.	n.a.	3.88	n.a.	1.36	2.30	2.70	0.53	0.76	4.43
P_2O_5	0.14	0.16	0.09	n.a.	n.a.	0.13	n.a.	0.05	0.04	0.06	1.25	0.14	0.34
Total	99.84	99.56	99.32			100.00		99.50	99.90	99.98	99.60	100.19	99.48

Total iron is reported as Fe_2O_3. m: mixed host and coticule (inseparable). The samples were analyzed at The Ronald B. Gilmore X-ray Laboratory at the University of Massachusetts in Amherst. Symbols: NN: Northwood Narrows, New Hampshire; CS: Center Strafford, New Hampshire; CC: Coos Canyon, Maine; CM: central Maine (Camden); NM: northern Maine (Calais); n.a.: not analyzed.

Discussion

The major-element data may indicate the possible protolith of these coticules. Bonatti (1975) delineated three fields on a plot of weight percent SiO_2 *versus* weight percent Al_2O_3 to indicate whether the source environment of coticules is hydrothermal, hydrogenous, or pelagic. Spry (1990) noted that the data from within a small geological region do not fall within just one of the three fields delineated on Bonatti's plot. Spry concluded that the data from a specific region might represent a mixing line between hydrothermal and pelagic sediment. Coticule data from this study as well as others in the New England Appalachians (Docka 1985) and southern Appalachians (Wonder *et al.* 1988) are plotted in Figure 9. The data of Wonder *et al.* (1988) may well represent a mixing line between hydrothermal and pelagic sediment. However, these data illustrate that coticules associated with iron formation and sulfide deposits are silica-poor compared to New England coticules which are not associated with these kinds of deposits. Most of Docka's (1985) data and the Perry Mountain and Megunticook data (with the exception of the Calais, Maine coticule NM–1) from this study plot primarily within the hydrogenous field. In fact, the "mixing-lines" drawn for the data established in this study (Fig. 9) may not be indicative of mixing of different sediment types at all. The SiO_2 *versus* Al_2O_3 ratio for an individual coticule could just as easily represent the modal proportion of garnet to quartz. For example, the sample from Calais, Maine (NM–1/c) consists of nearly 80% garnet and 20% quartz. The relationship between SiO_2 and Al_2O_3 alone is not conclusive concerning the protolith of these garnet-rich rocks. Furthermore, coticules in the Perry Mountain Formation cannot be distinguished from those from the Megunticook Formation on a SiO_2 *versus* Al_2O_3 diagram. Perhaps the *REE* data can tell us more.

Marchig *et al.* (1982) noted that chondrite-normalized *REE* patterns of diagenetic metalliferous sediments, deep-sea sediments and hydrothermal metalliferous sediments exhibit distinct trends; they proposed that

TABLE 3. TRACE-ELEMENT CONCENTRATIONS IN COTICULES (c), NEW ENGLAND APPALACHIANS

Sample No.	NN-2/c	NN-3/c	CS-1/c	CS-2/c	CS-3/c	CS-4/c	CS-6/c	CS-7/c	CC-1/c	CC-2b/c	CM-3a/c	CM-4b/c	NM-1/c
La ppm	15.61	11.59	8.16	34.93	11.81	14.45	27.02	4.85	12.90	22.98	31.93	31.85	44.46
Ce	30.80	24.09	17.37	75.42	24.80	29.50	59.83	11.24	26.30	45.34	82.38	85.82	91.93
Pr	3.36	2.71	2.07	7.80	2.68	3.17	6.27	1.39	2.90	4.90	6.96	6.83	9.01
Nd	13.18	10.58	9.05	32.73	11.00	12.51	26.76	6.64	12.02	19.18	28.51	27.20	35.98
Sm	2.90	2.52	2.97	8.97	3.03	3.16	7.25	2.74	4.31	5.02	7.24	6.74	8.66
Eu	0.94	0.90	0.60	1.59	0.73	0.74	1.36	0.53	1.26	1.43	1.40	1.45	1.79
Gd	3.59	3.00	4.27	10.59	4.65	4.55	9.08	4.65	10.16	7.27	8.06	7.16	8.06
Tb	0.81	0.69	1.00	1.92	1.14	1.04	1.67	1.11	2.16	1.54	1.47	1.37	1.36
Dy	6.05	5.46	7.12	11.77	8.90	7.74	10.70	6.94	12.98	9.97	9.02	8.65	7.89
Ho	1.39	1.28	1.51	2.27	2.05	1.78	2.17	1.17	2.36	1.97	1.79	1.72	1.62
Er	4.23	4.03	4.21	5.84	6.20	5.59	5.72	2.60	6.12	5.13	4.77	4.56	4.17
Tm	0.71	0.67	0.62	0.83	0.97	0.94	0.80	0.34	0.91	0.73	0.68	0.64	0.61
Yb	4.96	4.63	3.89	4.85	6.18	6.47	4.84	1.91	5.95	4.58	4.22	3.88	3.93
Lu	0.80	0.73	0.58	0.69	0.91	1.02	0.71	0.25	0.88	0.67	0.63	0.60	0.63
Ba	35	16	14	16	46	84	14	2	80	65	78	189	557
Th	3.16	1.77	1.39	5.52	2.17	3.18	4.14	0.13	4.35	8.19	5.85	7.24	9.81
Nb	7.17	3.99	2.80	26.30	2.80	3.60	26.71	2.87	7.56	8.25	10.19	17.57	14.36
Y	43.11	39.60	45.04	68.36	62.55	50.90	63.27	38.47	73.00	54.73	44.03	44.06	45.19
Hf	1.70	1.12	1.05	2.31	1.01	1.15	1.98	0.73	3.04	5.01	2.54	3.02	3.10
Ta	2.13	1.18	1.70	3.85	1.88	1.86	3.48	1.15	0.59	0.78	2.26	3.32	2.80
U	1.17	1.05	0.64	2.38	0.94	1.00	1.93	0.68	1.44	1.80	1.92	1.69	1.46
Pb	14.70	11.04	2.70	1.53	3.31	4.45	1.65	6.41	3.13	7.19	1.41	7.34	7.44
Rb	20.8	12.7	5.9	6.1	8.0	29.9	9.4	7.5	75.2	62.1	14.4	15.2	61.5
Cs	2.34	1.45	0.45	0.42	0.27	1.91	1.06	0.67	9.18	7.08	1.89	0.44	4.21
Sr	81	52	18	14	17	17	12	10	11	46	9	21	46
Sc	23.4	19.9	23.4	29.5	28.0	34.8	30.2	17.6	28.4	24.6	20.9	22.0	23.9

Symbols: NN: Northwood Narrows, New Hampshire; CS: Center Strafford, New Hampshire; CC: Coos Canyon, Maine; CM: central Maine (Camden); NM: northern Maine (Calais). The samples were analyzed at The GeoAnalytical Laboratory at Washington State University, Pullman, Washington.

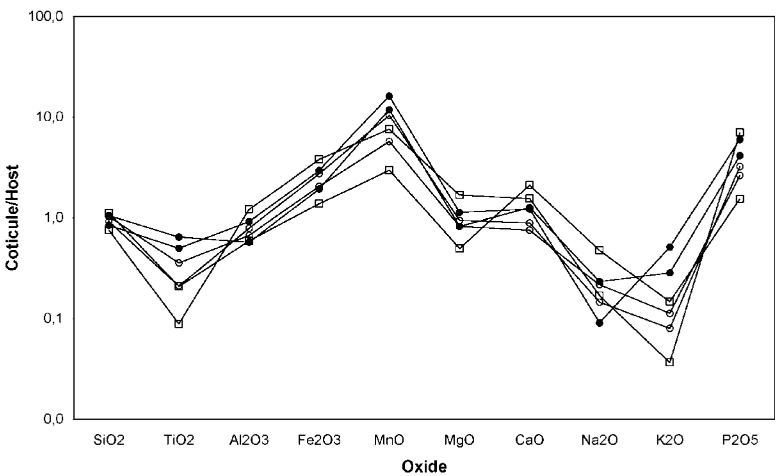

FIG. 7. Host-normalized major-element composition of coticules from the Perry Mountain Formation. Open circles: Northwood Narrows, open squares: Center Strafford, closed circles: Coos Canyon.

TABLE 4. TRACE-ELEMENT CONCENTRATIONS IN HOST ROCKS (h) NEW ENGLAND APPALACHIANS

Sample No.	NN-2/h	NN-3/h	CS-1/h	CS-2/h	CS-3/h	CS-4/h	CS-6/h	CS-7/h	CC-1/h	CC-2b/h	CM-3b/m	CM-4a/m	NM-1/h
La ppm	50.04	56.72	29.62	n.a.	n.a.	46.34	n.a.	48.24	44.57	44.85	41.32	40.33	59.58
Ce	93.40	107.82	58.87	n.a.	n.a.	93.05	n.a.	96.51	86.85	87.19	119.56	105.17	123.45
Pr	9.82	11.33	6.32	n.a.	n.a.	9.64	n.a.	10.26	9.65	9.56	9.05	8.58	12.26
Nd	36.62	42.29	24.77	n.a.	n.a.	37.71	n.a.	39.96	37.62	37.68	39.17	33.39	48.33
Sm	7.07	8.27	5.36	n.a.	n.a.	8.38	n.a.	8.23	8.20	8.29	10.43	7.75	10.58
Eu	2.49	2.56	0.74	n.a.	n.a.	1.60	n.a.	1.53	1.92	2.08	2.38	1.35	2.32
Gd	6.05	6.75	4.20	n.a.	n.a.	7.26	n.a.	6.89	6.79	6.86	11.88	8.01	9.92
Tb	1.15	1.13	0.59	n.a.	n.a.	1.40	n.a.	1.16	1.10	1.13	2.04	1.76	1.59
Dy	7.54	7.13	2.95	n.a.	n.a.	9.47	n.a.	7.52	6.43	6.52	11.68	12.78	9.39
Ho	1.61	1.46	0.56	n.a.	n.a.	2.09	n.a.	1.58	1.29	1.32	2.23	2.59	1.90
Er	4.50	4.02	1.50	n.a.	n.a.	5.83	n.a.	4.42	3.64	3.58	5.37	6.50	4.84
Tm	0.74	0.60	0.23	n.a.	n.a.	0.85	n.a.	0.64	0.54	0.53	0.73	0.88	0.71
Yb	5.04	3.82	1.62	n.a.	n.a.	5.09	n.a.	3.90	3.40	3.36	4.17	5.13	4.27
Lu	0.86	0.62	0.28	n.a.	n.a.	0.79	n.a.	0.61	0.54	0.52	0.61	0.75	0.67
Ba	354	267	99	n.a.	n.a.	498	n.a.	167	267	314	153	137	2278
Th	13.43	13.77	8.10	n.a.	n.a.	11.98	n.a.	10.44	13.37	14.73	5.26	8.51	13.13
Nb	15.71	13.64	8.48	n.a.	n.a.	18.42	n.a.	14.64	18.41	19.15	9.24	15.53	19.28
Y	43.43	36.93	13.66	n.a.	n.a.	52.72	n.a.	44.03	33.14	33.74	55.76	68.48	52.82
Hf	4.23	4.10	5.52	n.a.	n.a.	3.77	n.a.	8.00	8.25	6.25	2.57	3.52	4.22
Ta	2.30	6.43	6.09	n.a.	n.a.	2.87	n.a.	2.44	1.36	1.40	5.25	2.81	2.40
U	2.80	2.98	1.50	n.a.	n.a.	2.58	n.a.	2.49	3.32	3.14	1.94	1.82	1.82
Pb	68.74	68.62	5.18	n.a.	n.a.	20.36	n.a.	26.30	33.57	35.48	3.51	11.52	36.78
Rb	148.6	137.5	46.6	n.a.	n.a.	187.9	n.a.	66.2	137.7	159.8	41.3	41.4	166.6
Cs	12.43	11.55	2.82	n.a.	n.a.	11.18	n.a.	3.23	14.65	13.40	7.77	1.69	10.19
Sr	414	398	26	n.a.	n.a.	70	n.a.	188	296	269	12	15	138
Sc	20.3	17.0	6.9	n.a.	n.a.	23.1	n.a.	17.6	16.6	24.4	17.6	32.3	20.6

m: mixed host and coticule (inseparable). Symbols: NN: Northwood Narrows, New Hampshire; CS: Center Strafford, New Hampshire; CC: Coos Canyon, Maine; CM: central Maine (Camden); NM: northern Maine (Calais). The samples were analyzed at The GeoAnalytical Laboratory at Washington State University, Pullman, Washington.

these differences can serve as geochemical disciminants between hydrogenous and hydrothermal sediments. Figure 10 illustrates *REE* trends of typical seawater (Elderfield & Greaves 1983), the North American Shale Composite (NASC) (Gromet *et al.* 1984, McLennan 1989), oceanic pelagic clays (Rankin & Glasby 1979), manganese nodules (hydrogenous) (Haynes *et al.* 1986), and hydrothermal sediments both near and far from active vents (Ruhlin & Owen 1986). Hydrothermal metalliferous sediments are characterized by a large negative Ce anomaly that is greater in hydrothermal sediments in the vicinity of the vent system. On the other hand, diagenetic metalliferous sediments (manganese nodules) show a positive Ce anomaly and overall *REE* abundances greater than those of hydrothermal metalliferous sediments. In fact, it has been suggested that a positive Ce anomaly is indicative of a hydrogenous origin, whereas pure hydrothermal sediments are characterized by a negative Ce anomaly. The only very slight positive Ce anomaly in the Megunticook Formation samples is inconclusive. The general lack of a Ce anomaly in the Perry Mountain Formation suggests that these are neither hydrothermal nor hydrogenous. Fur-

TABLE 5. ADDITIONAL INFORMATION ON TRACE-ELEMENT CONCENTRATIONS IN COTICULES (c) AND HOST ROCKS (h) NEW ENGLAND APPALACHIANS

Sample No.	NN-2/c	NN-3/c	CS-1/c	CS-2/c	CS-3/c	CS-4/c	CS-6/c	CS-7/c	CC-1/c	CC-2b/c	CM-4b/c	NM-1/c
Zr ppm	46	n.a.	46	84	34	39	71	25	117	187	110	116
Ga	6	n.a.	4	4	4	6	4	8	9	6	11	20
Zn	47	n.a.	28	30	12	17	33	31	60	33	68	129
Ni	7	n.a.	0	2	0	2	3	3	55	49	46	125
Cr	42	n.a.	17	60	26	34	61	28	51	72	91	90
V	85	n.a.	69	84	40	45	82	96	87	50	116	158
Sample No.	**NN-2/h**	**NN-3/h**	**CS-1/h**	**CS-2/h**	**CS-3/h**	**CS-4/h**	**CS-6/h**	**CS-7/h**	**CC-1/h**	**CC-2b/h**	**CM-4a/m**	**NM-1/h**
Zr ppm	158	151	243	n.a.	n.a.	139	n.a.	340	313	232	123	161
Ga	21	17	5	n.a.	n.a.	26	n.a.	14	16	26	13	25
Zn	123	116	38	n.a.	n.a.	96	n.a.	55	76	227	55	157
Ni	39	37	13	n.a.	n.a.	22	n.a.	31	46	52	17	118
Cr	104	91	34	n.a.	n.a.	98	n.a.	77	97	120	106	94
V	119	96	39	n.a.	n.a.	165	n.a.	114	109	146	120	136

m: mixed host and coticule (inseparable); n.a.: analyzed. Symbols: NN: Northwood Narrows, New Hampshire; CS: Center Strafford, New Hampshire; CM: central Maine (Camden); NM: northern Maine (Calais). The samples were analyzed at The GeoAnalytical Laboratory at Washington State University, Pullman, Washington.

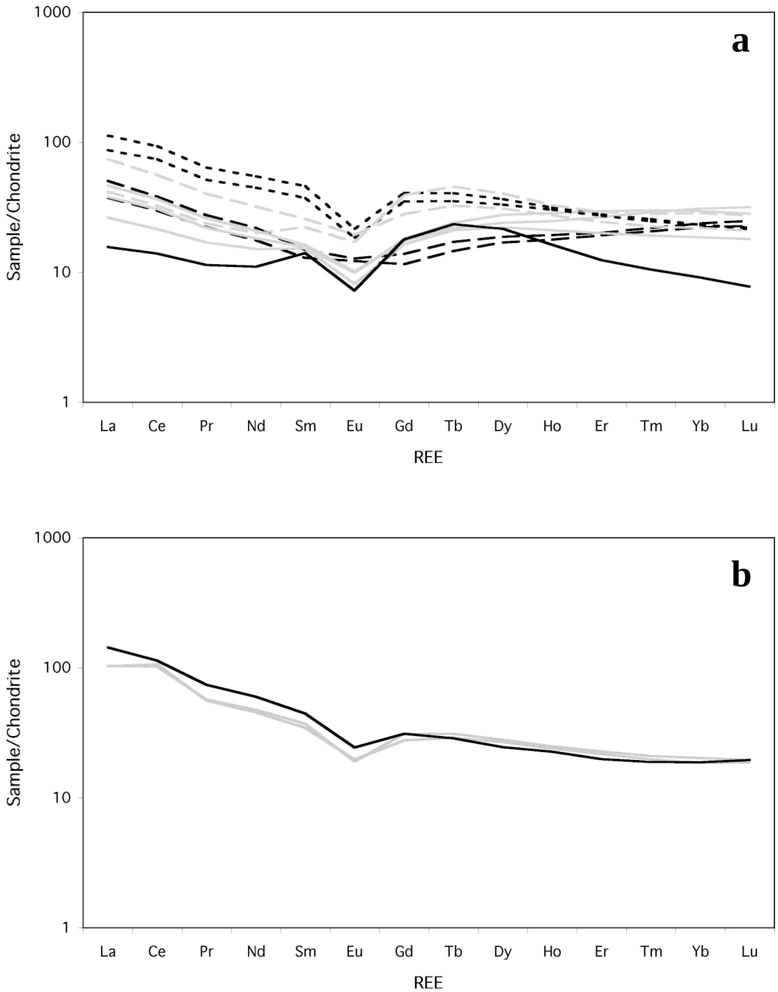

FIG. 8. Chondrite-normalized *REE* data for coticules and average host-rock. (a) Perry Mountain Formation. Short-dashed black lines are intermediate thickness coticules (CS–6/c, CS–2/c). Long-dashed gray lines are thin pink coticules (CC–1/c, CC–2b/c). Long-dashed black lines are thin black coticules (NN–2/c, NN–3/c). Solid gray lines are thin pink coticules (CS–1/c, CS–3/c, CS–4/c). Solid black line is a thick pink coticule (CS–7/c). b) Megunticook Formation. Black line: NM–1/c. Gray lines: CM–3a/c, CM–4b/c. The chondrite data are taken from Boynton (1984).

thermore, hydrogenous sediments typically have *REE* concentrations between 100 and 1000 times those of chondrites, whereas the *REE* concentrations in hydrothermal sediments are between 20 and 100 times those of chondrites. The data in Figure 10 suggest that the coticules investigated here more closely resemble oceanic pelagic clays and the North America Shale Composite than the range of concentrations illustrated for hydrothermal or hydrogenous sediments on Figure 10. In fact, the *REE* patterns of the coticules plot between those of oceanic pelagic clays and typical seawater. However, the lack of a clay component in the coticules is rather curious and "perplexing". Furthermore, coticules of the Perry Mountain Formation cannot be distinguished from coticules of the Megunticook Formation on a *REE* diagram.

Trace-element data may also be used to evaluate the nature of the protolith of coticules of the Perry Mountain and Megunticook formations. Marchig *et al*. (1982) discussed various geochemical indicators as discriminants between hydrogenous (diagenetic) and hydrothermal sediments. Both deep-sea sediments and diagenetic

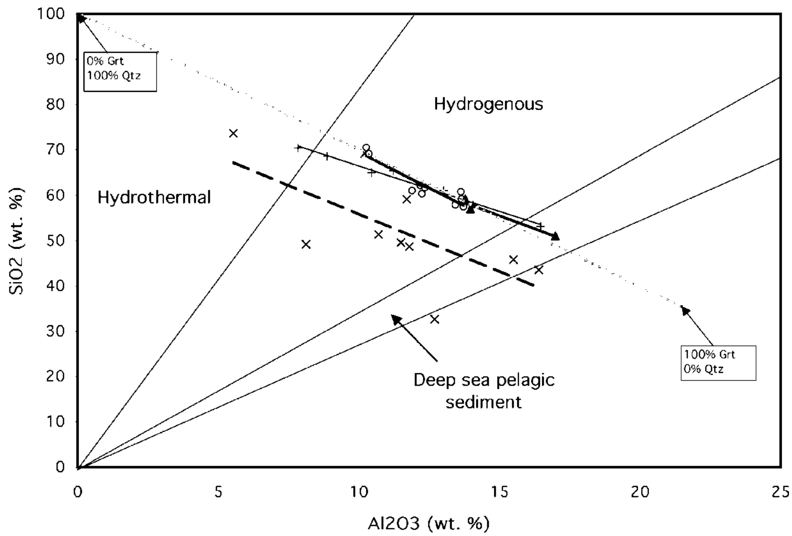

FIG. 9. SiO_2 *versus* Al_2O_3 (wt.%) for coticules from this study (open circles: Perry Mountain, filled triangles: Megunticook), the northern Appalachians (+ Docka 1985), and the southern Appalachians (Wonder *et al.* 1988). Compositional fields from Bonatti (1975). Heavy long-dashed line: trend line from Wonder *et al.* (1988). Thin solid line: trend line from Docka (1985). Short dashed lines: trend lines for data from this study. Dotted line: mixing line between quartz and garnet.

sediments may contain chromium derived from continental detritus. If the chromium is derived from continental detritus, then it should show a positive correlation with other elements presumed to be derived from continental detrital material, such as Ti, Mg, K, Rb, and Zr. On the other hand, Cr is more mobile in hydrothermal metalliferous sediments and may not be correlated with these terrigenous components.

The Cr data acquired in this study show a statistically significant correlation with the terrigenous component Zr (Fig. 11), at least in the Perry Mountain samples. Similar trends are obtained for plots of Cr *versus* Mg, K, Rb, and Ti. However, the trend of the Zr data, compared to that of Marchig *et al.* (1982), suggests that the coticules studied here are neither purely hydrogenous nor purely hydrothermal, and perhaps have another origin. Other geochemical indicators presented by Marchig *et al.* (1982), including weight % P_2O_5 *versus* Sc, Y, and La, are inconclusive owing to the small number of samples. One interesting conclusion from the data in Figure 11 is that the Coos Canyon coticules seem to have a greater continental component than Perry Mountain coticules in the Rochester area. This could very well be a reflection of the closeness of the Coos Canyon area to the basin edge (proximal), compared to the Rochester area (distal).

In summary, the major-element data acquired in this study hint at a hydrogenous origin, though the conclusion is not definitive. The *REE* data suggest that the coticules are more similar to oceanic pelagic sediment, North American Shale Composite, and typical seawater, but to suggest a possible protolith is difficult at best. Perhaps more importantly, the data demonstrate that none of the coticules of this study formed by hydrothermal processes.

Coticules from both the Perry Mountain and Megunticook formations were collected from sites within each formation that are considered to be stratigraphic equivalents. The intention was to see if coticule geochemistry within a specific formation could be used to confirm the stratigraphic correlation. It is clear that coticule geochemistry does not appear to be unique to a particular formation and may, in fact, be variable within a formation and perhaps within an individual coticule horizon. Clearly, the geochemistry of coticules is useful in evaluating sedimentary protoliths. However, a similar composition does not necessarily indicate stratigraphic correlation, and a different composition does not necessarily preclude stratigraphic correlation. Used by itself, the composition of coticules does not seem to be an effective tool for short- or long-range stratigraphic correlation.

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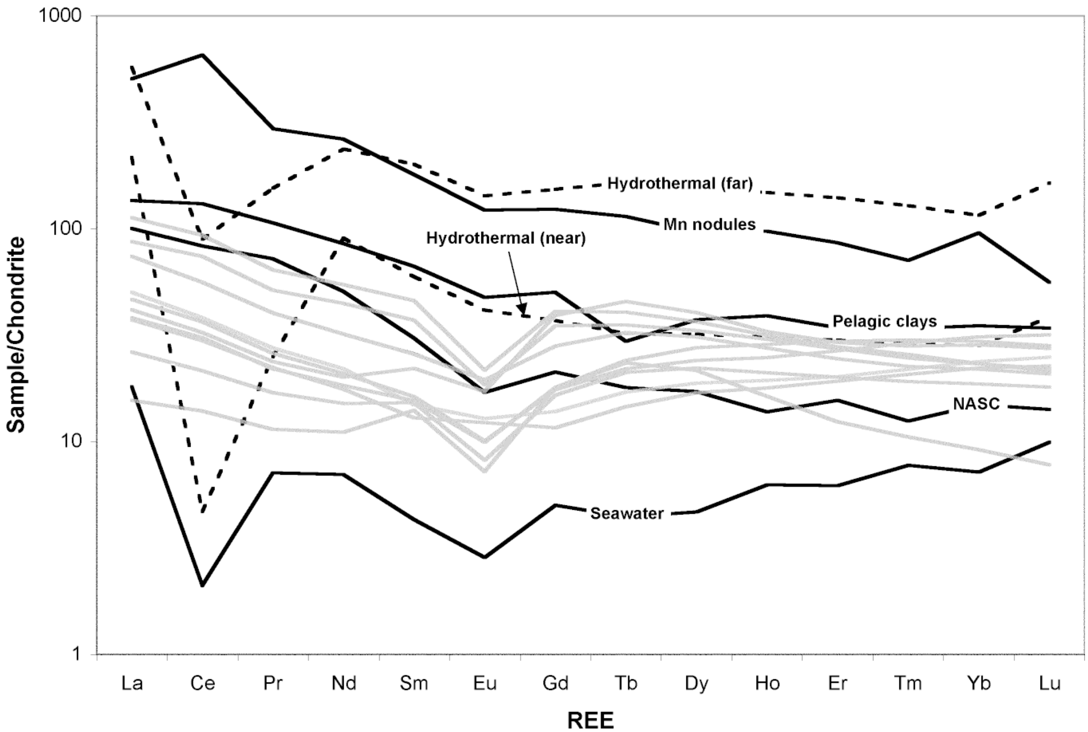

FIG. 10. Chondrite-normalized *REE* data for coticules of this study (gray). Also shown: North American Shale Composite (NASC) (Gromet *et al.* 1984, McLennan 1989), average Mn-nodule (Haynes *et al.* 1986), oceanic pelagic clay (Rankin & Glasby 1979), hydrothermal sediment (Ruhlin & Owen 1986), and typical seawater (Elderfield & Greaves 1983).

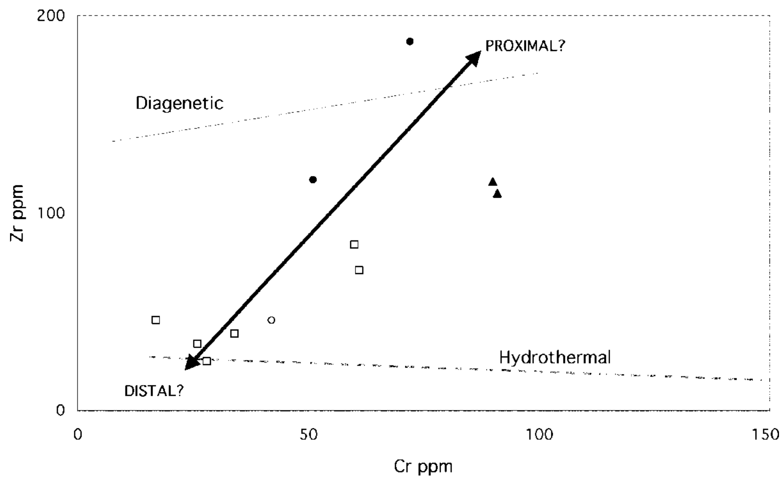

FIG. 11. Cr (ppm) plotted against Zr (ppm) for all coticules analyzed. Open circle: NN, open squares: CS, closed circles: CC, triangles: Megunticook Formation. Approximate trends for hydrothermal metalliferous sediments and diagenetic metalliferous sediments are from Marchig *et al.* (1982). Regression line is calculated for the coticules of the Perry Mountain Formation.

tion of thin sections. Whole-rock, major, and trace-element analyses were conducted at the University of Massachusetts with the assistance of Peter Dawson. Analyses by inductively coupled plasma – mass spectrometry for trace- and rare-earth elements and electron-microprobe analyses were conducted at the WSU GeoAnalytical Laboratories with the assistance of Diane Johnson, Charles Knaack and Scott Cornelius. Samples from Coos Canyon, Maine were collected by Charles V. Guidotti in the summer of 1998. Reviews by Henry Berry, IV and Peter Thompson greatly improved the manuscript.

References

BENNETT, M.A. (1987): Genesis and diagenesis of the Cambrian manganese deposits, Harlech, north Wales. *Geol. J.* **22**, 7-18.

________ (1989): Quartz–spessartine metasediments (coticules) and their protoliths in north Wales. *Geol. Mag.* **126**, 435-442.

BERRY, H.N., IV (1986): *Bedrock Geology of the Camden Hills, Maine*. M.S. thesis, Univ. of Maine, Orono, Maine.

________ & OSBERG, P.H. (1989): A stratigraphic synthesis of eastern Maine and western New Brunswick. *In* Structure and Stratigraphy – Studies in Maine Geology **2** (R.D. Tucker & R.G. Marvinney, eds.). Maine Geological Survey, Augusta, Maine (1-32).

BONATTI, E. (1975): Metallogenesis at oceanic spreading centers. *Annu. Rev. Earth Planet. Sci*. **3**, 401-431.

BOYNTON, W.V. (1984): Cosmochemistry of the rare earth elements: meteorite studies. *In* Rare Earth Element Geochemistry (P. Henderson, ed.). Elsevier, New York, N.Y. (63-114).

CLIFFORD, T.N. (1960): Spessartine and magnesium biotite in coticule-bearing rocks from Mill Hollow, Alstead Township, New Hampshire, USA: a contribution to the petrology of metamorphosed manganiferous sediments. *Neues Jahrb. Mineral., Abh.* **94**, 1369-1400.

DOCKA, J.A. (1985): *Petrology and Origins of Mn–Fe Metasediments in New England.* Ph.D. thesis, Harvard Univ., Cambridge, Massachusetts.

ELDERFIELD, H. & GREAVES, M.J. (1983): Determination of the rare earth elements in sea water. *In* Trace Metals in Sea Water (C.S. Wong, E.A. Boyle, K.W. Bruland, J.D. Burton & E.D. Goldberg, eds.). Perseus Books, Cambridge, Massachusetts (427-445).

EMERSON, B.K. (1898): Geology of Old Hampshire County, Massachusetts, comprising Franklin and Hampden counties. *U.S. Geol. Surv., Monogr.* **29**.

EUSDEN, J.D., JR. (1988): Stratigraphy, structure, and metamorphism across the "dorsal zone", central New Hampshire. *In* Guidebook for Field Trips in Southwestern New Hampshire, Southeastern Vermont, and North-Central Massachusetts (W.A. Bothner, ed.). *New England Intercoll. Geol. Conf. Guidebook*, 40-59.

________, BOTHNER, W.A., HUSSEY, A.M., II & LAIRD, J. (1984): Silurian and Devonian rocks in the Alton and Berwick quadrangles, New Hampshire and Maine. *In* Geology of the Coastal Lowlands Boston, MA to Kennebunk, ME (L.S. Hanson, ed.). *New England Intercoll. Geol. Conf. Guidebook*, 325-351.

________ & LYONS, J.B. (1993): The sequence of Acadian deformations in central New Hampshire. *In* The Acadian Orogeny; Recent Studies in New England, Maritime Canada, and the Autochthonous Foreland (D.C. Roy & J.W. Skehan, eds.). *Geol. Soc. Am., Spec. Pap.* **275**, 51-66.

GROMET, P.L., DYMEK, R.F., HASKIN, L.A. & KOROTEV, R.L. (1984): The "North American shale composite": its compilation, major and trace element characteristics. *Geochim. Cosmochim. Acta* **48**, 2469-2482.

GUIDOTTI, C.V. (1970): Metamorphic petrology, mineralogy and polymetamorphism of a portion of NW Maine. *In* Guidebook for Field Trips in the Rangeley Lakes – Dead River Basin region, western Maine (G.M. Boone, ed.). *New England Intercoll. Geol. Conf. Guidebook* **B2**, 1-29.

________ (2000): The classic high-T – low-P metamorphism of west central Maine. Is it post-tectonic or syntectonic? Evidence from porphyroblast–matrix relations: discussion. *Can. Mineral.* **38**, 995-1006.

________, TEICHMANN, F. & HENRY, D.J. (1991): Chlorite-bearing polymetamorphic metapelites in the Rangeley area, Maine; evidence for equilibrium assemblages. *Am. Mineral.* **76**, 867-879.

HATCH, N.L., JR., MOENCH, R.H. & LYONS, J.B. (1983): Silurian – Lower Devonian stratigraphy of eastern and south-central New Hampshire; extensions from western Maine. *Am. J. Sci.* **283**, 739-761.

HAYNES, B.W., LAW, S.L. & BARRON, D.C. (1986): An elemental description of Pacific manganese nodules. *Mar. Mining* **5**, 239-276.

KRAMM, U. (1976): The coticule rocks (spessartine quartzites) of the Venn–Stavelot Massif, Ardennes, a volcanoclastic metasediment? *Contrib. Mineral. Petrol.* **56**, 135-155.

KROSSE, S. & SCHREYER, W. (1993): Comparative geochemistry of coticules (spessartine-quartzites) and their Redschist country rocks in the Ordovician of the Ardennes Mountains, Belgium. *Chem. Erde* **53**, 1-20.

LAMENS, J., GEUKENS, F. & VIAENE, W. (1986): Geological setting and genesis of coticules (spessartine metapelites) in the Lower Ordovician of the Stavelot Massif, Belgium. *J. Geol. Soc.* **143**, 253-258.

LOTTERMOSER, B.G. (1989): Rare earth element study of exhalites within the Willyama Supergroup, Broken Hill block, Australia. *Mineral. Deposita* **24**, 92-99.

LUDMAN, A. (1978): Stratigraphy, structure, and progressive metamorphism of lower Paleozoic rocks in the Calais area, southeastern Maine. *New England Intercoll. Geol. Conf. Guidebook*, 78-101.

MARCHIG, V., GUNDLACH, H., MÖLLER, P. & SCHLEY, F. (1982): Some geochemical indicators for discrimination between diagenetic and hydrothermal metalliferous sediments. *Marine Geol.* **50**, 241-256.

MCLENNAN, S.M. (1989): Archean sedimentary rocks and the Archean mantle. *In* Workshop on the Archean Mantle (L.D. Ashwal, ed.). *Lunar and Planetary Inst., Tech. Rep.* **89-05**, 57-59.

OSBERG, P.H. & BERRY, H.N., IV (1991): Stratigraphy of lithotectonic belts west of Penobscot Bay, Maine. *In* Geology of the Coastal Lithotectonic Block and Neighboring Terranes, Eastern Maine and Southern New Brunswick (A. Ludman, ed.). *New England Intercoll. Geol. Conf. Guidebook*, 358-373.

RANKIN, P.C. & GLASBY, G.P. (1979): Regional distribution of rare earth and minor elements in manganese nodules and associated sediments in the southwest Pacific and other localities. *In* Marine Geology and Oceanography of the Pacific Nodule Province (J.L. Bischoff & D.Z. Piper, eds.). Perseus Books, Cambridge, Massachusetts (681-693).

RENARD, A. (1878): Sur la structure et la composition minéralogique du coticule et sur ses rapports avec le phyllade oligistifère. *Mém. couronnés de l'Académie Royale de Belgique* **41**.

RUHLIN, D.E. & OWEN, R.M. (1986): The rare earth element geochemistry of hydrothermal sediments from the East Pacific Rise: examination of a seawater scavenging mechanism. *Geochim. Cosmochim. Acta* **50**, 393-400.

SCHILLER, E.A. & TAYLOR, F.C. (1965): Spessartine–quartz rocks (coticules) from Nova Scotia. *Am. Mineral.* **50**, 1477-1481.

SCHREYER, W., BERNHARDT, H.-J. & MEDENBACH, O. (1992): Petrologic evidence for a rhodochrosite precursor of spessartine in coticules of the Venn–Stavelot Massif, Belgium. *Mineral. Mag.* **56**, 527-532.

SPRY, P.G. (1990): Geochemistry and origin of coticules (spessartine–quartz rocks) associated with metamorphosed massive sulfide deposits. *In* Regional Metamorphism of Ore Deposits and Genetic Implications (P.G. Spry & L.T. Bryndzia, eds.). VSP, Utrecht, The Netherlands (49-75).

________, PETER, J.M. & SLACK, J.F. (2000): Meta-exhalites as exploration guides to ore. *Rev. Econ. Geol*. **11**, 163-201.

________ & WONDER, J.D. (1989): Manganese-rich garnet rocks associated with the Broken Hill lead–zinc–silver deposit, New South Wales, Australia. *Can. Mineral.* **27**, 275-292.

THOMPSON, P.J. (1985): *Stratigraphy, Structure, and Metamorphism in the Monadnock Quadrangle, New Hampshire.* Ph.D. thesis, Univ. Massachusetts, Amherst, Massachusetts.

THOMSON, J.A. (1992): A mineralogically and chemically zoned granulite-facies coticule from the lower Silurian Rangeley Formation, south-central Massachusetts. *Can. Mineral.* **30**, 393-413.

WONDER, J.D. (1987): *The Origin of Manganese-Rich Metasediments and their Relationship to Iron Formation and Base Metal Deposits, Western Georgia Piedmont.* M.S. thesis, Iowa State Univ., Ames, Iowa.

________, SPRY, P.G. & WINDOM, K.E. (1988): Geochemistry and origin of manganese-rich rocks related to iron-formation and sulfide deposits, western Georgia. *Econ. Geol.* **83**, 1070-1081.

WOODLAND, A.W. (1939): The petrography and petrology of the Lower Cambrian manganese ore of west Merionethshire. *Quart. J. Geol. Soc. London* **95**, 1-35.

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