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Pumpellyite-dominated metadomain alteration at Builth Wells, Wales—evidence for a fossil submarine hydrothermal system?

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ABSTRACT. Two metamorphic alteration styles have been observed in basic lavas and volcaniclastic rocks of the Builth Volcanic Series of Ordovician age exposed at Llanelwedd Quarry, in the Builth Inlier, Wales. The first alteration style is characterized by the development of a mineral assemblage comprising albite + chlorite + sphene + pumpellyite ± prehnite ± calcite ± white mica, although original textures are largely preserved. This alteration pattern corresponds with that developed in volcanic and volcaniclastic rocks elsewhere in Wales, and which is attributed to the effects of burial. The second alteration style is characterized by the development of metadomains dominated by pumpellyite or, more rarely, prehnite. In the metadomains no original textures are seen in hand specimen. This alteration pattern has, so far, not been observed elsewhere in Wales, and it is here suggested that it might result from a submarine hydrothermal circulation system.

KEYWORDS: pumpellyite, metadomains, metamorphism, Builth Wells, Wales.

THE Builth Inlier, Powys, Wales, exposes a sequence of sedimentary, volcanic and volcaniclastic rocks which accumulated at the margin of the Welsh Basin during Ordovician times. The extensive quarry at Llanelwedd (National Grid Ref. SO 051 521), 1 km northeast of Builth Wells, provides an excellent section through westward-dipping basic lavas and associated volcaniclastic rocks of Llanvirn age, which form a part of the Builth Volcanic Series (Jones and Pugh, 1949). Elsewhere within the series, but not exposed at Llanelwedd, are andesitic to dacitic lavas and intrusions, and rhyolitic tuffs. Minor intrusions of basic composition occur in the inlier, but these invade strata of Caradoc age, and are not related to volcanism associated with the development of the Builth Volcanic Series.

Basic lavas of the Builth Volcanic Series are typically feldspar-phyric, strongly vesicular and generally massive, although pillowed flows and hyaloclastites are locally developed. This, coupled with the nature of the associated volcaniclastic sediments both above and below the lavas, suggests that, for much of the time, lava eruption and emplacement occurred in a submarine environment. Jones and Pugh (op. cit.), however, described the presence of a shore-line environment during accumulation of the sequence, and it is likely that 'volcanic islands' existed periodically during development of the pile. The volcaniclastic sandstones which overlie the lava pile (the Newmead Sands) appear to have been deposited in a shallow marine environment and provide clear evidence of contemporaneous erosion.

Chemically the basalt-andesite-dacite-rhyolite volcanic rocks of the Builth Volcanic Series show calc-alkaline affinities (Furnes, 1978), which contrasts with the predominantly tholeiitic basalt-rhyolite character of lavas from other Ordovician sequences in the Welsh Basin (Kokelaar *et al.*, 1984; Bevins *et al.*, 1984).

The Lower Palaeozoic rocks of the Welsh Basin and adjacent areas have suffered low-grade regional metamorphism, generally within the prehnite-pumpellyite facies (Bevins and Rowbotham, 1983), although there is probably a complete range from zeolite facies through to greenschist facies (Bevins *et al.*, in press). Metamorphic alteration has been attributed to the effects of burial (Bevins and Rowbotham, op. cit.), although it has been suggested that in Snowdonia and Llŷn the grade may have been influenced, at least in part, by the intensity of deformation (Merriman and Roberts, 1985).

Metamorphic alteration in the Builth Inlier. Two metamorphic alteration styles can be observed within the volcanic and volcaniclastic rocks of the Builth Volcanic Series at Llanelwedd. First, and most widely developed, is a mineral assemblage in basic lavas and volcaniclastics comprising albite + chlorite + sphene ± pumpellyite ± prehnite ± calcite ± white mica, sometimes with relict clinopyroxene. These secondary minerals occur in varying amounts in rocks which, although altered, generally preserve original textures which are visible in both hand specimen and thin section. The second alteration style, occurring in basic lavas and associated volcaniclastics, is characterized by the presence of metadomains dominated by pumpellyite or, more rarely, prehnite. In these rocks, virtually no original textures are seen in hand specimen, but are visible when observed microscopically.

The first alteration style corresponds with that present in volcanic rocks (particularly those of basic composition) from other parts of Wales, including the basic intrusions of Caradoc age in the Builth Inlier; this alteration is attributed to the effects of burial (Bevins and Rowbotham, 1983). In the Builth lavas, as elsewhere, prehnite and pumpellyite occur in a variety of forms, most commonly as small inclusions or aggregates of crystals in albitized feldspar and, more rarely, in the groundmass. When calcite is abundant, prehnite and pumpellyite are usually absent and the secondary mineral assemblage is then calcite-chlorite-albite-sphene ± white mica, which is non-diagnostic of metamorphic grade. Chlorite is found in cloudy, albitized feldspar grains, as well as lining vesicles, and also in the groundmass, whilst sphene forms small, dusty, granular aggregates of high relief, usually in, or closely associated with, altered ore grains, or in chloritic areas within the groundmass. Original mafic minerals are invariably pseudomorphed by chlorite, although relict clinopyroxene has been identified. Both actinolite and epidote are rare to absent and the mineral assemblage is diagnostic of the prehnite-pumpellyite facies. In contrast, however, the Ordovician shales exposed in the inlier, and lying both above and below the lavas of the Builth Volcanic Series, contain white micas with illite crystallinity values characteristic of the diagenetic zone (D. Robinson and R. E. Bevins, unpublished data), i.e. below the prehnite-pumpellyite facies. Some support for a lower metamorphic grade in the Builth Inlier is the presence of rare laumontite-calcite veins cutting the basic lavas in Llanelwedd Quarry; however, these are interpreted as being related to a late-stage, retrogressive event, although no such veins have

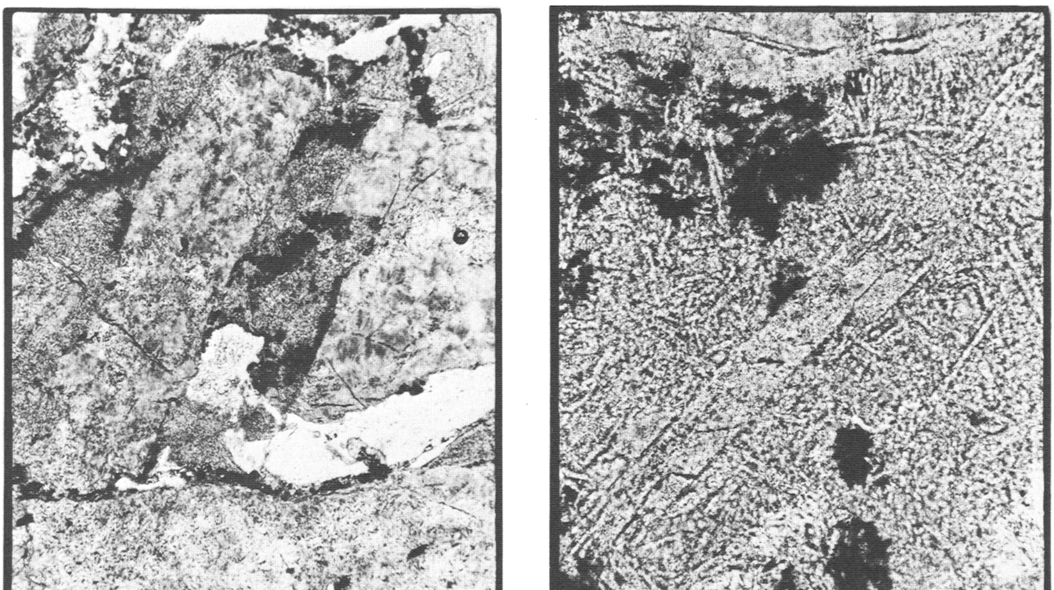

FIG. 1. (*a*) Plagioclase feldspar phenocryst and groundmass replaced by fine grained pumpellyite. Sample BW82/3. Magnification ×55. PPL. (*b*) Plagioclase feldspar microphenocryst and groundmass microlites entirely pseudomorphed by pumpellyite. Sample BW82/3. Magnification ×140. PPL.

been observed so far elsewhere in Wales. In view of the widespread occurrence of prehnite and pumpellyite in the inlier, not only in the basic lavas but also in the associated volcaniclastic sediments and in the basic intrusions of Caradoc age, it is considered that the metamorphic grade in the Builth Inlier lies in the prehnite-pumpellyite facies, and is related to burial of the sequence under a cover of younger Ordovician and Silurian strata.

The second alteration style contrasts markedly with that described above and has, so far, not been observed elsewhere in Wales. It is characterized by the development of metadomains dominated by pumpellyite or prehnite and is found in basic lavas and associated volcaniclastic rocks exposed in the eastern part of the main level, Llanelwedd Quarry. Nicholls (1958) described aspects of this alteration, attributing the development of the Ca-rich areas to the segregation of an immiscible liquid from the magma. This suggestion is not thought to be tenable and an alternative model is suggested here.

In hand specimen the rock is fine grained and structureless with no original textures and a distinctive pale blue-green or a milky, pale green hue when pumpellyite or prehnite, respectively, are dominant. Contacts between the alteration metadomains and normal basic lava (showing the first described alteration style) are generally sharp and are readily seen in the loose blocks on the quarry floor. The metadomains commonly define large, crudely ovoid, alteration patches, which show poorly defined concentric zones. Irregular calcite veins, some containing carbon, cut through these metadomains and in most cases appear to post-date the pumpellyitization and prehnitization episodes; in a number of cases, however, the evidence for the timing of events is ambiguous. When extensively developed these veins give the rock a brecciated appearance, particularly on weathered surfaces, following leaching of the calcite. No field evidence has so far been found to demonstrate the relationship of these veins with the laumontite-calcite veins.

In thin section the pumpellyite metadomains are characterized by the extensive development of dark green to colourless, strongly pleochroic pumpellyite which replaces virtually all of the primary minerals. Under crossed polars no primary igneous textures can be seen, but in plane light original crystal outlines can be observed (fig. 1). In the pumpellyite metadomains sparse prehnite is present along with calcite and chlorite in vesicles and also large, irregular patches that resemble vesicles, but which, in detail, are more vein-like. Such 'veins' are seen only in lavas which have suffered pumpellyitization and these features are thought not to be primary vesicles, but to have originated in the alteration episode. More rarely prehnite along with calcite and minor pumpellyite (fig. 2) infills former plagioclase phenocrysts. It is typically coarse and forms large, radiating, sometimes bow-tie shaped, masses. In thin section, when coarse, it is colourless but where fine grained it assumes a pale brown colour. Areas which have suffered prehnitization are relatively rare in comparison with the pumpellyite-dominated metadomains. However, loose blocks of coarse volcaniclastic sandstone, most probably from the underlying Pebbly Feldspar Ash (Jones and Pugh, 1949), collected from the main level at Llanelwedd Quarry, are pale green in colour with milky white to pale-green, irregular to ovoid prehnitized patches, up to 2 cm in diameter. Thin section examination reveals variable prehnite development, both in the clasts and the matrix. Many clasts are totally replaced by large, radiating prehnite crystals. Original textures and clast outlines are, for the most part, only observed in plane polarized light and

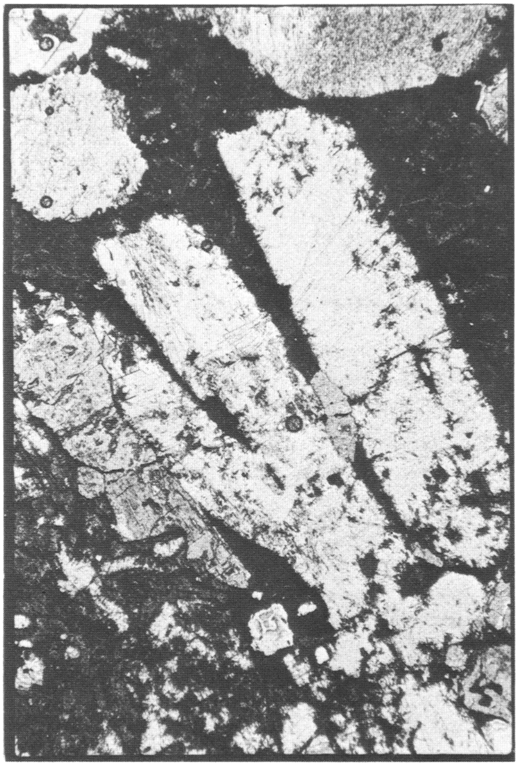

FIG. 2. Plagioclase feldspar phenocrysts pseudomorphed by coarse, radiating, colourless prehnite. Groundmass between feldspar phenocrysts composed predominantly of fine-grained pumpellyite. Specimen BW82/3. Magnification ×23. PPL.

TABLE I. Representative analyses of pumpellyite

SiO_2	36.42	35.32	35.00	35.72	35.88	35.50	35.74	35.35	35.24	35.16	35.86
TiO_2	n.d.	n.d.	n.d.	n.d.	n.d.	0.18	n.d.	n.d.	n.d.	n.d.	n.d.
Al_2O_3	15.36	16.57	16.73	18.67	18.44	16.22	16.06	16.21	16.51	15.79	18.30
Fe_2O_3*	16.21	15.93	15.23	14.04	13.74	15.84	15.48	15.82	15.81	16.10	14.58
MgO	2.73	2.14	2.26	1.97	2.08	1.98	2.39	2.40	2.18	2.04	2.06
CaO	21.58	21.29	20.98	21.82	22.07	21.72	21.39	21.35	21.40	21.48	22.04
Na_2O	n.d.	n.d.	n.d.	n.d.	n.d.	n.d.	n.d.	n.d.	0.43	n.d.	n.d.
K_2O	n.d.	n.d.	n.d.	n.d.	n.d.	n.d.	n.d.	n.d.	n.d.	n.d.	n.d.
Total	92.30	91.25	90.20	92.22	92.21	91.44	91.06	91.13	91.57	90.57	92.84
Recalculation based on 16 cations											
Si	6.21	6.09	6.08	6.04	6.06	6.11	6.16	6.09	6.02	6.12	6.03
Al	-	-	-	-	-	-	-	-	-	-	-
ΣZ	6.21	6.09	6.08	6.04	6.06	6.11	6.16	6.09	6.02	6.12	6.03
Al^Y	3.09	3.37	3.43	3.72	3.67	3.29	3.27	3.29	3.33	3.24	3.63
Fe^Y	0.91	0.63	0.57	0.28	0.33	0.69	0.73	0.71	0.67	0.76	0.37
Ti^Y	-	-	-	-	-	0.02	-	-	-	-	-
ΣY	4.00	4.00	4.00	4.00	4.00	4.00	4.00	4.00	4.00	4.00	4.00
Al*	-	-	-	-	-	-	-	-	-	-	-
Fe*	1.17	1.44	1.42	1.51	1.42	1.36	1.28	1.34	1.36	1.35	1.48
Mg	0.69	0.55	0.59	0.50	0.52	0.51	0.61	0.62	0.56	0.53	0.52
ΣX	1.86	1.99	2.01	2.01	1.94	1.87	1.89	1.96	1.92	1.88	2.00
Ca	3.94	3.93	3.91	3.95	3.99	4.01	3.95	3.94	3.92	4.01	3.97
Mn	-	-	-	-	-	-	-	-	-	-	-
Na	-	-	-	-	-	-	-	-	0.14	-	-
K	-	-	-	-	-	-	-	0.02	-	-	-
ΣW	3.94	3.93	3.91	3.95	3.99	4.01	3.95	3.96	4.06	4.01	3.97

* Total iron as Fe_2O_3

n.d. = not detected

pumpellyite is only sparsely developed in these prehnite metadomains.

Mineral Chemistry. Electron probe microanalyses of the important mineral phases in a sample (BW 77/3) of a pumpellyite metadomain from Llanelwedd Quarry have been determined at the Department of Geology, University of Manchester, using a Geoscan Mk. II, operating at 15 kV and 3.5×10^{-8} amps.

Pumpellyite analyses (Table I) have been recalculated assuming a total of sixteen cations (see Coombs *et al.*, 1976). Recalculated in this manner Si is generally close to the ideal six cations. Fe contents (assumed to be Fe^{3+}; see Liou, 1979) are noticeably high compared with other pumpellyites in Wales (reported in Bevins and Rowbotham, 1983), with up to 16.2 wt. % Fe_2O_3 and, indeed, are the most Fe-rich samples reported from Wales (fig. 3). Slight variations are present in Fe contents in BW 77/3 pumpellyites and are reciprocal to variations in Al. MgO is generally low, reaching a maximum of 2.7 wt. %. The pumpellyites compare closely with those from other low-pressure zeolite and prehnite-pumpellyite facies terrains, including Vancouver Island (Surdam, 1969; Kuniyoshi and Liou, 1976) and the East Taiwan Ophiolite (Liou, 1979). Pumpellyites of similar composition also occur in the zeolite and pumpellyite zones of the Del Puerto Ophiolite submarine hydrothermal system (Evarts and Schiffman, 1983).

Analyses of prehnite in BW 77/3 (Table II) similarly show high Fe contents, with between 6.2 and 8.2 wt. % Fe_2O_3, again in excess of that reported from elsewhere in Wales (Bevins and Rowbotham, 1983). The high Fe^{3+} contents of the pumpellyites and prehnites described here probably reflect high oxygen fugacity at the time of crystallization.

Two analyses of chlorite have been made and, following the classification of Hey (1954), are diabantites. They are relatively Si-rich in comparison with other chlorites from Wales, which generally plot in the brunsvigite, pycnochlorite, or ripidolite fields (see Bevins and Rowbotham, 1983). Chlorites similar in composition to those reported here have been recorded from other low-pressure zeolite or prehnite-pumpellyite facies terrains, including the zeolite and pumpellyite zones of the Del Puerto Ophiolite (Evarts and Schiffman, 1983).

Origin of the Builth metadomains. As the regional metamorphic grade in the Builth Inlier is in the prehnite-pumpellyite facies, the simplest model is to relate generation of the metadomains to the burial episode. However, the field and petrographic evidence described above strongly suggests that the two alteration types belong to separate events. The metadomain alteration is not regional in extent, being only observed in Wales to date at Builth; it is more typical of a hydrothermal type of alteration event. This implies an origin at an early stage in the alteration history of the Builth sequence.

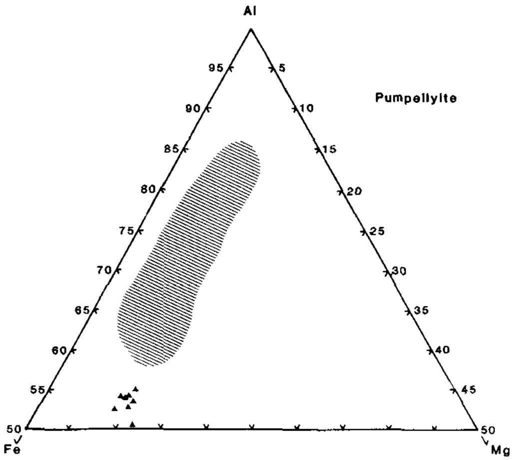

FIG. 3. A portion of the Al:total Fe:Mg diagram, showing compositions of pumpellyites from Builth Wells (triangles), along with the field of other pumpellyites from Wales (shaded).

TABLE II. Representative analyses of prehnite

	1	2	3	4	5	6
SiO_2	41.97	42.09	42.21	41.98	42.18	42.05
TiO_2	n.d.	n.d.	n.d.	n.d.	n.d.	n.d.
Al_2O_3	18.00	18.57	18.66	18.60	19.42	19.23
Fe_2O *	8.23	7.79	7.02	7.11	6.25	6.57
MgO	n.d.	n.d.	n.d.	n.d.	0.25	n.d.
CaO	25.38	25.60	25.46	25.48	25.91	25.96
Na_2O	n.d.	n.d.	n.d.	n.d.	n.d.	n.d.
K_2O	n.d.	n.d.	n.d.	n.d.	n.d.	n.d.
Total	93.58	94.05	93.35	93.17	94.01	93.81
Structural formulae based on 22 oxygens						
Si	6.06	6.04	6.09	6.07	6.03	6.03
Al^{iv}	-	-	-	-	-	-
ΣZ	6.06	6.04	6.09	6.07	6.03	6.03
Al^{vi}	3.07	3.14	3.16	3.17	3.27	3.25
Ti	-	-	-	-	-	-
Fe^{3+}	0.90	0.84	0.76	0.77	0.67	0.71
ΣY	3.97	3.98	3.92	3.94	3.94	3.96
Mg	-	-	-	-	0.05	-
Ca	3.93	3.94	3.94	3.95	3.97	3.99
Na	-	-	-	-	-	-
K	-	-	-	-	-	-
ΣX	3.93	3.94	3.94	3.95	4.02	3.99

* total iron as Fe_2O_3

n.d. = *not detected*

Pervasive alteration, involving the production of monomineralic metadomains or metadomains dominated by a limited number of phases, as described here, provides evidence of extensive element mobility (Smith, 1968), involving high water/rock ratios. Elsewhere, these alteration patterns have been related to areas of increased permeability, such as in the vicinity of faults or fractures, or at the vesicular margins of lava flows, as described by Smith (1968) and Jolly and Smith (1972). At Llanelwedd Quarry, no relationship between the development of metadomains and flow tops or bases is observed. In addition, although faults are an attractive method for control of fluid passage, and there are a number of important faults in the quarry area, field evidence to support this is, again, lacking.

If the metadomain alteration pattern at Llanelwedd Quarry is the result of hydrothermal alteration, a heat source must have been available to drive a small, localized, hydrothermal circulation system. Evidence for this heat source can be found in the Felsite Agglomerate of Jones and Pugh (1949), which is one of the volcaniclastic horizons interbedded with the lava pile, and which forms the western face of the main level at Llanelwedd Quarry. The origin of this unit is problematical (see Furnes, 1978; Baker and Hughes, 1979), but of relevance here is the fact that it contains abundant clasts, generally in the range 10–15 cm in diameter but reaching up to 1 m, of intrusive and extrusive igneous rocks which vary in composition from basic through intermediate to acid. The intrusive clasts suggest the presence of an igneous complex or magma chamber at depth. Heat from this source may have been responsible for driving a small, localized, hydrothermal circulation system, the effects of which were possibly concentrated along a fault or fault system(s) with fluid being derived either from seawater which had penetrated into the roof rocks overlying the heat source or from the still wet, unlithified sediments and volcaniclastics. Support for this model is provided by the fact that some of the clasts incorporated in the Felsite Agglomerate are extensively altered, containing abundant pumpellyite, in contrast to adjacent clasts which, although of similar composition, do not show such intense alteration. This is here interpreted as indicating an early stage of pumpellyite development, occurring before the clasts were incorporated into the Felsite Agglomerate deposit.

Conclusions. It is suggested here that generation of the pumpellyite- and prehnite-dominated metadomains in basic lavas of Ordovician age in the Builth Inlier was related to a submarine hydrothermal circulation system, the heat to drive the system being provided by a high level magma chamber or intrusion complex. A similar model has recently been forwarded to account for extensive alteration of the Del Puerto Ophiolite (Evarts and Schiffman, 1983) and although the Builth volcanic rocks do not form part of an ophiolite system, a similar heat source to that at Del Puerto can be envisaged. Evarts and Schiffman (op. cit.) suggested that the Del Puerto Ophiolite was generated in an island arc environment, with an intrusion of basic magma within 3 km of the floor resulting in closed-system fractionation. This caused heating of seawater which had descended through the roof rocks, leading to the development of a hydrothermal circulation system. Although the scale of alteration is much larger in the case of the Del Puerto Ophiolite than at Builth there are obvious similarities.

More detailed work is in progress, aimed particularly at gaining an understanding of the nature of the fluids responsible for the metadomain alteration at Builth.

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REFERENCES

Baker, J. W., and Hughes, C. P. (1979) *Proc. Geol. Assoc.* **90,** 65–79.

Bevins, R. E., and Rowbotham, G. (1983) *Geol. J.* **18,** 141–67.

——Kokelaar, B. P., and Dunkley, P. N. (1984) *Proc. Geol. Assoc.* **95,** 337–47.

——Oliver, G. J. H., and Thomas, L. J. (in press) *Earth Evolution Sciences.*

Coombs, D. S., Nakamura, Y., and Vuagnat, M. (1976) *J. Petrol.* **17,** 440–71.

Evarts, R. C., and Schiffman, P. (1983) *Am. J. Sci.* **283,** 289–340.

Furnes, H. (1978) Unpubl. D.Phil. thesis, Univ. of Oxford.

Hey, M. H. (1954) *Mineral. Mag.* **30,** 277–92.

Jolly, W. T., and Smith, R. E. (1972) *J. Petrol.* **13,** 273–309.

Jones, O. T., and Pugh, W. J. (1949) *Q. J. geol. Soc. London,* **105,** 65–99.

Kokelaar, B. P., Howells, M. F., Bevins, R. E., Roach, R. A., and Dunkley, P. N. (1984) In *Marginal Basin Geology: Volcanic and associated sedimentary and tectonic processes in modern and ancient marginal basins.* Spec. Publ. Geol. Soc. London, **16,** 245–69.

Kuniyoshi, S., and Liou, J. G. (1976) *Am. J. Sci.* **276,** 1096–119.

Liou, J. G. (1979) *Am. Mineral.* **64,** 1–14.

Merriman, R. J., and Roberts, B. (1985) *Mineral. Mag.* **49,** 305–19.

Nicholls, G. D. (1958) *Q. J. geol. Soc. London,* **114,** 137–62.

Smith, R. E. (1968) *J. Petrol.* **9,** 191–219.

Surdam, R. C. (1969) *Am. Mineral.* **54,** 256–66.

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