Oxygen and Hydrogen Isotope Compositions as Indicators of Granite Genesis in the New England Batholith, Australia

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Abstract. Oxygen and hydrogen isotope studies of a number of granite suites and mineral separates from the New England Batholith indicate that δO^{18} can be used to discriminate the major granite protoliths. The granite suites previously subdivided on the basis of mineralogical and geochemical criteria into S-type (sedimentary) and I-type (igneous) have δO^{18} values consistently higher in the S-type granites (10.4-12.5) than in the spatially related I-type plutons (7.7–9.9). There appears to be a systematic variation in δO^{18} from the most S-type to the most I-type granites, the dividing point between the two occuring at δO^{18} equal to 10. A group of leucocratic granites that form about half of the batholith and difficult to classify mineralogically and geochemically is found to have low δO^{18} values (6.4–8.1), suggesting an affinity to the most I-type granites. A single leucogranite pluton with minor muscovite has a δO^{18} of 9.6 which is significantly higher than other leucogranites indicating a different origin perhaps involving amphibole fractionation.

The behavior of δD in the plutonic rocks is much less systematic than δO^{18} . Excluding samples collected adjacent to major faults, the δD values show a rough positive correlation with water content similar to, but less pronounced than, the trend previously observed in the Berridale Batholith, southeastern Australia. This relation is considered to reflect an interaction between meteoric water and the granites, the largest effect being observed in samples with the least amount of water. Of note is the generally lower δ D values of the upper Paleozoic New England Batholith compared with the Silurian Berridale Batholith. This difference may be related to a near equatorial paleolatitude of 22° S in the Silurian and near polar paleolatitudes in the late Carboniferous that have been inferred for these regions. Granite samples collected from near major faults, and one ignimbrite sample of rhyodacite composition, have very low δ D values (less than -120) suggesting a much greater degree of interaction with meteoric water.

1. Introduction

This paper documents the oxygen and hydrogen isotope chemistry of a large upper Paleozoic calc-alkalic batholith. Field mapping and petrological studies of the New England Batholith indicate that at least four major suites of granites can be recognized in addition to a large group of leucocratic granite plutons. Isotopic data emphasize the differences between the suites and are used to evaluate aspects of the genesis of these rocks.

2. The New England Batholith

The New England Batholith is predominantly granite and leucocratic granite (Wilkinson, 1969) in contrast with many Cordilleran batholiths where granodiorite, tonalite and diorite from a significant proportion (Bateman and Dodge, 1970; Reed and Lanphere, 1974; Gastil et al., 1974; Pitcher, 1974). In New England over 120 plutons or plutonic complexes have been recorded (Pogson and Hitchins, 1973) and 2 distinctive granite suites named: the Hillgrove Plutonic Suite (Binns et al., 1967) and the Bundarra Plutonic Suite (Flood and Shaw, 1975). Each of these suites consists of spatially related plutons with mineralogical and geochemical characteristics that indicate derivation from a pelitic parentage. These suites have many of the S-type properties that have been described by Chappell and White (1974). Preliminary Rb/Sr dating (Flood and Shaw, 1977a) indicates that these suites are late Carboniferous in age and predate the main hornblende-biotite and biotite granites and leucogranites of late Permian and Early Triassic age (Evernden and Richards, 1962; Cooper et al., 1963).

Flood and Shaw (in preparation) propose further subdivisions of the remainder of the Batholith into two hornblende-biotite suites (Moonbi and Uralla Plutonic Suites) with properties similar to the I-type of Chappell and White (1974) and a less well defined group of leucocratic granites. For convenience in this paper the leucocratic granites will be referred to as the leucogranite suite, although preliminary data indicate that the plutons may have diverse origins and in this sense are not regarded as equivalent to the other suites in rank. Figure 1 shows the approximate distribution of the five suites together with sample localities.

The granite suites have been distinguished on the basis of detailed studies of parts of the Batholith and subsequently extended to include the remainder. Recent and more extensive sampling has shown that some plutons have characteristics intermediate between two suites, specifically between the Moonbi and Uralla Suites and perhaps between Uralla and Hillgrove Suites. As some areas of the Batholith are not well documented, a degree of uncertainty exists as to the affinity of some specific plutons. However, these uncertainties are not likely to affect greatly the areas outlined in Figure 1 for the four main suites. The samples selected for this study are considered to typify most closely examples of the various suites and as a consequence the sampling may be biased against the transitional types.

Fig. 1. Map showing the major subdivisions of the New England Batholith and sample locations. The sample numbers are those listed in Table 1

3. Petrology of the Granite Suites

Bundarra Plutonic Suite. This suite comprises a group of granite plutons occupying 25% of the area of the batholith within a 300-km north-south belt on the western margin of the batholith (Flood and Shaw, 1975). The minor minerals, which average 4%, are an aluminous biotite, cordierite, ilmenite and minor, possibly sub-solidus, muscovite. The suite is coarse grained and in part porphyritic with megacrysts of K-feldspar up to 8 cm long. Chemically the Suite shows only limited variation in composition about the low temperature minimum and analyses are consistently corundum normative. The biotite is low in Fe₂O₃/FeO and reflects the "reduced" nature of the Suite. The properties of the Bundarra Suite, apart from a low initial Sr^{87}/Sr^{86} ratio of 0.706, are considered to reflect a strong S-type character.

Hillgrove Plutonic Suite. Members of this suite comprise a series of discrete foliated biotite-granodiorite and granite plutons forming 6% of the batholith (Binns et al., 1967). The color index is somewhat higher than in the Bundarra Suite and consists of a russet-red aluminous biotite, minor ilmenite, and a pyrope-almandine garnet that has been noted in at least three plutons. Some plutons included in the Hillgrove Suite contain minor amounts of low alumina amphibole and may reflect a transition within the suite as defined towards an I-type character. Aluminous biotite and a moderately corundum normative composition point to an S-type character of this suite, but not so strongly as the Bundarra Suite. The initial Sr^{87}/Sr^{86} ratio of 0.706 is similar to the Bundarra Suite. The Hillgrove Suite is marked by a low ("reduced ") $Fe₂O₃/FeO$ ratio.

Moonbi Plutonic Suite. This suite forms 11% of the batholith and consists of several plutons that are widely distributed through the batholith. Dominant features of this Suite include the abundance of pink megacrysts of orthoclase microperthite, biotite and hornblende and accessory sphene and magnetite. Chemically the rocks have a high K_2O/Na_2O ratio in the more mafic types, high Sr, Ba and Rb and high Fe_2O_3/FeO ratios in the biotite and hornblende. The Moonbi Suite is considered to have a very marked I-type character, in that it has a high hornblende/biotite ratio, magnetite and sphene as accessory phases and relict clinopyroxene as cores in some hornblende grains. The initial Sr^{87}/Sr^{86} ratio is low at 0.7047.

Uralla Plutonic Suite. The Uralla Plutonic Suite consists of a group of plutons that range from gabbro through diorite and granodiorite to granite, and constitutes 9% of the Batholith. In age they either predate or are coeval with the Moonbi Suite at approximately 240–255 m.y. Mafic minerals are actinolitic hornblende, an alumina poor redbrown biotite and variable amounts of clino- and orthopyroxene. Ilmenite is an accessory mineral, and trace amounts of sulphide also appear. Both hornblende and biotite have "reduced" or low ratios of $Fe₂O₃/FeO$. The Uralla Suite has less strongly developed I-type features. Specifically the amphibole is sub-aluminous, and most plutons contain abundant biotite and have ilmenite and a trace of sulphide as accessory phases. The initial Sr^{87}/Sr^{86} ratio at 0.706 is somewhat higher than in the Moonbi Suite.

Leucogranites. The plutons known to belong to the four defined suites above constitute only about one-half of the batholith. Some leucogranites are present in the Bundarra Suite as the color index of this suite is around 5, but because of the presence of cordierite and the spatial, textural and compositional relations of all rock types, there can be little doubt as to the affinity of these leucogranites and they are therefore not included in this discussion.

The remaining granite in the Batholith consists largely of biotite leucogranite with some biotite granite. There is a wide distribution of these rock types, and isotopic dating suggests a considerable range in age. The close approach of these granites in composition to the minima of the granite system, together with the paucity of xenoliths and distinctive mineralogy, makes these plutons more difficult to subdivide. At least three subtypes can be recognized: (1) Biotite-hornblendeleucogranite. These granites have up to 5% biotite and a trace (less than 1%) of hornblende and occur as small plutons, some of which predate the Moonbi Suite while others are associated with a biotite-leucogranite intrusive episode around 220 m.y. They contain an iron-rich biotite and an iron-rich tschermakitic hornblende. (2) Biotite-leucogranite. This group has biotite alone as the mafic mineral and includes some biotite granite plutons. The age of this group is probably similar to some of the hornblende-bearing leucogranites with which they are often associated. The biotites of these plutons are partly altered to chlorite, and preliminary analytical data suggest that they have low alumina characteristics similar to those in the hornblende-bearing granite. (3) Biotite-muscovite-leucogranite. Several smaller plutons in the northern part of the batholith contain minor muscovite and an alnminous biotite. K/Ar dating suggests an Late Permian to Early Triassic age for these leucogranites. Mineralogically they have an S-type character but are not related spatially or temporally to the main late Carboniferous period of S-type plutonism.

4. Presentation of Data

The oxygen and hydrogen isotope analyses of whole-rock samples are given in Table 1. Mineral data are listed in Tables 2 and 3. Samples have been grouped by suite, and where possible, the name of the pluton is included in Table 1.

Sample (see Fig. 1)	Field No.	Lat. S.	Long. E.	δO^{18}	δD	H ₂ O $\%$	Rock	Unit				
Bundarra Plutonic Suite (S-type)												
23	FS111	30°23'	150°53′	12.25	-192	0.41	Granite					
24	FS115	30°13'	150°55'	11.94	-90	0.61	Granite					
25	FS129	29°26'	150°59′	12.43	-104	0.36	Granite					
26	FS143	29°16′	150°55'	12.29	-99	0.49	Granite	$\overline{}$				
27	FS420	30°40'	151"17"	12.49	- 92	0.50	Granite	<u>.</u>				
28	FS432	30°34'	151"14"	11.02	-106	0.52	Granite					
29	FS114	30°16'	150°55′	12.25	- 90	0.42	Granite	$\overline{}$				
Hillgrove Plutonic Suite (S-type)												
18	FS231	30°24'	152°10'	10.95	-110	1.03	Granodiorite	Abroi				
19	FS235	30°26'	152°02′	11.42	-89	0.92	Granodiorite	Rockvale				
20	FS246	30°33'	151°52′	10.58	-85	0.63	Granite	Hillgrove				
21	FS250	30°43'	151°46'	11.82	- 68	0.66	Granite	Blue Knobby				
22	FS252	31°12'	$151^{\circ}45'$	10.35	-72	0.85	Granodiorite	Tia				
Uralla Plutonic Suite (I-type)												
11	FS227	29°54′	151°57′	7.90	-98	0.69	Granite	Wards Mistake				
12	FS244	30°26'	151°43'	9.60	-68	1.08	Granodiorite	Tilbuster				
13	FS240	30°27'	151°21'	9.92	-87	0.63	Granite	Gwydir River				
14	BDN5	30°38′	151°21′	8.93	86 $\overline{}$	0.66	Granodiorite	Balala				
15	DA1	30°48'	151°27'	8.76	61 -	1.42	Diorite	Kentucky				
16	UQ	30°38′	151°29′	9.88	-76	0.87	Granodiorite	Uralla				
17	MD16	30°48'	151°27'	8.64	71	0.84	Diorite	Kentucky				
Moonbi Plutonic Suite (I-type)												
1	FS199	29°10'	152°00′	7.70	-92	0.42	Granite	Bungulla				
\overline{c}	FS204	28°38′	152°08′	7.70	-92	0.62	Granite	Undercliffe Falls				
3	FS224	29°36'	152°06′	8.21	-128	0.56	Granite	Mt. Mitchell				
$\overline{4}$	229	29°07'	152°03′	7.70	-92	0.55	Granite	Bungulla				
5	CC4	30°56′	151°19'	9.12	-91	0.42	Granite	Congi Creek				
6	WR1	30°58′	151°21'	8.62	-97	0.47	Granite	Walcha Road				
7	D439	28°58′	152°11′	8.69	-126	0.96	Granite	Bungulla				
Moonbi Xenoliths												
38	D435	28°58'	$152^{\circ}11'$	6.82	-130	1.02	Xenolith	Bungulla				
39	FS204X	28°38′	152°08′	7.54	- 84	1.23	Xenolith	Undercliffe Falls				
40	WR8XA	30°58'	151°21′	8.91	-91	0.69	Xenolith	Walcha Road				
Leucogranites (mostly I-type)												
30	FS205	28°39′	152°02′	9.60	-102	0.41	Granite	Ruby Creek				
32	FS214	29°15'	151°42′	7.78	-106	0.27	Granite	Mole				
33	FS222	29°38′	$152^{\circ}00'$	7.62	-112	0.29	Granite	Kingsgate				
34	C ₂₉₁	29°04'	$151^{\circ}58'$	6.39	-105	0.29	Granite	MacKenzie				
35	D352	28°53'	$152^{\circ}06'$	7.62	-122	0.27	Granite	Stanthorpe				
36	E283	29°20'	151°54'	8.09	-110	0.24	Granite	Bolivia Range				
37	FS233	30°30'	152°17′	7.74	-98	0.23	Granite	Round Mtn.				
Volcanic Ignimbrite (I-type)												
8	DRVI	29°34'	151°52'	7.03	-121	0.62	Rhyodacite	Dundee				
9	2211	29°05'	152°03′	7.42	-106	0.39	Rhyodacite	Dundee				
10	41712	29°26'	151°40′	7.80	-97	0.52	Rhyodacite	Dundee				

Table 1. Whole-Rock Oxygen and Hydrogen Isotope Compositions and Water Contents

Sample	Suite	Ouartz	K-spar	Biotite	$\Delta Q - Kf$	$\triangle Q - \text{Bi}$
FS252	Н	12.06	10.55	5.56	1.5	6.4
FS420	B	13.88	11.90	8.02	1.9	5.8
FS204	Μ	9.72	7.42	3.62	2.3	6.1
FS240	U	13.48	10.12	6.03	3.3	7.4
FS244	U	11.18	9.88	6.35	1.3	4.8
UQ.	U	10.98	-	6.00	$\overline{}$	4.9
FS205	L.	10.04	8.65	5.52	1.4	4.5
FS222	L	10.47	5.88	1.91	4.6	8.5
C ₂₉₁	L.	7.90	6.35	2.35	1.5	5.5
D352	L	8.65	8.33	2.43	0.3	6.2

Table 2. δO^{18} Relations of Coexisting Minerals

 $H = Hillgrove$; $B = Bundarra$; $U = Uralla$; $L = Leucogranite$; $Kf = Potassium$ feldspar; $A(A-B) = 10^3 \ln (1000 + \delta A)/(1000 + \delta B)$

In addition to the plutonic rocks listed in Table 1 are isotopic results from three samples of the Dundee Rhyodacite (Wilkinson et al., 1964; Flood et al., in press), an ignimbritic flow that is widespread over the northern part of the batholith and that has mineralogical and geochemical properties in common with the Moonbi Suite.

5. Oxygen Isotope Results

Whole Rocks. Values of δO^{18} from the Bundarra and Hillgrove Suites are without exception higher those in the Moonbi and Uralla Suites. Values of the Bundarra Suite (11.0-12.4) overlap but tend to be higher than δO^{18} values of the Hillgrove Suite (10.4-11.8), while those of the Uralla Suite (7.9-9.9) overlap values from the Moonbi Suite (7.7-9.1). These relations are shown diagrammatically in Figure 2, and with the suites ranked vertically from most I-type to most S-type there is a systematic change in the average δ O¹⁸ value increasing from the most I-type (Moonbi) to the most S-type (Bundarra). The higher δO^{18} values in the S-type granites are consistent with an origin involving a protolith that has undergone substantial surface weathering and/or equilibration with sea water (Taylor, 1968 ; Savin and Epstein, 1970; O'Neil and Chappell, 1977).

The petrologic relation of the leucogranites to the four main suites of the batholith is not clear. Absence of distinctive mineralogy and the close approach of the plutons to the ternary minimum composition make it difficult to assess their origin. The leucogranites have a range of δO^{18} values (6.4–9.6) that are much lower than the S-type plutons and lower than many I-type plutons. On the basis of this criterion alone, the leucogranites, with the possible exception of the Ruby Creek pluton, have an I-type origin. The Ruby Creek pluton differs from many other leucogranites in that it contains minor amounts of muscovite and has associated tin mineralization, a feature in common with

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Fig. 2. Variations in δO^{18} values of whole-rock samples, showing the O^{18} enrichments in the S-type rocks relative to the I-type rocks and the similarity between δO^{18} values of leucogranites and I-type rocks

some S-type granites (Chappell and White, 1974). A sample of this leucogranite (FS205) has a δO^{18} value of 9.60 that, although higher than found in other leucogranites, is nevertheless within the range characteristic of most I-type plutons of southeastern Australia. Another leucogranite with similar affinities of muscovite and tin mineralization is the Mole Granite, but a single sample from this body (FS214) has a δ O¹⁸ value of 7.78, which is well within the I-type range. Clearly, more data are required on these two bodies.

Mineral Separates. The chemical composition of biotite is probably a strong indicator of the peraluminous character of the granite (Nockolds, 1947; Albuquerque, 1973). Biotites from the Bundarra and Hillgrove Suites have octahedral A1 in excess of 0.6 atoms, while in the Moonbi and Uralla Suites octahedral A1 is typically less than 0.1 atoms. The hornblende-biotite and biotite leucogranites have low alumina biotites similar to I-type plutons, while the muscovitebiotite leucogranites have exceptionally high octahedral A1 (about one atom). As shown in Figure 3, δO^{18} values of biotite are distinctive for each suite and provide a criterion for origin which may prove better than that based on whole rock values. δO^{18} values of both biotite and whole rock have a range of about 6% ₀₀. The biotites from the Bundarra and Hillgrove Suites are richest in O^{18} primarily because of the S-type character of their protoliths. All else being equal, peraluminous biotites would tend to be richer in O^{18} than normal or subaluminous biotites for thermodynamic reasons, but this effect is minor compared to the obvious differences in O^{18} contents of the parent materials at New England. Biotites from the leucogranites are in general the lowest in O^{18} in keeping with the whole rock data. These samples may have been lowered in O^{18} content by extensive interaction with isotopically light ground water, as discussed below.

Oxygen isotope fractionations between quartz and biotite, $A(O - Bi)$, are relatively large in all rocks: Permil fractionations (Table 2, Fig. 3) are typically around 6% ₀₀ and range from 4.5 to 8.5. On the basis of the quartz-muscovite expression of Bottinga and Javoy (1973) and the observation that in hornblendebearing rocks $\Delta(Q-Mu)=0.49\Delta(Q-Bi)$ (Shieh and Taylor, 1969), the oxygen

Fig. 3. Plot of δO^{18} of biotite versus the permii-fractionation between quartz and biotite. The various suites have distinctive δO^{18} values for biotite, the most O^{18} -rich samples coming from S-type rocks. Isotopic temperatures based on $\Delta(Q - Bi)$ are much too low, implying a subsolidus exchange process

isotope temperatures range from approximately 380° C to 620° C, with a distinct clustering of values around 500° C. Temperatures for the Bundarra and Hillgrove Suites are probably about 40° C lower than indicated on Figure 3, since for muscovitebearing rocks Δ (Q-Mu)=0.54 Δ (Q-Bi) (Garlick and Epstein, 1967). The temperaturesindicate equilibration about 200° C below the water-saturated granite solidus. As many of the northern plutons of the batholith are intruded into volcanic rocks of similar age and chemistry, it is considered that they were high-level granites with solidus temperatures of 700 \degree C or greater at 2 Kb P_{H₂₀} or less (Tuttle and Bowen, 1958). Values of $\Delta(Q-Bi)$ that are seemingly too large for plutonic rocks have been measured by several investigators (Shieh and Taylor, 1969; Taylor and Epstein, 1962; Turi and Taylor, 1971; Shieh and Schwarcz, 1974; Honma and Sakai, 1975; O'Neil and Chappell, 1977). This observation could indicate that oxygen isotopes continue to re-equilibrate between quartz and biotite at temperature below the solidus during prolonged cooling. Alternatively, where indicated temperatures are low, continued interaction with hydrothermal fluids may have differentially lowered the O^{18} content of the biotite. In all the studies of plutonic rocks cited above, and in others,

values of $\Delta(O - B_i) \leq 5$ have been measured. Values between about 3 and 5 are expected if equilibrium oxygen isotope distributions are frozen-in at igneous temperatures. In the present study, 3 of the 10 quartz-biotite pairs analyzed have these low values, two from the Uralla Suite and one leucogranite (Table 2). These rocks have "normal" quartz-feldspar fractionations as well. Thus retrograde exchange between constituent minerals in the granites does not take place in all cases.

As with biotite and whole rocks, the δO^{18} values of potassium feldspar from S-types, I-types, and leucogranites decrease generally in that order. In addition, the δO^{18} values of the feldspars are very close to those of the whole rocks, an observation made in many other studies. Of particular interest are the permil fractionations between quartz and potassium feldspar. Equilibrium values of $\Delta(Q - Kf)$ in igneous rocks should fall in the approximate range of 0.8 to 2.0 (Taylor and Epstein, 1962; O'Neil and Taylor, 1966). Values outside of this range indicate non-equilibrium assemblages and commonly occur because of preferential exchange between feldspar and meteoric water at subsolidus temperature (e.g. Taylor and Turi, 1976; Magaritz and Taylor, 1976). Of the nine pairs analyzed in this study, five appear to be in near equilibrium, one (D352) has a $\Delta(Q-Kf)$ which is too small, and the remaining three have the values 2.3, 3.3, and 4.6. The corresponding values of $\Delta(O-Bi)$ in these latter three are 6.1, 7.4, and 8.5, respectively, corroborating the gross disequilibrium nature of these rocks. Some rocks examined in this study have normal values of $\Delta(Q-Kf)$ and high values of $\Delta(Q-Bi)$ indicating that biotite may be more susceptible than feldspar to the exchange process operative in these systems. On the basis of many measurements of natural and laboratory samples, it is assumed that quartz is considerably more resistant to isotope exchange processes than either feldspar or biotite (e.g. O'Neil and Taylor, 1967; Garlick and Epstein, 1968; Clayton et al., 1968). The relative rates of oxygen isotope exchange between aqueous fluids and either feldspar or biotite are expected to depend markedly on the chemical composition of the fluid (e.g. O'Neil and Taylor, 1967). In a laboratory exchange experiment between a granite gneiss and 0.1 m NaCl solution at 200° C, Cole and Ohmoto (1976) observed that biotite exchanged at a faster rate than did feldspar. In the New England Batholith it is clear that some postmagmatic exchange process has altered the isotopic ratios originally present in many rocks and that biotite was generally more affected than feldspar.

6. Hydrogen Isotope Results

The pattern of δD variations shown in Figure 4 are not as systematic as those of δO^{18} . The Bundarra Suite is generally depleted in deuterium and has a more narrow range of δD values (-90 to -106) than the Hillgrove Suite (-68 to -130). The I-type Uralla Suite is in general the richest in deuterium, and the leucogranites are the most deuterium depleted in the groups analyzed. These two groups essentially do not overlap in δ D values. The two most deuteriumdepleted samples (Moonbi Suite D439 and FS224) were collected near major

Fig. 4. Variations in δ D values of whole-rock samples, showing no clear distinction between S- and I-type rocks. In comparison to the nearby Berridale Batholith the New England Batholith is generally depleted in deuterium

fault structures, and if they are excluded because of possible extensive interaction with ground water, there is no significant difference in δ D values between S- and I-type granites in the New England Batholith. Of the three xenoliths analyzed (Moonbi Suite, Table 1), D435 from the Bungulla unit has a very low δ D value similar to the host granite D439 and must be excluded from discussion for the same reason as given above for D439. The other two xenoliths are slightly enriched in deuterium relative to the host similar to those studied from the nearby Berridale Batholith (O'Neil and Chappell, 1977).

The most noteworthy feature of the deuterium data is the fact that only six δ D values (four from the Uralla Suite alone) are more positive than -85 , a value commonly chosen as a lower limit for "normal" rocks. Of the many hundreds of analyses of deuterium in granitic rocks that have appeared in the literature, those of the New England Batholith can be considered generally low. This is in contrast to the Berridale Batholith where most δD values are more positive than -85 . The Berridale data are shown for comparison in Figures 4 and 5. Interaction with isotopically light meteoric water is generally invoked to explain such deuterium depletion. However, the parent materials could have been low in deuterium, either because of a previous history near the surface of the earth or because there are localized spots in the mantle or lower crust that are relatively low in deuterium.

Chemical compositions of the hydrous minerals also affect the hydrogen isotope compositions of the granites. Suzuoki and Epstein (1976) have shown that the equilibrium distribution of deuterium between either biotite or hornblende and water is mainly a function of Fe/Mg ratio. The higher the Fe/Mg ratio, the more depleted in deuterium is the biotite or hornblende, provided the water retains a constant D/H ratio. This relation has been demonstrated in a series of biotite-hornblende granites from Japan where the hydrous minerals are progressively depleted in deuterium with increasing Fe/Mg ratio (Kuroda

et al., 1974, 1975). In both the Berridale and New England Batholiths (as with most granites), the more mafic types generally contain the most Mg-rich hydrous phases. The leucogranites as well as containing the least amount of mafic minerals (and therefore water), usually have the most Fe-rich mafic phases. Therefore a general relation between δ D and water content might be expected.

Such a relation was observed in the Berridale Batholith (O'Neil and Chappell, 1975) and is shown in Figure 5 along with the data from the New England Batholith. There is much more scatter in the New England data, but apart from three samples very depleted in deuterium (D439 and FS224) there is a rough positive correlation between δD and water content. Isotopically light **ground waters may be expected to affect the granites in a manner proportional to their water contents, the granites with low water being affected to a greater degree than the more mafic types, where the larger "reservoir" of water offers a greater resistance to the isotopic effects of ground water interaction. The three samples that depart significantly from the general pattern were collected near major fault structures, a logical accessway for ground water. In these cases, as well as at least one ignimbrite (DRVI) there is good geologic evidence** to suggest that ground water is responsible for the lowering of the δD values.

7. Significance of Whole Rock 3018 Variations

The following factors are considered to be important in producing the observed bulk rock δO^{18} values from the New England Batholith.

(1) As shown by O'Neil and Chappell (1977), granites with mineralogical and geochemical characteristics indicative of a "sedimentary" source rock (i.e. Bundarra and Hillgrove Suites) have higher δO^{18} values than granites derived from "igneous" source rock (i.e., Moonbi and Uralla Suites). The two groups can be conveniently subdivided at a δO^{18} value of 10.0. This suggests that bulk rock δO^{18} may provide the single most diagnostic criterion for recognizing S and I-type granites.

(2) Studies have shown that metamorphism and the concommitant loss of fluids, dominantly H₂O and CO₂, leads to a reduction in the bulk rock δO^{18} (e.g., Shieh and Taylor, 1969; Shieh and Schwarcz, 1974). Published data suggest that variations in δO^{18} values from about 18-20 in low grade rocks to about 12-15 in high grade rocks are typical. Considering that zones of granite melting will be in high grade rocks, it might be expected that melts would have lower δO^{18} values than the original metasediment. Whole rock δO^{18} values in S-type granites would therefore be expected to vary according to the amount of fluids lost during metamorphism. In a comparison of the New England and Berridale S-type granites of eastern Australia, an initial water content in the very peliticrich rocks of the Lachlan Mobile Belt would be expected to be greater than in the quartz-rich semi-pelites of the New England source region. This may in part explain the generally higher δO^{18} values of the New England S-type granites relative to similar rocks at Berridale.

(3) Like other geochemical parameters, δO^{18} values will depend on the degree of partial melting and the fractionation of O^{18}/O^{16} between the melt and the refractory residue. Inasmuch as O^{18} fractionations become smaller with an increase in temperature, this effect will be less for high temperature "dry" granite melts than for lower temperature "wet" granite melts. The refractory minerals differ between S and I-type granites, and therefore differences in O^{18} content will probably be emphasized between the two granite types. The refractory residue in I-type melting will be mainly calcic plagioclase (andesine), pyroxenes and pargasitic hornblende and possibly biotite and/or magnetite. As these minerals have δO^{18} values similar to or lower than the whole rock in which they occur, a melt with a δO^{18} value greater than the protolith could be predicted. In contrast, the refractory residues of S-type melting would be rich in quartz, with some cordierite, garnet, sillimanite and possibly biotite. As quartz is typically the most O^{18} rich mineral in a rock, its presence in part of a refractory phase may more than offset any tendency of the partial melt to develop δO^{18} values heavier than the protolith. Thus, it is to be expected that I-type granite magma would be richer in O^{18} than its protolith, while for S-type granite magma the O^{18} content would be roughly equal to or less than the protolith. In the same way, the less mafic I-type magma would be expected to be richer in O^{18} than the more mafic I-type magma from the same source.

(4) Related to (3) above, any crystal settling that occurs at higher levels

as the pluton is intruding could also alter the whole rock δO^{18} values. The Kentucky Diorite (Table 1, DA1, MD16) has recently been interpreted as the mafic cumulate-rich base of the Uralla pluton (Table 1, UO; Flood and Shaw, 1977b) and, as might be expected, the hornblende-biotite-rich cumulate has a lower δO^{18} value than the more extensive granodiorite. It is now considered that samples CC4 and WR1 (Table 1) from the Walcha Road pluton may also belong to a single zoned pluton, and although the variation is less marked, the more mafic rock type WR1 has a lower δ O¹⁸ value than the more felsic type CC4. Clearly this effect would be more noticeable in magmas that crystallized only phases with low δO^{18} values like magnetite, ilmenite, pyroxenes and amphiboles, and would become less pronounced as feldspars and quartz begin to crystallize. This type of variation could provide an excellent test for volcanic island arc suites where amphibole fractionation has been suggested to play a dominant role in producing strongly silica oversaturated rocks from "potassic" andesites. Experimental studies (Helz, 1975; Cawthorn and Brown, 1976; Cawthorn and O'Hara, 1976) have shown that amphibole fractionation could be an important mechanism in producing corundum-normative compositions, a feature of S-type granites. Although this is not considered to be a factor in producing the main S-type granites of New England, it could well be a mechanism responsible for generating the muscovite-bearing Ruby Creek leucogranite (Table 1, FS205) spatially and temporally related to the main I-type granites of the Batholith. The δO^{18} value of this intrusion is one of the highest values found for I-types and may well be the result of amphibole fractionation.

(5) The first four points do not explain the very low δO^{18} values of the leucogranites (with the exception of the muscovite-bearing Ruby Creek pluton) in relation to the more mafic I-type granites. Certainly the leucogranites sampled in this study have δO^{18} values suggestive of an I-type protolith, but from the points discussed above the I-type leucogranites might be expected to have higher δO^{18} values than the more mafic I-type granites. The leucogranites may well result from low degrees of partial melting at high temperature. Low degrees of partial melting are inferred from the bulk rock chemistry, and high temperatures inferred from (a) the high level of intrusion of some of these bodies implying dry melting, and (b) the almost complete separation of any phases that may have been refractory, reflecting the lower viscosities of higher temperature melts. The low degrees of partial melting and low δO^{18} values of these leucogranites are consistent with derivation from an I-type protolith that has already been depleted by an earlier partial melting episode.

8. Conclusions

This study emphasizes the results of O'Neil and Chappell (1977) that δO^{18} values can be used to distinguish various granite types previously considered on other geochemical and mineralogical grounds to belong to different protoliths. The isotopic differences arise mainly from the different O^{18} contents of the source materials which may be further modified by the degree of partial melting, separation of refractory minerals and possible fractionation of crystallizing phases.

The relation of the leucogranites to the main Batholith is not clear but the range of δO^{18} values obtained for these rocks indicates an affinity to the I-type plutons. The leucogranites, with the exception of the Ruby Creek pluton, are in the lower range of δO^{18} values and could be interpreted as having developed by low degrees of "dry" partial melting from an I-type source which had previously undergone a partial melting stage. Alternatively the low δO^{18} values of the leucogranites could reflect extensive groundwater interaction, although it is difficult to reconcile the amount of interaction required with the paucity of hydrous minerals present and the apparent lack of alteration. Selective groundwater activity with the leucogranites in preference to other units of the Batholith also pose problems, particularly as many of the leucogranites are among the youngest members of the Batholith. The Ruby Creek pluton, and perhaps the Mole pluton, may have had an origin related to an I-type protolith in which the magma has undergone some degree of amphibole fractionation so as to produce a corundum-normative muscovite-biotite leucogranite as suggested by Cawthorn and Brown (1976), Cawthorn and O'Hara (1976), and Helz (1975).

The general depletion of deuterium in the New England Batholith, the unusually high values of $\Delta(Q-\text{Bi})$ presumably due to a lowering of δQ^{18} values of biotite, and the more scattered relation between δD and water content relative to the Berridale Batholith point to ground water interactions with the cooling plutons as a major factor in the development of the batholith. A relation between Δ (Q-Bi) and δ D might be expected if these values were controlled dominantly by the intensity of ground water interaction, but no clear relation was observed. The leucogranites are relatively depleted in both deuterium and O^{18} , and this could be related to extensive ground water interactions rather than to the original isotopic ratios of the parent materials.

The difference between the overall deuterium content of the upper Paleozoic New England and Silurian Berridale Batholiths is striking, and on the basis of the argument concerning meteoric water interactions is probably related to the near polar paleolatitude in the Carboniferous and the paleolatitude of 22° S in the Silurian that have been inferred for these regions (Embleton et al., 1974).

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