Oxygen and hydrogen isotope disequilibria in the Landsverk I pegmatite, Evje, southern Norway: Evidence for anomalous hydrothermal fluids

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Oxygen isotope analysis of certain quartz, amazonite feldspar, and epidote from the Landsverk I pegmatite near Evje, southern Norway, indicates extreme local ^{18}O depletion. The $\delta^{18}O$ values of massive, milky white breccia-filling quartz are as light as -7.4 ‰. Marked oxygen isotope exchange was found for amazonite, but not for fragments of microcline perthite in the breccia. ^{18}O depletion of this magnitude is characteristic of meteoric-hydrothermally altered rocks, and the isotopic composition of the appropriate meteoric water is estimated to have been $\delta^{18}O = 16.0$‰ and $\delta D = -125$‰. However, the δD values of water in only one fluid inclusion sample provide supportive evidence for the presence of meteoric water, and no large-scale hydrothermal alteration systems are as yet known for the area. The oxygen and hydrogen isotope compositions of Precambrian (?) meteoric water may not correspond to the Epstein-Craig equation for present-day waters. ^{18}O depletion by CO_2 exsolution from a late magmatic fluid provides an alternative explanation.

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The granitic pegmatite exposed in the Landsverk Quarry I, near Evje, S. Norway (Fig. 1) contains evidence of marked ^{18}O depletion and isotope disequilibrium of an unusual nature between minerals in pegmatite breccia formed subsequent to mainstage pegmatite crystallization. By comparison with stable isotope studies of other pegmatites (B. E. Taylor et al. 1979, B. E. Taylor 1982, B. E. Taylor & Friedrichsen 1983), interaction between a cooling pegmatite and meteoric water does not appear to be a common phenomenon, in contrast to alteration observed in some other geologic environments (e.g. H. P. Taylor 1977). The isotopic disequilibrium between the most ^{18}O-depleted quartz yet reported, and coexisting K-feldspars demonstrated in this study, sheds some light on the nature of amazonite feldspars and retrograde isotopic exchange of microcline, and raises questions regarding the involvement and isotopic composition of ancient (pre-Permian, possibly Precambrian) meteoric water. An alternative mechanism to explain the marked ^{18}O depletion of quartz is also considered.

B. E. Taylor & Friedrichsen (1983) determined the oxygen, hydrogen, and carbon isotope compositions of minerals and fluid inclusions from a number of granitic pegmatites from the Evje-Iveland area. Bjørlykke (1935) suggested a magmatic origin on the basis of contact relationships, while Barth (1947) preferred a metasomatic origin for the pegmatites. The oxygen isotope fractionations between primary, magmatic stage minerals, and the lack of isotopic equilibrium with the immediate wall rocks (B. E. Taylor & Friedrichsen 1983) lend strong support for a magmatic origin, i.e., crystallization of hydrous silicate melts over a range of temperature.

Neumann (1960) concluded from K/Ar, Pb^{207}/Pb^{206}, and Re/Os ages that Iveland pegmatites were formed 880–920 Ma b.p. These data compare well with the Rb/Sr ages of 900 ± 53 Ma for granitic and monzonitic rocks, respectively, of the Høvringsvatn complex immediately to the north (Pedersen 1973, 1975, and 1981). A genetic relationship between the magmatism that produced the Høvringsvatn complex and the pegmatites is compatible with both age relationships (Pedersen 1981) and oxygen isotope data (B. E. Taylor & Friedrichsen 1983).

Aspects of the geology, mining history and mineralogy of the Landsverk pegmatite are described by Andersen (1926) and by Bjørlykke (1935). Of importance to our discussion is the
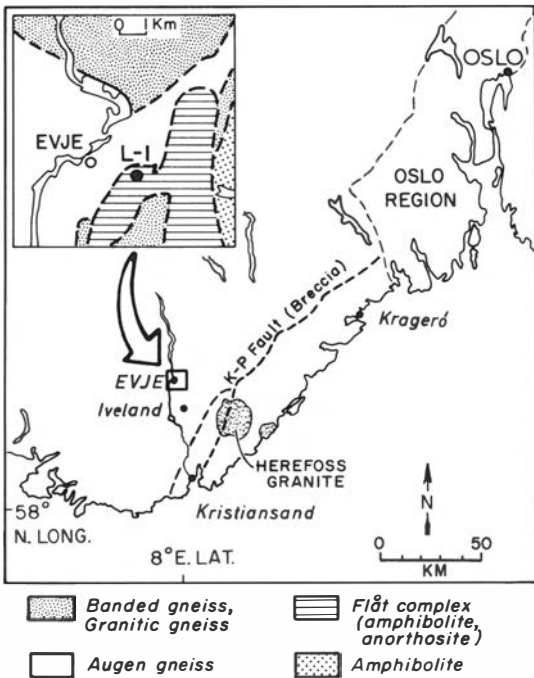
Fig. 1. The location of the Landsverk I pegmatite quarry (L-1) is shown in the inset map, near Evje, South Norway. The geology of the inset map is modified after Pedersen (1981); the geologic legend applies to the inset map. The locations of the Kristiansand – Porsgrunn (K-P) fault (breccia zone) and the Oslo Region, which contains lower Paleozoic sedimentary and Permian igneous rocks, are after Fig. 1 of S. R. Taylor et al. (1960).

occurrence within the pegmatite of a breccia zone containing quartz-cemented pegmatite breccia. Inclusions of the gabbroic host rock are also reported from the brecciated areas (Bjørlykke 1935). S. R. Taylor et al. (1960) measured the trace abundances of Li, Na, K, Rb, Cs, Pb, Tl, Ca, Sr, and Ba in a number of feldspar samples from the Landsverk I quarry. They distinguished three generations of microcline, (1) common pink microline (with albite); (2) amazonite (plus cleavelandite); and (3) a brick-red microcline found as overgrowths on other feldspars and on fractured surfaces in brecciated pegmatite. From a study of the distributions of Fe, Cr, Mn, Cu, Co, Ni, V, Sn, and F in a sample of microcline which graded into amazonite, S. R. Taylor et al. concluded that the amazonite formed by the metasomatic replacement of generation (1) microcline by late-stage 'pegmatitic rest liquid'. On the basis of contrasting trace element compositions, especially Ba, the brick-red microcline (generation 3) was inter-

preted to be of secondary origin, unrelated to the primary crystallization of the generation 1 and 2 pegmatite feldspar.

In this paper we compare oxygen, hydrogen, and carbon isotope composition of minerals and fluid inclusions in normal (generation (1) and (2)) and brecciated (generation (3)) pegmatite from the Landsverk I quarry. The variations in isotopic compositions, and some trace element patterns of S. R. Taylor et al. (1960), will be seen to be due in part to chemical exchange with an ^{18}O-depleted hydrothermal fluid during growth of the generation (3) stage feldspar.

Since the isotopic anomalies described in this paper were measured in pegmatite breccia, the age of the brecciation is critical to our discussion. Unfortunately, this age cannot be precisely determined, although details of the oxygen isotope data discussed below would be compatible with a close relationship between the very latest hydrothermal stages of pegmatite crystallization and the deformation. Although several younger radiometric ages (K/Ar, Pb^{207}/Pb^{206}, Re/OS) are recorded by Neumann (1960), major thermal events younger than 900 Ma have not been recognized in the Evje-Iveland area (Pedersen 1980).

On a regional scale, the breccia zone shown in Fig. 1 (after S. R. Taylor et al. 1960) cuts the post-kinematic Herefoss granite dated at 961 + 60 Ma (Rb/Sr, Brueckner 1972). A slightly lower K/Ar age of 860 Ma for biotite from the granite was reported by O'Nions et al. (1969). Fissure-filling copper mineralization has been described in the Telemark-Setesdal region as post-kinematic (900–950 Ma) by Nordrum (in Bugge 1978), but the relationship of this hydrothermal activity and fracture-producing deformation to that at the Landsverk I quarry is unknown. Brittle deformation like that observed in the Landsverk pegmatite has not been described in other pegmatites in the Evje-Landsverk area. The absolute age must remain equivocal, but brecciation could have occurred during cooling and uplift of the Evje-Iveland area in the Precambrian.

Analytical methods

Oxygen and hydrogen were extracted from pegmatite minerals using standard techniques as described in B. E. Taylor et al. (1979) and B. E. Taylor & Friedrichsen (1983), and analyzed as CO_2 and H_2, respectively, by mass spectrometry

to determine the isotopic composition relative to V-SMOW. The carbon isotope composition of fluid inclusion CO_2 is reported relative to PDB. Results are reported in the usual δ notation.

Samples analyzed for their oxygen isotope content in this study constituted less than 0.1 cm^3, and were either hand-picked or cut from traverses in one hand sample, as a series of subsamples within ≤ 1 cm of each other to elucidate the detailed relationships discussed below. Notes regarding the preparation and extraction of fluids from fluid inclusions may be found in B. E. Taylor et al. (1979). Because the extraction process requires the *in vacuo* crushing of sample volumes of several cm^3, selective extraction of fluids from different generations of inclusions is generally not possible.

Results and discussion

Oxygen and hydrogen isotope compositions of pegmatite minerals from the Landsverk I pegmatite are reported in Table 1, and descriptions of the samples are given in the appendix. The $\delta^{18}O$ values in Table 1 include those of magmatic stage (generation (1)) minerals with primary isotopic compositions (compare with the ranges of $\delta^{18}O$ values shown in Table 2 for other Evje-Iveland pegmatites), and isotopic data for some minerals which indicate oxygen isotope exchange with heated ^{18}O-depleted fluids. Hydrogen isotope data for biotite and muscovite from other Evje-Iveland pegmatites and fluid inclusion water are also summarized from B. E. Taylor & Friedrichsen (1983) in Table 2. The data in Table 2 will serve as the isotopic characteristics of unaltered pegmatites to which the unusual isotopic compositions reported here for certain Landsverk I samples may be compared.

Very late-stage minerals in the pegmatite (generation (2) of S. R. Taylor et al. 1960) usually include cleavelandite, quartz, and stilpnomelane. Oxygen isotope fractionations between quartz and these minerals in samples NP-2 and NP-7 indicate lower temperatures (based on fractionation factors of various authors cited in B. E. Taylor & Friedrichsen 1983) than expected for most of the magmatic-stage pegmatite crystallization history, i.e., less than about 550°C. Crystallization from and extensive replacement of earlier portions of some pegmatites by an exsolved aqueous fluid are often characteristic of this 'hydrothermal-stage' of pegmatite crystallization

(e.g. Jahns 1955). Mutual oxygen isotope equilibrium between these later and sometimes sequentially crystallized mineral phases is rarely achieved, despite possible isotopic equilibrium between the aqueous fluid and newly crystallized minerals (B. E. Taylor et al. 1979). Thus, oxygen isotope fractionations between very late mineral phases, especially those formed in pegmatite pockets, or voids, are generally of little value for geothermometry. Nevertheless, preserved isotopic zonation in some minerals does provide useful information regarding late-stage crystallization processes, and can under certain circumstances provide information on temperature variations.

Oxygen isotope zonation parallel to the c-axis in the doubly terminated quartz crystals of sample NP-7 (Table 1) was found to vary from $+ 5.0‰$ in the core, to $+ 11.8‰$ at the outermost tip. This 6.8‰ variation in $\delta^{18}O$ would imply a range of crystallization temperature of approximately 300°C, based on the quartz-water fractionation expression of Matsuhisa et al. (1979). With the assumption that the quartz crystal grew from a fluid of constant oxygen isotope composition, this could have corresponded to crystallization from near a solidus temperture of 550°C down to ca 250°C.

Isotopic zonation studies of large individual crystals are few. B. E. Taylor et al. (1979) reported base-to-tip enrichments in ^{18}O of up to 2.1‰ in pegmatite quartz crystals from pockets. In all instances, the oxygen isotope composition of the first-formed (basal) part of the quartz crystal was the same or very similar to quartz in nearby pegmatite. The $\delta^{18}O$ of the quartz core of the crystal of NP-7 described above is lighter than any primary quartz, excluding the breccia discussed below. This observation suggests subsolidus growth from an ^{18}O-depleted fluid, whereas most of the massive quartz referred to as 'primary' crystallized at higher temperatures from a melt. Continued, closed-system oxygen isotope exchange between entrapped aqueous fluid and pegmatite minerals during cooling could deplete the fluid in ^{18}O due to isotopic fractionation. In this case, the estimated 300°C temperature range for the hydrothermal stage would be a minimum. However, the marked increase in $\delta^{18}O$ along the c-axis nearer the tip of the crystal may also reflect an increase in the ^{18}O content of the fluid, such as would occur to the liquid phase upon steam separation during boiling.

Table 1. Oxygen and hydrogen isotope analyses of pegmatite minerals from the Landsverk pegmatite quarry and other occurrences, Evje-Iveland area, Southern Norway

| Sample No. | Description* | $\delta^{18}O[\delta D]$ | | | | | |
		Qtz.	Ksp.	Amaz.	Ab.	Mu.	Other
Landsverk Pegmatite Quarry							
L-3-1	Quartz	+ 8.8					
L-3-2	Quartz	+ 9.2					
L-1-A	Quartz	− 7.06					
FPN-6	Epidote with feldspar		+6.70		+6.33		− .34
NP-2	Quartz and stilpnomelane	+14.66 (Qtz-1)					+ 9.84
1	Pegmatite				+6.95		
NP-6	Pegmatite breccia	− 1.21	+9.3		+7.12(Ab-1)		
					+7.70(Ab-2)		
NP-7	Quartz crystal; feldspar	+10.12			+8.00		
		+10.99			+7.70		
		+ 4.96					
		+ 5.48					
		+ 6.66					
		+11.81					
NP-13	Amazonite			+3.61			
AM-1-1	Amazonite			+2.26(−1)			
AM-1-2				+1.87(−2)			
AM-2-1	Amazonite			+5.90			
AM-2-2				+5.32			
AM-2-3				+4.54			
				+6.59			
AM-3-1	Amazonite			+6.29			
AM-3-2				+7.34			
AM-3-3				+7.51			
AM-3-4				+7.13			
AM-3-5				+7.35			
AM-3-6				+8.06			
LB-1	Pegmatite breccia		+7.10				
LB-2			+7.00				
LB-3			+7.42				
LB-4			+7.30				
LB-5			+7.00				
LB-6		−7.43					
LB-7		−6.20					
LB-8		−6.24					
LB-9		−4.40					
LB-10		−5.03					
LB-10		−5.03					
LB-11		−0.72					
LB-12		−3.93					
LB-2-1	Pegmatite Breccia	−6.36					
LB-2-2			+7.50				
LB-2-3		−3.10					
F-La	Magnetite + Quartz	+ 8.9					
F–Lb		+ 8.9					
F-Lc		+ 1.6					
F-Ld		+ 1.6					
F-Le		+ 1.6					
F-Lf		+ 9.0					
F-Lg		+ 8.9					
Other Pegmatite Quarries							
NP-10	Cleavelandite, quartz stilpnomelane	+ 8.42			+6.18		+10.03 [−108]
NP-3	pegmatite	+ 8.30			+6.75	+5.65	

* See appendix for more detailed descriptions

Pegmatite breccia. – Oxygen isotope data collected on quartz and feldspar from quartz-cemented pegmatite breccia (Table 1) show the largest variations and greatest ^{18}O depletions for any quartz yet reported. These unusual compositions record oxygen isotope exchange with, and crystallization from, very ^{18}O-depleted fluids. The samples of breccia analyzed included that pictured in Figure 2a. Fragments of large microcline perthite crystals up to several centimeters across are enclosed within a matrix consisting of intergrown, euhedral milky white quartz crystals and massive white quartz.

The LB-series of subsamples reported in Table 1 is a particularly good example of the extent of ^{18}O-depletion in quartz from the breccia matrix adjacent to isotopically 'normal' perthite ($\delta^{18}O$ = +7.00 to +7.42). The $\delta^{18}O$ values of quartz (LB-6 – 12) as negative as −7.43‰ in sample LB- (Table 1) contrast markedly with other pegmatite quartz which typically has positive $\delta^{18}O$ values similar to those in Table 2.

Not all massive quartz in the pegmatite crystallized from the ^{18}O-depleted fluid. Gray, vitreous quartz (e.g. L#3(1), L#3(2), Table 1), which typically comprises a quartz core to many pegmatite bodies, has a normal (i.e. primary) oxygen isotope composition. The depleted oxygen isotope compositions associated with quartz-cemented brecciation have not been observed among other pegmatites of the district, and may denote an event of specific significance to the Landsverk I pegmatite.

Subsolidus oxygen isotope exchange by diffusion between quartz and a grain-boundary fluid cannot account for the marked ^{18}O depletion described above. This is illustrated by the constant $\delta^{18}O$ values measured for quartz adjacent to a vein of magnetite in sample F–L (Table 1, Appendix). Subsamples of quartz adjacent to, and within several millimeters of, the magnetite ($\delta^{18}O$ = +1.6) do not vary significantly from $\delta^{18}O$ = +8.9, an isotopic composition typical of other massive pegmatite quartz (see Table 2). Resistance to subsolidus oxygen isotope exchange is, of course, not the same for all minerals.

A relatively rapid rate of oxygen isotope exchange between water and feldspar has been observed in experiments of several workers (O'Neil & Taylor 1967, Yund & Anderson 1978, Matsuhisa et al. 1979, Matthews et al. 1983). The preservation of primary oxygen isotope compositions in feldspar fragments within several milli-

Table 2. Oxygen, hydrogen, and carbon isotope compositions of mineral and fluid inclusions from undisturbed Evje-Iveland pegmatites[1]

Sample type	$\delta^{18}O$	δD	$\delta^{13}C$
quartz	+8.3 to +10.1		
microcline	+6.7 to + 9.9		
albite[2]	+6.5 to + 8.5		
muscovite	+5.7 to + 6.9	−37 to −59	
biotite	+2.2 to + 5.6	−53 to −79	
magnetite	+0.5 to + 2.0		
fluid inclusion (H_2O)[2]		−32 to −67	
fluid inclusion (CO_2)			−2.2 to −6.1

[1] based on data in B. E. Taylor and Friedrichsen (1983).
[2] isotopic compositions reported only for waters released by second crushing

meters of the ^{18}O-depleted quartz matrix suggests that some quartz had perhaps already been deposited at the time of influx of the ^{18}O-depleted fluid, shielding the perthitic feldspar fragments from contact and isotope exchange with the ^{18}O-depleted fluids.

An epidote rosette from a fracture which cross cuts a crystal of microline perthite (sample FPN-6, Table 1) has an oxygen isotope composition of −0.34‰ which indicates that it, too, crystallized from an ^{18}O-depleted fluid. The $\delta^{18}O$ values of associated feldspar are 2‰ depleted in ^{18}O relative to normal feldspar in the pegmatite body due to partial exchange with the fracture-filling fluid. Thus, the interaction of the hydrothermal fluid with the pegmatite involved not only the crystallization of quartz, but also the replacement of feldspar by epidote.

The local nature of the ^{18}O depletions is further emphasized by the fact that not all very late stage (hydrothermal) minerals show isotopic depletion. Stilpnomelane (e.g. NP-2, Table 1; see also NP-10) found growing on the tips of euhedral, vug-filling, milky quartz crystals, exhibits ^{18}O-rich isotopic compositions (+9.84, +10.03, Table 1) similar to hydrothermal alteration phases found in other pegmatites (e.g. B. E. Taylor et al. 1979) where no evidence for ^{18}O-depleted fluids exists.

Amazonite. – Amazonite is thought to form in pegmatites during the hydrothermal stage (e.g. Andersen 1926, S. R. Taylor et al. 1960) when Na-metasomatism of earlier formed minerals may result in large-scale replacement of pegmatite.
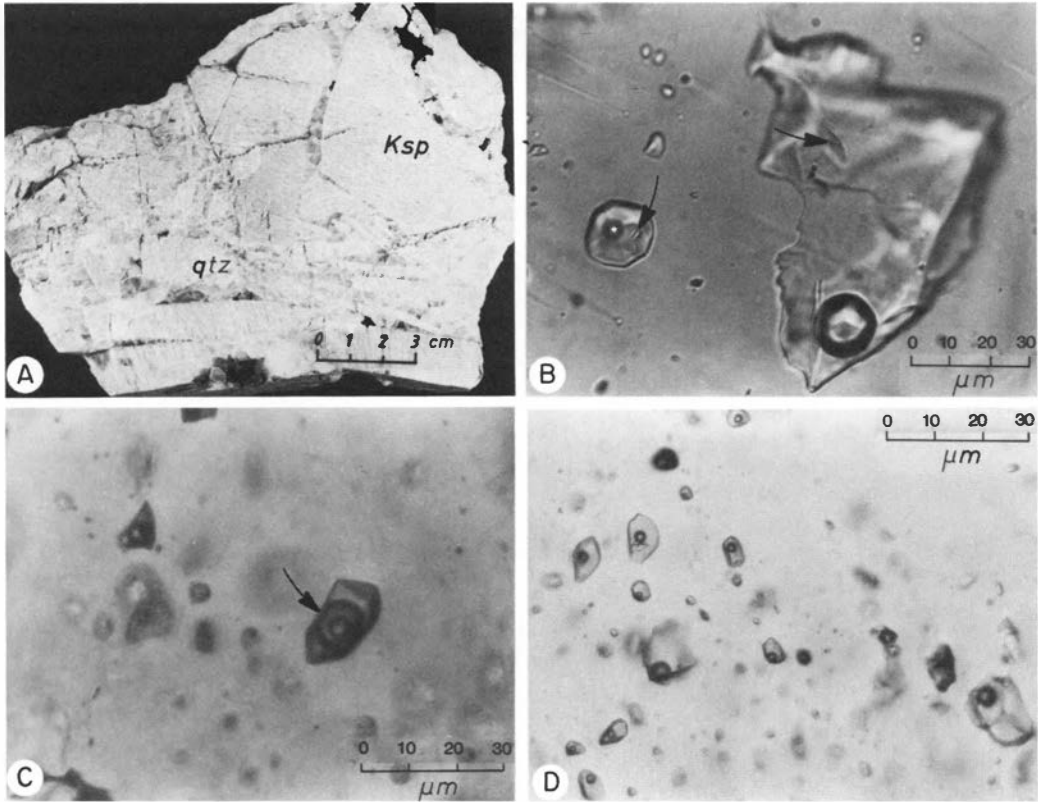
Fig 2a-d.
(a) Photograph of quartz-cemented quartz-feldspar breccia from the Landsverk quarry. Ksp = microline perthite, qtz = quartz. Bar scale is in centimeters.
(b) Photomicrograph of large and small primary two-phase fluid inclusions in isotopically normal, non-brecciated quartz (sample Brua #2). Note elongate solid inclusions shown by arrows.
(c) Photomicrograph of primary three-phase ($H_2O + CO_2$) inclusion (see arrow) exhibiting negative crystal outline in non-brecciated pegmatite quartz (sample Brua #2). Variable vapor/liquid ratios may be seen in other, slightly out of focus inclusions.
(d) Photomicrograph of two phase fluid inclusions in quartz breccia pegmatite sample L#1A. Inclusions in similar breccia samples are typically more numerous and have lower vapor/liquid ratios compared to fluid inclusions in normal pegmatite samples. Fluid inclusions in breccia samples contain no liquid CO_2 and solid inclusions only rarely.

The micro-texture of amazonite is suggested by Tibballs & Olsen (1977) to have been acquired during very slow cooling. The fact that some crystals of amazonite (e.g. Am-3, Table 1) exhibit normal oxygen isotope compositions for potassium feldspar found elsewhere in the Landsverk and other pegmatites indicates that the late-stage hydrothermal fluid responsible for amazonite growth was derived from the pegmatite itself, and probably represents at least part of the water exsolved from the hydrous pegmatite melt. Similar isotopic relationships have been observed for gem-quality feldspars that grew in pockets of granitic pegmatites in southern California (B. E. Taylor et al. 1979).

Comparison of oxygen isotope depletions in pink potassium feldspar with those found in amazonite fragments in breccias suggests that amazonite may have exchanged oxygen isotopes more readily with the solutions present during brecciation than did the pink microcline. Samples NP-13 and AM–1, –2, and –3 have δ18O values as low as +1.87‰. In addition, we note a qualitative correlation between a darker blue-green color and more extensive ^{18}O depletion in the amazonites analyzed. Is the reason for this apparently more extensive oxygen isotope exchange related to the origin of color in amazonite?

A close spacing (< 3nm) of Al-Si defects in

two amazonite specimens was interpreted by Tibballs & Olsen (1977) from structural state X-ray studies. They also observed very fine twinning in amazonite with the transmission electron microscope. For the two specimens compared, the amazonite with the greater Al/Si framework defect density discolored more rapidly upon heating to 520°C, suggesting the presence of a submicroscopic texture on a scale even finer than 3 nm. The specimen which showed the more rapid bleaching was also characterized by a stronger bluegreen color. Thus, the presence of defect structures may have aided the diffusion of oxygen in the amazonite, and thereby have accelerated oxygen isotope exchange.

Fluid inclusions. Figures 2b–d show typical fluid inclusions in quartz from both undisturbed and brecciated pegmatite. The inclusions in both occurrences are typically 10–20 microns in their longest dimension, although rare inclusions as large as 100 microns (e.g. Fig. 2b) may be observed in normal pegmatite quartz. Although generally similar in size, the mode of occurrence of the inclusions in quartz differs in the brecciated and non-brecciated pegmatite. The inclusions in the normal, or non-brecciated quartz typically occur along discontinuous planar zones, presumably healed fractures. Inclusion shapes may be irregular, but a great many are subrounded or exhibit negative crystal shapes. The enclosed fluids are locally CO_2-bearing, as indicated by 3-phase inclusions with liquid CO_2 (e.g. Fig. 2c) which are abundant in some zones. Solid inclusions are occasionally observed, either as isotropic cubic forms (sodium chloride?) or as elongate anisotropic (muscovite?) crystals. The apparent vapor/liquid volume ratios in both 3-phase and sometimes 2-phase inclusions are locally variable, and may suggest either quartz growth from a heterogeneous (two-phase) fluid, or fracturing and local recrystallization of quartz during boiling of a once homogeneous, trapped C-O-H fluid. The possible consequences of the separation of a CO_2-rich vapor and an H_2O-rich liquid during boiling for the oxygen isotope composition of the H_2O and crystallizing quartz are discussed later.

Fluid inclusions in the milky white quartz breccia are abundant throughout the quartz and do not exhibit any preferential localization along either growth or fracture planes. Vapor/liquid ratios appear to be constant within the quartz, and no 3-phase, liquid CO_2-bearing inclusions have

been observed. Also no inclusions were found containing solid inclusions of any kind. Although no homogenization temperatures or freezing-point-depression data are presently available for these inclusions, the physical characteristics of the inclusions in the breccia and non-breccia samples are sufficiently distinct to indicate that different conditions of entrapment must have reigned during their formation. Samples such as LB (Table 3), which exhibit large inhomogeneities in ^{18}O content, are seen to contain fluid inclusions characteristic of both of the types described above.

The hydrogen and carbon isotope compositions of H_2O and CO_2 released from fluid inclusions in pegmatite quartz and feldspar are listed in Table 3. Each sample analyzed was subjected to partial crushing in an evacuated tube as described in B. E. Taylor et al. (1979) in an attempt to preferentially release secondary inclusions. As may be seen from the data in Table 3, each first crushing released water which was depleted in deuterium relative to the larger quantities of water released upon subsequent crushings. This variation in δD with subsequent crushing would be compatible with the preferential release of D-depleted waters from secondary fluid inclusions. However, the possibility that isotopic fractionation occurred during the extraction due to incomplete recovery of the water released during the first, partial crush, and to isotopic fractionation of small (< 30 μm) H_2 samples collected using activated charcoal has been discussed by B. E. Taylor & Friedrichsen (1983).

Pegmatite quartz samples in Table 3 which exhibit 'normal' oxygen isotope compositions contain fluid inclusions with waters having δD values in the range of −32 to −67‰. These data are similar to isotopic compositions observed for water from fluid inclusions in a number of unaltered pegmatites (B. E. Taylor et al. 1979, B. E. Taylor & Friedrichsen 1983) where δD values as negative as −90‰ may reflect isotopic fractionation during vapor loss from the hydrous melt. Several samples of ^{18}O-depleted quartz in Table 3 yielded waters from fluid inclusions with hydrogen isotope compositions similar to fluid inclusions analyzed from other, 'normal' pegmatites (Table 2). One exception is sample LB, for which a marked ^{18}O gradient is demonstrated; it contains fluid inclusion water with a hydrogen isotope composition of −110‰. Note that this sample was rather small (47 μm), and may have been fractionated.

Table 3. Hydrogen and carbon compositions of inclusion fluids in brecciated and non-brecciated pegmatite

Sample No.	Description[1]	N.C.[2]		μmH$_2$[3]	μmCO$_2$	X_{CO_2}	δD	$^{13}C_{CO_2}$
Landsverk Pegmatite Quarry								
FPN-6	small vein in	(1)[4]	<0.1	10	<0.1		− 63	
	perthite	(2)[5]	0.1	502	~0.1	0.001	− 50	
LB	breccia	(1)	<0.1	5	<0.1	<0.02	−125	
		(2)	<0.1	47	~0.1	0.002	−110	
L#1	breccia	(1)	<0.1	45			− 85	
		(2)	<0.1	270	~0.1	<0.001	− 63	
L#1	breccia	(1)	<0.1	3	0.5	0.007		
		(2)	<0.1	68			− 77	−4.35
L#1a	quartz	(1)	<0.1	40	n.d.		− 85	
		(2)	0.1	670	~0.1	<0.001	− 52	
L#3*	quartz	(1)	<0.1	2				
		(2)	<0.1	36	0.5	0.001	− 56	−7.42
NP-6	breccia	(1)	<0.1	41	n.d.		−104	
		(2)	<0.1	630	~0.2	<0.001	− 64	
Landsverk Milky Qtz.	quartz	(1)	<0.1	26	n.d.	<0.001	− 85	
		(2)	<0.1	96	n.d.	<0.001	− 66	
		(3)	<0.1	600	<0.11		− 50	
Landsverk Milky Qtz.	quartz		<0.1	365	<0.1	<0.001	− 57	

[1] See Appendix for more detailed descriptions.
[2] N. C. = non-condensible at liquid nitrogen temperature
[3] μmH$_2$ = H$_2$O + H$_2$
[4, 5] = extraction number in sequence when more than one extraction performed on same sample in same crushing tube
* = quartz-bearing samples in which the quartz exhibits "normal" oxygen isotope compositions

The carbon isotope composition of CO$_2$ extracted from the inclusion fluids in several samples is given in Table 3. Sample L#3, a 'normal' pegmatite quartz sample, exhibits a δ^{13}C value of −7.42‰, typical of CO$_2$ from other pegmatites (B. E. Taylor & Friedrichsen 1983). CO$_2$ from pegmatite breccia Sample L#1 (Table 3) has a δ^{13}C-value commonly observed for CO$_2$ in undisturbed pegmatites. Such a carbon isotope composition is not sufficiently different so as to warrant a unique origin. Slight contamination of essentially primary CO$_2$ with another source of carbon, such as metamorphic (decarbonation) CO$_2$, is permissible, but carbon isotope fractionation during loss of a CO$_2$-rich vapor which could also enrich the residual dissolved carbon in ^{13}C (see Fig. 27, Friedman & O'Neil 1977) is also possible.

Conclusions

The previously presented stable isotope data record an unusual and marked depletion of ^{18}O in quartz and amazonite feldspar. The most isotopically depleted quartz yet reported occurs in quartz-cemented pegmatite breccia and has a δ^{18}O value of −7.43‰. Isotopic compositions of minerals thus depleted in ^{18}O are usually characteristic of high temperature interaction with meteoric water. The evidence for such isotopic and chemical exchange processes is numerous and has been demonstrated for a number of geologic environments (e.g. H. P. Taylor 1977). For certain geographic situations, large negative δD values are indicative of interaction with even small amounts of meteoric water because complete hydrogen isotope exchange between hydrothermal fluids and rocks is usually attained well before equilibration of oxygen isotopes.

The oxygen isotope composition of the hydrothermal fluid responsible for the isotopic depletion in the pegmatite breccia can be estimated using the data in Table 1 and a few assumptions. The maximum temperature of the brecciation and quartz crystallization may be limited by the occurrence of amazonite. Oftedal (1958) presented the results of a simple experiment in which the

rate of color loss in amazonite was determined as a function of temperature. He determined that coloration is stable below about 250°C, whereas at higher temperatures, the blue-green color of amazonite fades within minutes to hours. These results are in accord with those of Tiballs & Olsen (1977).

Assuming a temperature of 250°C and using the quartz-water oxygen isotope equilibrium equation of Matsuhisa et al (1979), we calculate a $\delta^{18}O$ value for the water in equilibrium with sample LB-6 to be −16.0‰. Such an ^{18}O-depleted fluid could be most readily explained as meteoric water. From the Epstein-Craig equation for meteoric waters, we estimate the hydrogen isotope composition to have been −118‰. Both the oxygen and hydrogen isotope compositions must be minimum estimates if isotopic exchange between country rocks and meteoric water occurred prior to interaction with the pegmatite. The lack of close agreement between the δD value of −118‰ and the hydrogen isotope compositions of the fluid inclusions water (Table 3) requires some comment.

Of the fluid inclusion samples analyzed in Table 3, only one (LB) contained water with a hydrogen isotope compositions ($\delta D = -110‰$) close to that expected for a breccia fluid of meteoric origin. A mixture of fluid inclusion types was noted in some breccia samples, and therefore isotopic analyses might also indicate a mixture of isotopically distinct fluids. However, breccia samples in which quartz exhibits distinctly light oxygen isotope compositions (e.g. L#1A; Np-6) typically yielded inclusion water with δD values in the 'magmatic' range, i.e. close to −50‰, permitting admixture of only a few percent of a low deuterium water. The similarity of results for nearly all samples from Landsverk quarry and for samples from younger pegmatites (B. E. Taylor et al. 1979) precludes any alteration of the original isotopic composition of the inclusion fluids, such as by diffusional loss of hydrogen, or *in situ* formation of CH_4.

If the oxygen isotope composition of the ocean during the Precambrian (the assumed age of brecciation) were significantly different from the present-day (cf. Perry et al. 1978), the hydrogen isotope composition of the meteoric water could not be estimated with the Epstein-Craig equation. Sea-floor hydrothermal convection and isotopic exchange provide strong evidence against major changes in the oxygen isotopic composition of ocean water (Gregory & Taylor 1981). The

oxygen and hydrogen isotope data are difficult to reconcile in terms of known meteoric water compositions. Further, there are at present no other data indicating any major meteoric hydrothermal alteration in the Landsverk area. The possibility suggested by these data that the isotopic composition of early meteoric waters does not correspond to present Epstein-Craig equation is, nevertheless, intriguing.

One alternative to the meteoric water hypothesis that has potential based on observation of heterogeneous C-O-H fluids in primary fluid inclusions is the possible oxygen isotope depletion of magmatic water upon exsolution of CO_2 as the H_2O-CO_2 solvus is passed during cooling. The temperature at which H_2O and CO_2 separate depends on the NaCl content of the fluid, increasing with NaCl, and may even reach magmatic temperatures (Bowers & Helgeson, in press). By this mechanism, water with a $\delta^{18}O$ values as light as −16.0‰ could only reasonably be produced by preferential loss of CO_2 which concentrates ^{18}O at elevated temperatures (Truesdell 1974), leaving water locally depleted in ^{18}O. It was noted previously that fluid inclusions in the breccia samples were water rich, and contained no signs of either abundant CO_2 or salt (NaCl) inclusions in the samples examined microscopically. The 'salt effect' on oxygen isotope compositions of fluids and solids (Truesdell 1974) can be ignored.

To recommend ^{18}O-depletion by a process of CO_2-boiling is based on the lack of evidence elsewhere in the rocks for any large-scale meteoric hydrothermal systems, the lack of strong evidence in the hydrogen isotope compositions of fluid inclusions of the presence of meteoric water, and petrologic continuity of the development of significant internal pressures in a pegmatite with the exsolution of a fluid phase during crystallization in a closed system (cf. Burnham 1979). Internal deformation associated with the latter process has been observed elsewhere in pegmatites (e.g. Jahns 1982), although not with similar isotopic effects (B. E. Taylor et al. 1979). Unfortunately, both the uncertainties involved in estimating the isotopic effect of CO_2-H_2O unmixing and the difficulty of proving such a process greatly hinder its testing in this case.

Thus, the isotopic data suggest, on the one hand, the influx into a brecciated pegmatite of hot (∼250°C) meteoric water, perhaps isotopically anomalous relative to present-day meteoric waters. The distinct trace element compositions

of a brick-red potassium feldspar analyzed by S. R. Taylor et al. (1960) in the Landsverk I pegmatite were interpreted by them as the result of late-stage processes unrelated to magmatic differentiation. These 'late-stage processes' included crystallization in a chemically open system, permitting communication with adjacent wall rocks.

On the other hand, not all of the isotopic data are readily explained by the presence of meteoric water with oxygen- and hydrogen isotope relationships as we presently know them. An alternative explanation, involving the inevitable separation and preferential loss of a CO_2-rich vapor from a magmatic C-O-H fluid may be considered as a mechanism by which to deplete the residual H_2O-rich liquid phase (and crystallizing quartz) in ^{18}O. Such a scenario might also be accompanied, or even initiated by local brecciation.

Note added in proof. Harmon & Schwarcz (1981; Nature, 290, pp. 125–128) noted that δD–$\delta^{18}O$ relationships for paleowaters from fluid inclusions in speleothems did not correspond with isotopic relationships for present day meteoric waters due to small shifts in $\delta^{18}O$ of ocean water during glaciation. The glacial effect changed the δD intercept of the Epstein-Craig equation by $-10‰$. The isotopic shift inferred from our data would be opposite in sign, but nearly of the same magnitude.

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Appendix: Descriptions of samples analyzed in Tables 1 and 3

FPN-1
: Epidote rosette in 2 mm-thick veinlet cross-cutting large (>3 cm) microcline perthite crystal.

NP-2
: Densely intergrown white quartz crystals; clear quartz occurs as tips on white quartz; stilpnomelane euhedra grow in interstices and on clear quartz.

NP-6
: Quartz-feldspar pegmatite breccia, cemented by quartz. Albite occurs as 7 × 7 cm fragment in quartz matrix (Ab-1) and as 1 × 1 mm euhedral crystals in small yug (Ab-2). K-spar is typical pink microcline perthite. Fluorite is present in matrix interstices.

NP-7
: Doubly-terminated quartz crystal (6.5 cm long) with pink microcline, albite and stilpnomelane (which occurs as overgrowth on quartz).

NP-13
: Amazonite; medium blue color.

AM-1-1
AM-1-2
: Amazonite, dark-medium blue color; subsamples −1 and −2. Taken 1 cm apart from slice through amazonite crystal. Am-1-2 located within 3 mm of outer edge of crystal.

AM-2-1
through
AM-2-4
: Amazonite, medium blue-green color; subsamples taken from slice through crystal, within 1 cm of each other. Fluorite crystals coat outer surface of amazonite. Am-2-1 is located closest to outer edge of crystal.

AM-3-1
through
AM-3-6
: Amazonite, medium to light blue color. Subsamples cut from slice through crystal, AM-3-1 located closest to outer edge of amazonite.

LB-1
through
LB-12
: Quartz-microcline pegmatite breccia, cemented by densely intergrown milky white quartz crystals. Subsamples collected at approximately 1 cm intervals along traverse through fragment of K-spar and adjacent quartz of matrix (see Fig. 2a).

LB-2-1
through
LB-2-3
: Quartz-microcline pegmatite breccia similar to LB-1 (above).

F-La
through
F-Lg
: 0.75 to 1.0 cm thick magnetite vein cross cutting densely, but irregularly fractured gray quartz. Quartz is typical of primary (generation −1) quartz.

1
: Coarse quartz-cleavelandite pegmatite. Quartz occurs as dark gray, isolated grains (to 1 cm diameter) in matrix of blueish cleavelandite (Fsp-1) and white cleavelandite (Fsp-2).

L#3(1)
: Massive, irregularly and moderately densely fractured gray quartz.

L#3(2)
: Massive dark gray quartz; similar to L#3(1).

L#1(A)
: Densely intergrown subhedral to euhedral milky white quartz crystals. Similar to quartz found as breccia matrix.

NP-10
: Quartz-cleavelandite pegmatite with late stilpnomelane growing on quartz surfaces adjacent to small vug.

NP-3
: Quartz-cleavelandite-muscovite-spessartite pegmatite; Frikstad.