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# Composition of hydrous melts in equilibrium with quartz eclogites

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With 1 Figure

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#### Summary

Compositions of the hydrous melts in equilibrium with garnet, omphacitic clinopyroxene and quartz have been investigated experimentally at 28.5 and 35 kbar. They are represented by silica-rich liquids (> 70% SiO<sub>2</sub>) with low MgO, FeO and CaO contents. The removal of ca 10–15% of the magma of this composition may be sufficient to convert quartz eclogite formed after subduction of altered MORB into a quartz-free bimineralic eclogite assemblage, which is a common type of xenoliths in kimberlites.

At 28.5 kbar the solidus temperature is between 700 and 750  $^{\circ}$ C in the system quartz eclogite—water, and the high pressure amphibole-out boundary lies at ca 25 kbar in accord with the previous studies.

#### Zusammenfassung

#### Die Zusammensetzung wasserhältiger Schmelzen im Gleichgewicht mit Quarz-Eklogiten

Um Prozesse zu simulieren, die bei der Subduktion von Ozeanbodenbasalten durch partielle Anatexis im Stabilitätsfeld von Eklogiten ablaufen, wurde die Zusammensetzung wasserhältiger Schmelzen in Gleichgewicht mit Granat, Omphacit und Quarz bei 28.5 und 35 Kbar experimentell untersucht. Diese Schmelzen sind reich an SiO<sub>2</sub> (>70 Gew %) und arm an MgO, FeO and CaO. Die Extraktion von ca. 10–15% derartiger Schmelzen würde genügen, um quarzführende Eklogite, die durch die Subduktion von alteriertem MORB Material entstanden sind, in quarzfreie bimineralische Eklogite umzuwandeln wie sie häufig als Xenolithe in Kimberliten beobachtet werden.

Im System Quarz-Eklogit—Wasser liegt die Solidustemperatur bei 28.5 Kbar zwischen 700 und 750 °C. Die obere Stabilitätsgrenze von Amphibol liegt in diesem Temperaturbereich bei ca. 25 Kbar.

#### Introduction

In recent years considerable efforts have been devoted towards identifying magmas which may be produced by partial melting of subducted oceanic crust at the depths where the stable mineral assemblage for this composition is represented by eclogites. These processes have been invoked to explain the origin of acid and intermediate magmatic rocks both in Archean cratons and in modern island arcs (e.g. Ireland et al., 1994; *Peacock* et al., 1994). High pressure equivalents of tholeiitic basalts are represented by quartz eclogites or coesite eclogites (Green and Ringwood, 1968). For example, all high-Ti eclogites produced by the high-pressure metamorphism of MORB-like basalts from the Eastern Alps contain quartz (Miller et al., 1988). Average MORB recalculated into eclogite norm contains about 8% of quartz. However, eclogites from the mantle, such as xenoliths in kimberlites, do not contain quartz or coesite, being predominantly represented by so-called bimineralic eclogites (garnet, clinopyroxene and often accessory rutile). This has been explained by the loss of silica minerals during certain episodes of partial melting in the eclogite stability field (Ringwood, 1990). Judging from the widespread occurrence of bimineralic eclogites among mantle xenoliths, high pressure partial melting of subducted basaltic material must be an universal process not necessarily confined to specific geodynamic situations such as hot slab subduction.

The understanding of these processes requires the knowledge of melting relations in quartz eclogites. In spite of the growing number of high pressure experimental work on basaltic materials (*Green* and *Ringwood*, 1968; *Stern* and *Wyllie*, 1978; *Yaxley* and *Green*, 1994; *Yasuda* et al., 1994; *Rapp* et al., 1991; *Rapp* and *Watson*, 1995), the compositions of melts in equilibrium with quartz eclogites or coesite eclogites have not been investigated in detail, because the majority of experiments in the subliquidus region were performed at such high degrees of partial melting that silica minerals were completely resorbed. In the petrological literature it is usually assumed that melts produced during the partial melting of an oceanic slab are represented by intermediate to mildly acid compositions similar to andesites and dacites (so called adakites, e.g., *Peacock* et al., 1994), although *Stern* and *Wyllie*'s data (1978) suggest that hydrous melting at 30 kbar may produce melts with SiO<sub>2</sub> contents in excess of 70%. Recent experimental results of *Yaxley* and *Green* (1994), however, indicate less siliceous liquids under similar conditions.

The aim of this work is to resolve these inconsistencies. We investigated hydrous silicate liquids in equilibrium with the phase assemblage quartz (or coesite) + omphacite + garnet by adding excess silica to the composition of altered basaltic crust. We have performed our experiments at pressures just above the upper stability limit of amphibole in order to simulate situations when water released by amphibole decomposition causes the partial melting of anhydrous mineral assemblages.

### Starting materials

The composition chosen for high pressure experiments in the present work is based on the average composition of altered MORB (composition GA1 in Table 1) as estimated by *Yaxley* and *Green* (1994). We added 30% of a mixture of  $Qz_{70}Ab_8Or_{22}$ (weight %) to this composition in order to have larger proportions of the liquid

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	SiO <sub>2</sub>	TiO <sub>2</sub>	Al <sub>2</sub> O <sub>3</sub>	FeO	MgO	CaO	Na <sub>2</sub> O	K <sub>2</sub> O
GA1	50.35	1.49	16.53	9.83	7.94	9.60	3.49	0.44
E3	62.29	1.04	13.29	6.90	5.58	6.74	2.74	1.42
E4	67.41	1.39	15.71	4.37	2.36	5.85	2.37	2.54
E6	64.47	1.29	15.73	3.87	3.11	4.10	5.59	1.84
E7	74.44	0.93	11.32	2.78	2.24	2.95	4.02	1.32

 Table 1. Composition of starting silicate mixtures

phase in equilibrium with the quartz-bearing phase assemblage. The composition of this starting mixture (E3) is given in Table 1. It was prepared from reagent grade  $SiO_2$ ,  $TiO_2$ ,  $Al_2O_3$ , MgO,  $CaCO_3$ ,  $Na_2CO_3$ ,  $K_2CO_3$ ,  $Fe_2O_3$  and metallic iron. All ingredients except metallic Fe were mixed by grinding in an agate mortar under alcohol. The glass was then prepared by heating this mixture in air in a Pt crucible at 1100 °C. The necessary amount of metallic iron was then added to the ground glass, and this mixture was again homogenized by grinding under alcohol. After drying at 120 °C for 50 hours this material was used for the experiments with various amounts of added bidistilled water.

To produce material with larger amounts of glass, which can be more accurately analyzed by electron microprobe, we prepared three other starting compositions (E4, E6 and E7). These were calculated as mixture of ca. 70% melt and crystalline minerals from the run products of experiments with E3. These compositions are also given in Table 1.

#### **Experimental methods**

The conventional piston-cylinder apparatus has been used. The starting mixture together with measured amounts of water has been hermetically sealed by welding in gold tubes, which were placed in a cell made of NaCl with graphite heaters. Temperatures were measured with chromel-alumel thermocouples. Earlier it has been demonstrated that for NaCl cells friction correction for pressure is negligible (*Mirwald* et al., 1975).

Run products have been studied under the optical microscope, and the compositions of the synthesized phases were measured by electron microprobe (ARL-SEMQ II) with an attached energy dispersive system. Operating conditions were 15 keV accelerating voltage and 15 nA beam current. Both high pressure experiments and electron microprobe analyses were performed at the Institute for Mineralogy and Petrography, University of Innsbruck.

#### Results

Run conditions and observed phase assemblages in the quenched products are given in Table 2 and the results of the microprobe analyses of clinopyroxenes and garnets are shown in Tables 3 and 4. Crystalline phases are represented by Amph + Cpx + Qz

	P, kbar	T, °C	Duration, hours	Silicate mixture	Water added (wt %)	Phase assemblage
1	26	800	20	E3	6.58	Qz + Cpx + Ga + L
2	24	750	25.5	E3	6.81	Qz + Amph + Cpx + L
3	28.5	750	47	E3	6.70	Qz + Cpx + Ga + L
5	28.5	800	20.5	E3	6.61	Qz + Cpx + Ga + L
6	28.5	700	45	E3	6.22	Qz + Cpx + Ga + Qnch Fl
7	28.5	850	38	E3	7.44	Qz + Cpx + Ga + L
8	28.5	800	44.5	E3	13.24	Qz(sa) + Cpx + Ga + L
10	28.5	800	46	E4	8.75	Qz + Ky(sa) + Rut(sa) + Ga + L
11	28.5	800	38	E6	9.24	Cpx + Ga + Rut(sa) + L
12	28.5	820	53	E7	5.97	Cpx + Ga + Qz + L + Rut(sa)
13	35	900	46	E7	11.52	Cpx + Ga + Cs + L + Rut(sa)
14	35	820	48	E7	11.86	Cpx + Ga + Cs + L + Rut(sa)

 Table 2. Run conditions and resulting phase assemblages

Note: Qz quartz; Cs coesite; Cpx clinopyroxene; Ga garnet; L quenched liquid (glass); Ky kyanite; Rut rutile; Qnch Fl quenching products of fluid phase; sa small amount

Table 3. Compositions of clinopyroxenes from run products, determined by electron microprobe analyses (normalized to 100%)

	Run 1	Run 2	Run 3	Run 5	Run 6	Run 7	Run 8	Run 11	Run 12	Run 13	Run 14
SiO <sub>2</sub>	54.89	50.87	55.44	55.89	56.16	54.41	53.54	55.52	56.38	56.14	56.01
TiO <sub>2</sub>	0.35	0.68	0.30	0.14	0.73	0.43	0.35	0.36	1.01	0.43	0.65
Al <sub>2</sub> Õ <sub>3</sub>	8.73	7.14	9.02	9.22	12.53	9.19	10.19	14.09	14.51	12.12	15.84
FeO	8.05	9.00	4.53	3.30	8.45	4.57	7.52	6.07	5.35	3.96	5.83
MgO	10.54	11.84	12.08	11.84	6.09	11.82	10.01	7.45	6.33	8.60	5.04
CaO	13.92	18.40	14.89	15.38	10.72	15.90	15.08	9.65	8.68	11.95	8.77
Na <sub>2</sub> O	3.52	2.08	3.74	4.22	5.32	3.67	3.32	6.86	7.73	6.91	7.87

Table 4. Compositions of garnets from run products, determined by electron microprobe analyses (normalized to 100%)

	Run 7	Run 8	Run 10	Run 11	Run 12	Run 13	Run 14
SiO <sub>2</sub>	39.83	39.93	39.08	39.04	41.63	39.88	40.36
TiO <sub>2</sub>	0.98	0.71	1.36	0.98	1.11	1.45	0.67
$Al_2 \tilde{O}_3$	21.71	22.15	21.52	21.55	23.12	21.37	21.97
FeO	17.93	17.90	22.58	23.40	16.14	20.78	19.71
MgO	10.43	10.17	5.74	7.06	11.10	9.04	10.23
CaO	9.12	9.15	9.72	7.89	6.90	7.48	7.07

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Fig. 1. Back-scattered electron photomicrograph of the products of run 13. Light grey equant grains are garnets, darker grey elongated crystals are clinopyroxenes, much larger dark elongated grains are coesites, rare small very bright grains are rutiles, all surrounded by glass. Length of bar is  $50 \,\mu\text{m}$ 

at 24 kbar (run 2) and by Qz (or Cs) + Ga + Cpx at higher pressures. This places the amphibole-out boundary close to 25 kbar.

Unlike hydrous basaltic melts which are always quenched into a mixture of solid phases, the products of our experiments always contained clear glasses without quench crystals. There are also no noticeable quench overgrowths on the mineral phases immersed in the glass. Garnets were typically represented by euhedral equant crystals of varying sizes up to  $15-20 \,\mu\text{m}$  in cross section, clinopyroxenes were present as elongated grains up to  $5 \times 30 \,\mu\text{m}$ , rutile was represented by rare anhedral small grains, whereas quartz and coesite were in many cases represented by much larger crystals (tens of  $\mu\text{m}$  across). All minerals were usually rather uniformly distributed in the glass, but occasionally larger pools of quenched liquid (Fig. 1) were observed.

Microprobe analyses of hydrous silicate glasses in the products of the runs with smaller fractions of a liquid phase indicate significant loss of Na and K due to the volatilisation of alkalis. Scanning of large areas was not possible because of the fine grained character of the synthesized material. In order to estimate the composition of the melt we used the ratios between Si, Ti, Al, Mg and Ca from microprobe analyses of glasses, and calculated sodium, potassium and water contents based on mass balance considerations. For this purpose a computer programme was developed. It starts from the input of the first approximation of the weight fractions of melt, clinopyroxene and garnet for a given experiment ( $F^L$ ,  $F^{Cpx}$  and  $F^{Ga}$ ; the fraction of quartz is calculated as  $F^{Qz} = 1 - F^L - F^{Cpx} - F^{Ga}$ ). Then  $C^L_{H_2O}$  are calculated as  $C^{Blk}_i/F^L$  (assuming that these components are present only in melt;

	Run 7	Run 8	Run 10	Run 11	Run 12	Run 13	Run 14
SiO <sub>2</sub>	60.60	53.16	58.08	62.27	64.80	62.42	57.54
TiO <sub>2</sub>	0.28	0.24	0.29	0.22	0.17	0.38	0.12
$Al_2 \tilde{O}_3$	10.69	9.73	14.48	13.29	13.12	10.94	9.86
FeO	1.09	1.53	1.39	1.05	0.80	0.47	0.42
MgO	0.65	1.11	2.22	1.14	0.41	0.33	0.18
CaO	1.72	1.70	3.47	1.84	0.80	1.13	0.75
Na <sub>2</sub> O	4.43	3.38	3.28	5.24	4.22	4.43	2.52
K <sub>2</sub> Õ	3.08	2.48	3.51	2.29	2.71	1.85	2.60
H <sub>2</sub> O	17.46	26.66	13.27	12.64	12.97	18.04	26.01

Table 5. Compositions of melts calculated by mass balance based on microprobe analyses ofglasses from run products

 $C_i^{\text{BIK}}$  are bulk concentrations in the material sealed in the capsule) and for sodium we use the formula  $C_{Na_2O}^{\text{L}} = (C_{Na_2O}^{\text{BIK}} - C_{Na_2O}^{\text{Cpx}} \cdot F^{\text{Cpx}})/F^{\text{L}}$ . The fractions of these phases are then varied according to the SIMPLEX algorithm until the minimum of the squares of deviations of bulk concentrations from calculated values  $F^{\text{Cpx}} \cdot C_i^{\text{Cpx}} + F^{\text{Ga}} \cdot C_i^{\text{Ga}} + F^{\text{Qz}} \cdot C_i^{\text{Qz}} + F^{\text{L}} \cdot C_i^{\text{L}}$  is reached. The melt compositions estimated in this way are given in Table 5.

The examination of the products of run 6 (700 °C, 28.5 kbar) under the optical microscope has shown the presence of small amounts of glass-like material. This phase has also been observed under the electron microprobe as small isolated irregularly shaped segregations occupying the interstitial space between the minerals. EDS spectra of this material yielded high Al/Si ratios, and radiation damage was particularly severe. This compositional feature is very different from the analyses of glasses representing quenched silicate melts at 750 °C and at higher temperatures. This suggests that 700 °C at 28.5 kbar correspond to subsolidus conditions and that the amorphous phase represents material precipitated from a quenched water-rich fluid with alkali aluminosilicates as predominant components. These results show that the solidus in the quartz eclogite—water system lies at 28.5 kbar between 700 and 750 °C.

Run 8 was conducted with a larger amount of water compared to other experiments. Quenched glass in these run products contained small bubbles, which probably indicates the presence of a separate gas phase. In this case the water content in the melt would be lower than about 27% as shown in Table 5. It is also possible, that the observed bubbles are due to vesiculation which occurred during quenching of the water-rich melt and that a separate gas phase was absent during the run. The significant difference between the results of runs 5 and 8 (both at 800 °C and 28.5 kbar, but with different amounts of  $H_2O$ ) with a much larger proportion of glass and the almost complete disappearance of quartz in run 8 (higher  $H_2O$  content) favours the latter assumption.

The weighted average concentrations of  $TiO_2$  calculated from analyses of individual phases in the products of experiments with the E3 starting composition

were always lower than the bulk content in the starting material. This could be explained by the presence of ca. 0.5% of tiny grains of rutile which could not be identified by the methods used. The presence of a Ti-rich phase is also confirmed by very high Ti concentrations (sometimes in excess of 10%) obtained occasionally at some points on glass or crystalline minerals. From this we may conclude that the estimated concentrations of TiO<sub>2</sub> in the melts in the range of 0.2–0.3% correspond to saturation with respect to rutile. Rutile was actually observed in run products with E4, E6 and E7 starting compositions.

Run 10 which was conducted with E4 starting composition had no clinopyroxene in the run products. Instead, small amounts of kyanite and rutile were found together with garnet, quartz and glass.

Runs 10–14 were characterised by higher melt/solids ratios compared to previous runs, and there were patches of glass in the run products large enough to be analysed by scanning electron beam avoiding loss of alkalis. The concentrations of all elements including sodium and potassium measured by electron microprobe in the glass from this run are consistent within 0.5 wt% with the values calculated on the basis of mass balance considerations as described above.

#### Discussion

The high-pressure amphibole out boundary (ca 25 kbar) and solidus temperature (between 700 and 750 °C) observed in our experiments are in good agreement with the results of previous works for various basaltic and andesitic compositions (*Stern* and *Wyllie*, 1978; *Green*, 1982; *Rapp*, 1991; *Rushmer*, 1991; *Yaxley* and *Green*, 1994). The partial melt compositions are very similar to those estimated by *Stern* and *Wyllie* (1978). Although they did not report any silica minerals in the run products, their experiments in the andesite-H<sub>2</sub>O system were probably very close to saturation conditions with respect to quartz or coesite. Therefore, our experimental data favour the conclusion that hydrous silicate melts produced by partial melting of a basaltic composition after the breakdown of amphibole in quartz eclogite do not have andesitic or dacitic, but rather rhyolitic compositions.

Mass balance calculations based on the compositions of melt and minerals from run 12 and the bulk composition corresponding to altered MORB (GA1 in Table 1) show that at 820 °C and 28.5 kbar quartz is completely dissolved by melt (10–15%) and that the residual mineralogy is represented by bimineralic eclogite (garnet, clinopyroxene and 0.9% of rutile). The removal of approximately 10–15% of a partial melt similar to that observed in run 12 would result in an almost complete loss of H<sub>2</sub>O and K<sub>2</sub>O, reduction of SiO<sub>2</sub> content by about 2% and increasing concentrations of CaO, MgO, FeO and TiO<sub>2</sub>. The temperatures at which such melts may be produced lie close to the P-T paths estimated for parts of subducting slabs (*Peacock* et al., 1994).

The considered example requires about 2% H<sub>2</sub>O in the bulk basaltic composition. This is somewhat higher than the amount of water which may reside in amphiboles of eclogitic phase assemblages before their breakdown (ca 1%, *Poli*, 1993). This indicates that partial melting of quartz eclogites may require additional H<sub>2</sub>O. Melting of subducted basaltic crust at higher water contents has been also suggested by *Ringwood* (1990) who invoked the dehydration of serpentinites in the underlying parts of the subducting slab as a source for additional  $H_2O$ . Recently this hypothesis found experimental confirmation in the work by *Ulmer* and *Trommsdorff* (1996).

The SiO<sub>2</sub>-rich melts generated during the melting of basaltic material at mantle depths would be in disequilibrium with the surrounding peridotitic rocks. They could induce an enrichment of the overlying mantle wedge with incompatible elements and low melting components (*Ringwood*, 1990). The consequences of such processes would be an increasing content of orthopyroxene in peridotites accompanied by phlogopitization, the appearence of more sodic pyroxenes and possibly amphibolization. However, the possibility exists that some acid magmas above subduction zones may represent melts produced in the eclogite stability field. Comparison of melt compositions recalculated on anhydrous basis from runs made at 28.5 and 35 kbars shows a tendency for increasing SiO<sub>2</sub> content with rising pressure. If this is so, then eclogite derived granitic rocks should be looked for among anomalously siliceous compositions.

The analyses of glasses in the products of our experiments permitted also an extension the existing experimental data on rutile saturation in silicate melts (*Green* and *Pearson*, 1986; *Ryerson* and *Watson*, 1987) to more siliceous compositions and lower temperatures at high pressures. TiO<sub>2</sub> contents presented in Table 5 are consistent with extrapolation to the lower temperatures of experimental results of *Green* and *Pearson*, and they are in excellent agreement with the empirical formula proposed by *Ryerson* and *Watson*. Comparison of titanium contents in eclogite xenoliths from kimberlites, in melts which could have been in equilibrium with them and in various basaltic materials may impose some restrictions on the nature of protolith for mantle eclogites.

, According to the compilation of Ireland et al. (1994) mantle eclogites with MgO contents close to basaltic compositions (10% and less) are characterized by a range of TiO<sub>2</sub> concentrations between 0.5 and 0.9%. Archean basalts are favored by Ireland et al. (1994) as protoliths for mantle eclogites. At ca. 10% MgO their TiO<sub>2</sub> contents are in the range of 0.3-1.5%. The upper limit of this range is similar to modern MORBs, whereas the lower limit is closer to boninites. The separation of acid melts with low  $TiO_2$  contents (0.2–0.3%) similar to those observed in our experiments or even of less siliceous magmas of andesitic composition (about 1% TiO<sub>2</sub> in rutile saturated melts—Green and Pearson, 1986; Jenner et al., 1994) from MORB-like material should result in an increase in the concentration of titanium, and therefore would not yield typical eclogitic restite. Partial melting of boninite-like low-TiO<sub>2</sub> rocks seems to be a more likely explanation for mantle eclogite formation. Their initially low TiO<sub>2</sub> contents may be slightly increased during the separation of granite-like magmas. It is possible that very siliceous near-solidus liquids appear at the initial stage of the melting process and which are subsequently replaced by more mafic magmas. The order of events during this process would depend on the mechanism of partial melting: melting reactions induced by amphibole breakdown or magma generation in the subducting crust induced by the flux of volatiles derived from underlying dehydrating serpentinites (Ulmer and Trommsdorff, 1995).

#### Conclusions

- 1. Near-solidus hydrous melts in equilibrium with quartz eclogite are represented by rhyolitic magmas with SiO<sub>2</sub> contents exceeding 70% on a volatile-free basis.
- 2. The generation of ca 10-15% melt of this composition will eliminate quartz from the high pressure phase assemblage of the average altered MORB composition.
- 3. The solidus temperature for the system quartz eclogite—water lies between 700 and 750 °C at 28.5 kbar, just above the high-pressure amphibole-out boundary.

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