

## ARC CRUST FORMATION AND DIFFERENTIATION CONSTRAINED BY EXPERIMENTAL PETROLOGY

OTHMAR MÜNTENER<sup>\*,†</sup> and PETER ULMER<sup>\*\*</sup>

**ABSTRACT.** We review igneous experimental data designed to understand fractionation processes in the roots of magmatic arcs, focusing on fractional crystallization experiments in dry and hydrous basalts to andesites to simulate the liquid and cumulate lines of descent of primary mantle derived magmas. An evaluation of the major element composition indicates that the cumulate line of descent (CLD) of hydrous systems is fundamentally different from dry systems. Cumulates derived from hydrous experiments display elevated  $\text{Al}_2\text{O}_3$  and CaO contents at low  $\text{SiO}_2$ , producing voluminous andesitic to rhyolitic liquids that closely overlap with compositions of natural systems. The mineralogical and chemical composition of cumulates converges for very different hydrous primary mantle magmas, indicating that fundamental phase equilibria under the conditions prevailing in the roots of magmatic arcs exert a strong control on the compositions of derivative andesitic to rhyolitic liquids. Melting experiments on amphibolite or basaltic to andesitic compositions equally produce granitic – rhyolitic liquids of similar composition yet their restites do not present the same variability of the CLD of hydrous fractional crystallization experiments.

The comparison of our experimentally produced cumulates with natural rock suites from the Jurassic Talkeetna arc (South Central Alaska) and the Cretaceous Kohistan arc (Pakistan) show that fractionation experiments closely reproduce the natural rock compositions and in particular the cumulate sequences exposed in the roots of island arcs. Both dry and hydrous fractionation series are preserved yet only the hydrous fractionation series produces voluminous ultramafic cumulates (50–70%) that are negatively buoyant with respect to mantle peridotite. Delamination of dense ultramafic roots can explain the andesitic composition of continental crust. Our fractionation experiments show that generation of granites is a natural consequence of fractional crystallization of primary basaltic magmas. Discrepancies of liquid compositions produced in the lower crust and the arc volcanic record can readily be explained by further differentiation, partial assimilation and mingling and mixing processes during ascent and decompression in transcrustal magmatic systems. We therefore favor the interpretation that Earth-like continental crust is primarily produced by fractional crystallization in the lower crust and upper mantle, and that magma mixing, assimilation, metamorphism and crustal melting are important secondary processes that refine and amplify the difference and stratification between mafic lower crust and a granitic upper crust.

Keywords: fractional crystallization, experimental petrology, arc lower crust, cumulate lines of descent, liquid lines of descent

### INTRODUCTION

The composition of the continental crust of the Earth is unique in the solar system and is the most accessible witness of differentiation processes. As initially proposed and demonstrated by Bowen (Bowen, 1915a; Bowen, 1915b; Bowen, 1928) crystal liquid segregation is a fundamental mechanism that is able to separate/differentiate a chemically uniform silicate liquid into mineral assemblages or cumulates that have significantly different compositions than the residual melt. This mechanism commonly referred to as fractional crystallization generates the entire range of silica contents ranging from mantle-derived basaltic magmas to rhyolites (for example, Nandedkar

\* Institute of Earth Sciences, Géopolis 4897, CH-1015 Lausanne, Switzerland

\*\* Institute of Geochemistry and Petrology, ETH, Clausiusstrasse 25, CH-8092 Zurich, Switzerland

† Corresponding author: othmar.muntener@unil.ch

and others, 2014). Primary liquids originating from the mantle that are in equilibrium with at least olivine and orthopyroxene and one or more Ca and Al-rich phases (cpx, garnet, spinel, plagioclase) are generated either by near-adiabatic decompression melting beneath mid-ocean ridges and in the mantle wedge of subduction zones or/and by volatile-rich flux melting in subduction zones. Numerous natural and experimental studies of basalts established that primary, mantle-derived silicate liquids in equilibrium with the mantle are characterized by Mg# (molar  $\text{Mg}/\text{Mg}+\text{Fe}_{\text{tot}}$ ) between  $\sim 0.65$  and  $0.75$  and  $\text{SiO}_2$  contents between 45 and 57 weight percent (for example, O'Hara, 1965; Ulmer, 2001), substantially different than estimates of the bulk continental crust (for example, Arculus, 1981; Taylor and McLennan, 1985; Rudnick and Gao, 2003). Because continental crust is enriched in highly incompatible elements and the same elements are depleted in the upper mantle, there is a consensus that generating Earth's continental crust is ultimately related to melting of the mantle (Hofmann, 1988), yet it remains uncertain whether mantle melting alone can explain the diversity of Earth crust. Indeed, the incompatible trace element patterns and some trace element ratios such as Nb/U, Ce/Pb and Ce/Yb indicate that the bulk continental crust has some striking similarities with arc crust and, therefore, there is some consensus that large parts of the continental crust probably formed by subduction-related arc magmatism, that gave rise to the andesite crustal model (for example, Taylor and White, 1965; Gill, 1981) at least since the Proterozoic, while this subject is controversial for older rock suites (for example, Bédard, 2006; Bédard and others, 2013). However, most studies question the possibility that granitic rocks form directly by fractionation from basaltic magmas, based on ubiquitous inherited and/or antecrystic zircons in many granites (for example, Miller and others, 2007), and textural evidence for 'petrologic cannibalism' in siliceous plutonic and volcanic rocks (for example, Eichelberger, 1975; Davidson and others, 2007) and, therefore, call upon the necessity of at least two processes that are required for the formation of felsic continental crust: (i) primary liquids crystallize ultra-mafic and mafic cumulates to produce intermediate to silicic-magmas, (ii) and/or intra-crustal melting will produce mafic residues and evolved silicate liquids similar to continental crust (Davidson and Arculus, 2005). While we do not dismiss the evidence for mixing in the forms of basaltic to andesitic enclaves, resorbed phenocrysts, reversed phenocryst compositions and disequilibrium assemblages in arc volcanic rocks (for example: Hildreth, 1983), nor the evidence of assimilation of isotopically very different country rocks, we will show that experimentally derived cumulates from fractional crystallization experiments show striking similarities to arc lower crust. In any case, to balance the discrepancy between primary magma composition in equilibrium with residual mantle assemblages and the average continental crust some sort of density sorting is required [delamination, or subduction of material back into the mantle, (Bird, 1979; Kay and Kay, 1988; Arndt and Goldstein, 1989; DeBari and Sleep, 1991; Jull and Kelemen, 2001)]. Inspired by numerical models (for example Gerya and Yuen, 2003) so-called 'relamination' models have become popular in recent years in explaining the composition of the continental crust, yet such models have difficulties to explain the huge volumes of mafic and ultramafic plutonic rocks with near-mantle like isotopic compositions building the roots of juvenile island arcs (for example, Dhuime and others, 2007).

The major difficulties of accurately estimating the composition of island arc crust are threefold: (1) there are very few continuously exposed cross sections of island arc crust from volcanics to the Moho and below, (2) post-crystallization (subsolidus) metamorphic equilibration has affected the mineral assemblages of exposed intra-oceanic and continental lower arc crust (for example, Kohistan, Talkeetna, Sierra Nevada, Famatinian arcs) (a.k.a. granulitization), which generally increases modal

amounts of garnet and, thus, overestimates the density of the original arc cumulates. Cumulate fragments erupted in active arc volcanoes, however, are often ambiguous and potentially represent complex multiphase – multipressure assemblages (for example, Stamper and others, 2014; Melekhova and others, 2015). Calculated cumulates based on experimental studies between 0.7 and 1.2 GPa provide the initial adcumulate composition without subsolidus modifications; and (3) perhaps most important is the estimate of the intermediate to felsic composition of island arcs, since preserved examples such as the Sierra Nevada and Kohistan arc are plagued by representing a time-integrated picture of many different batches of melts that crystallized over 10s of millions of years. However, primary arc magmas in single volcanoes are likely to vary irregularly between tholeiitic and calc-alkaline compositions on the <1 Myr timescale (for example, Grove and others, 2002) and their LLD (liquid line of descent) and CLD (cumulate line of descent) are significantly different. Disentangling the different liquid lines of descents between hydrous and damp/dry magmas as a function of time in such settings is a formidable task, and integrating bulk volumes of hundreds of square kilometers are affected by errors that are not usually taken into account in studies that simply report mass balances to estimate the bulk compositions of arc crust. Our experimentally–constrained cumulate and liquid compositions are designed to further discuss and distinguish primary compositions from the complexly assembled igneous and metamorphic rocks from natural examples.

Experimental constraints on the generation of continental crust by fractional crystallization in the range of 0.7 to 1.5 GPa are few. Indeed most studies either focused on: (i) phase relationships of primitive basalts at pressures between 1 and 3 GPa to evaluate multiple saturation with a lherzolitic or harzburgitic residues (for example: Tatsumi, 1981; Baker and others, 1994; Parman and Grove, 2004; Wood and Turner, 2009) or (ii) on partial melting and advanced crystallization of non-primitive compositions (andesites, evolved basalts) that may be present in the lower crust (Wolf and Wyllie, 1994; Sisson and others, 2005; Alonso Perez and others, 2009; Blatter and others, 2013). In order to understand crystal fractionation processes in the sense of Bowen, it is, however, necessary to provide experimental data that follow as closely as possible the LLD and the CLD in order to estimate the critical compositions, pressure and temperature conditions from which quartz diorites (andesites) and more evolved arc magmas are extracted. Constraints from experimental petrology on arc lower crust are still rare, mainly due to experimental difficulties of performing H<sub>2</sub>O–undersaturated crystallization experiments in natural, Fe-bearing systems. Over the last 15 years we have explored the effects of variable bulk composition of parental melts, fO<sub>2</sub> and H<sub>2</sub>O on the phase proportions of ultramafic and mafic plutonic rocks. By applying modified experimental techniques (Hall and others, 2004; Kägi and others, 2005) and by simulating equilibrium and fractional crystallization (Müntener and others, 2001; Villiger and others, 2004; Villiger and others, 2007; Alonso Perez and others, 2009; Nandedkar and others, 2014; Ulmer and others, 2018) we calculated a variety of ‘experimental cumulates’ in dry and wet systems to simulate the CLD. We also compiled experimental studies on partial melting of hydrous mafic rocks at intermediate to lower crustal conditions (Wolf and Wyllie, 1994; Sisson and others, 2005; Blatter and others, 2013) to explore partial melting and fractional crystallization experiments to better understand lower crustal processing of arc magmas and the links to the chemical composition of the continental crust. This is relevant in particular for the discussion on the origin of granitic rocks. While sediment and amphibolite melting are important processes that contribute to the diversity of granites, fractional crystallization studies indicate that the formation of granites does not require crustal melting. This is fundamental to maintain the hypothesis that continental crust mainly formed in arcs, at least since the Archean.

The main objectives of our study are: (1) to provide a summary of experimentally produced plutonic rocks by fractional crystallization at pressures relevant for both moderately juvenile (25–35 km) and mature (35–45 km) island arc lower crust and compare these assemblages to residues of partial melting of basaltic to andesitic rocks; (2) to provide experimental data on plutonic rocks that crystallized both from ‘wet’ and ‘damped/dry’ magmas since both arc-tholeiitic and calc-alkaline fractionation trends are the dominant processes of active and fossil island arcs (Grove and others, 2002; Tatsumi and others, 2008) and (3) to quantify the amounts of granitoid liquids produced along the LLD with respect to mafic/ultramafic cumulates for both dry (damped) and wet magmas. By comparing the SiO<sub>2</sub> and CaO content of cumulates and liquids we further demonstrate that the extraction depth of the primary magmas has a surprisingly small influence on the variability of ultramafic to mafic plutonic rocks and their respective liquid and cumulate line of descent (LLD and CLD) thereby relaxing long-standing controversies on the relative importance of primary high Mg# andesite and basaltic rocks on the construction of island arc crust.

#### EXPERIMENTAL METHODS

High-pressure experiments were performed in end-loaded piston cylinder apparatus with a 14mm bore. NaCl-Pyrex-MgO assemblies with a friction correction of ~3 percent applied to the nominal pressure were used. The temperatures were monitored with Pt/Pt<sub>90</sub>-Rh<sub>10</sub> (S-type) and Pt<sub>70</sub>-Rh<sub>30</sub>/Pt<sub>94</sub>-Rh<sub>6</sub> (B-type) thermocouples and accurate to within ±10 °C. For experiments at relatively low fO<sub>2</sub> (constrained close to the C-CO-CO<sub>2</sub> equilibrium) the Pt-graphite double capsule technique was used (Ulmer and Luth, 1991). For experiments at higher fO<sub>2</sub> (~QFM+1) a sample-in-sample modified double capsule technique was applied (Kägi and others, 2005). Welded shut, Fe-preconditioned Au<sub>90</sub>Pd<sub>10</sub> capsules were placed in an outer Pt or AuPd capsule that contains the same starting material. A limited number of experiments cooler than 950 °C were performed in single Au capsules (fig. 1).

The starting compositions, representative of a broad range of primary and derivative arc-magmas, are listed in table 1. A natural high Mg-basaltic dike (Ulmer, 1986; Hürlimann and others, 2016), synthetic analogues of a high Mg basaltic andesite (Ba, Baker and others, 1994) an olivine tholeiite derivative of the high Mg-basalt (OT, Nandedkar and others, 2014), and an andesite derived by experimental fractional crystallization of the high Mg basalt were used for equilibrium crystallization experiments (an, Alonso Perez and others, 2009). The initial H<sub>2</sub>O content for the mantle derived Mg-basalt, olivine tholeiite and high Mg basaltic andesite varies between 2.6 and 5 weight percent, regarded as a reasonable range of H<sub>2</sub>O-contents of primary, arc-related calc-alkaline to tholeiitic basalts and picrites, for example (Baker and others, 1994). Some of the experiments were nominally dry (MT, Villiger and others, 2004; Villiger and others, 2007) to simulate tholeiitic differentiation, yet nearly dry olivine tholeiites formed by decompression melting are common arc magmas (for example, Sisson and Bronto, 1998) and are relevant for our understanding of crust formation in arcs. The initial H<sub>2</sub>O content of the synthetic andesite ranged between 4 and 8 weight percent (Alonso Perez and others, 2009). An approximation to simulate fractional crystallization was achieved by a stepwise approach (fig. 2). The liquid compositions were determined in each experiment. The adcumulate composition can be understood as instantaneous solid composition (ISC, Morse, 1976) for the temperature interval of interest between two successive experiments. The successive experiment tracking the evolution of the liquid composition was performed at a 30 °C (and in a few cases 50 °C) lower temperature starting with a synthetic mix of the liquid composition from the previous experiments (figs. 1 and 2). For details of the step-wise fractional crystallization approach, the reader is referred to Villiger and others (2004) and Nandedkar and others (2014). Fractional crystallization experiments under more

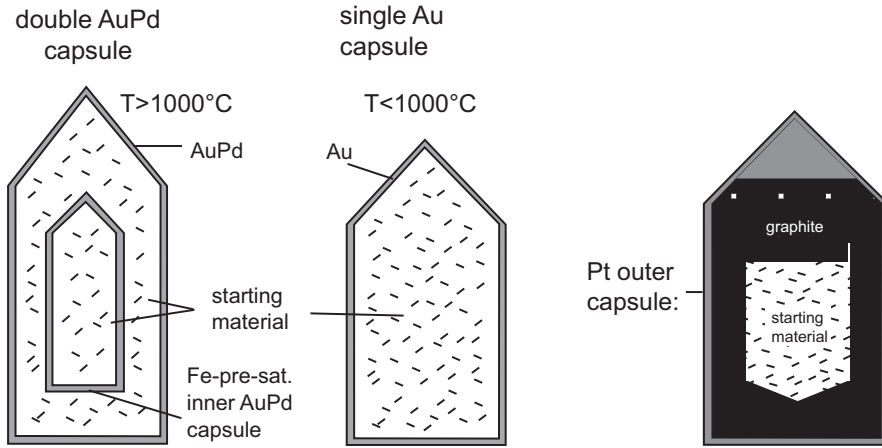
**(A) Oxidized Experiments (NNO)    (B) Reduced Experiments (C-COH)**

Fig. 1. Schematic illustrations showing the setup of the capsule techniques used in this study. (A) Hydrous experiments at temperatures  $> 1000\text{ }^{\circ}\text{C}$  are performed in AuPd double capsules with the same starting material in the inner and outer capsules. Fe-presaturated inner capsules are used (modified from Kägi and others, 2005). Hydrous experiments at temperatures lower than  $1000\text{ }^{\circ}\text{C}$  were mostly performed in single and/or double gold capsules. (B) Nominally anhydrous experiments were performed in conventional Pt-C double capsules (after Villiger and others, 2004).

oxidizing conditions were conducted by setting the ferric/ferrous iron ratio in the starting material to a value corresponding to  $f\text{O}_2$  conditions around  $\text{FMQ}+1$  at a given pressure and temperature according to the algorithm of Kress and Carmichael (1991). This approach only strictly applies/works when the amount of liquid in each experiment remains large, thus not modifying the ferric/ferrous ratio of the liquid phase significantly and can only be applied to experiments simulating fractional crystallization where the liquid proportions always exceeded 75 percent unlike the equilibrium crystallization experiments where the melt fraction decreases with decreasing temperature.

#### LIQUID LINES OF DESCENT (LLD) AND CUMULATE LINES OF DESCENT (CLD)

For each experiment, mineral and melt compositions were determined by electron microprobe and modal proportions of minerals and melt were determined by least squares regression. Here we provide a dataset of cumulate compositions that were obtained by renormalizing the crystal fraction to 100 percent and using the mineral compositions (Wolf and Wyllie, 1994; Müntener and others, 2001; Villiger and others, 2004; Sisson and others, 2005; Alonso Perez, ms, 2006; Villiger and others, 2007; Alonso Perez and others, 2009; Blatter and others, 2013; Nandedkar and others, 2014; Ulmer and others, 2018). Liquid compositions from these studies and Beard and Lofgren (1991) were calculated anhydrous in order to compare the different crystallization and fractionation studies. Experimental temperatures, pressure, modal proportions and melt and cumulate compositions for various experimental studies are summarized in tables EA1 and EA2 available as Excel spread sheets in the electronic Appendix ([http://earth.geology.yale.edu/%7eajs/Supplementary Data/2018/Muntener/](http://earth.geology.yale.edu/%7eajs/Supplementary%20Data/2018/Muntener/)).

#### *Cumulate Lines of Descent (CLD)*

In figure 3 we illustrate the bulk cumulate composition for fractional and equilibrium experimental studies in terms of Harker diagrams [ $\text{SiO}_2$  vs  $\text{CaO}$ ,  $\text{SiO}_2$  vs  $\text{Al}_2\text{O}_3$ , and  $\text{SiO}_2$  vs

TABLE 1  
*Compositions of starting materials used in various experimental studies*

Label	RC158c	RC156	85-44 syn	85-44	F8a	HK19	HK19.2	01SB-872	87S35A	AD19-93	YOS-55A	Owens Amph	HQ36	AGS11.1
Acronym	Mb	OT	ba	ba	AP09	MT	MT	V07	B13	S05	S05	S05	WW94	SPD95
Study	U17, AP06	N14	U17	M01	AP09	V04	V07	V07	B13	S05	S05	S05	WW94	SPD95
SiO <sub>2</sub>	47.52	50.02	52.34	51.62	57.29	49.06	48.70	48.70	50.29	51.31	50.75	53.96	49.34	55.30
TiO <sub>2</sub>	0.72	0.70	0.64	0.60	0.95	0.60	0.57	0.57	1.19	1.29	1.25	1.03	0.41	0.94
Cr <sub>2</sub> O	0.17			0.09		0.36	0.34							
Al <sub>2</sub> O <sub>3</sub>	12.83	15.69	15.73	16.38	18.86	15.28	14.56	14.56	17.10	19.37	19.71	17.85	14.88	13.88
FeO <sub>tot</sub>	9.38	8.39	8.38	7.92	7.77	7.57	7.57	7.57	9.31	8.82	9.69	8.11	8.56	12.36
MnO	0.19	0.15	0.19	0.16	0.30	0.14	0.14	0.14	0.16	0.17	0.15	0.15	0.20	0.20
NiO	0.04			0.03										
MgO	17.08	10.46	10.25	10.78	2.96	13.04	14.82	14.82	8.27	4.38	4.52	5.43	10.91	5.37
CaO	10.35	11.95	9.91	9.66	7.44	12.31	11.73	10.10	10.10	8.98	7.39	9.33	14.58	10.13
Na <sub>2</sub> O	1.22	1.92	2.18	2.24	2.96	1.57	1.50	2.92	2.92	4.29	3.76	2.74	1.02	1.11
K <sub>2</sub> O	0.40	0.61	0.38	0.42	1.47	0.08	0.08	0.48	0.48	1.01	2.32	1.19	0.10	0.61
P <sub>2</sub> O <sub>5</sub>	0.10	0.10		0.11				0.18	0.18	0.38	0.46	0.21		0.10
Total	100.00	100.00	100.00	100.00	100.00	100.00	100.00	100.00	100.00	100.00	100.00	100.00	100.00	100.00
xMg	0.764	0.690	0.686	0.708	0.404	0.754	0.777	0.777	0.613	0.470	0.454	0.544	0.694	0.436

Study: U17 - Ulmer and others (2018); AP06 - Alonso Perez (PhD, 2006); N14 - Nandedkar and others (2014); M01 - Müntener and others (2001); V04 - Villiger and others (2004); V07 - Villiger and others (2007); B13 - Blatter and others (2013); S05 - Sisson and others (2005); WW94 - Wolf and Wyllie (1994); SPD95 - Skjerlie and Patino Douce (1995).

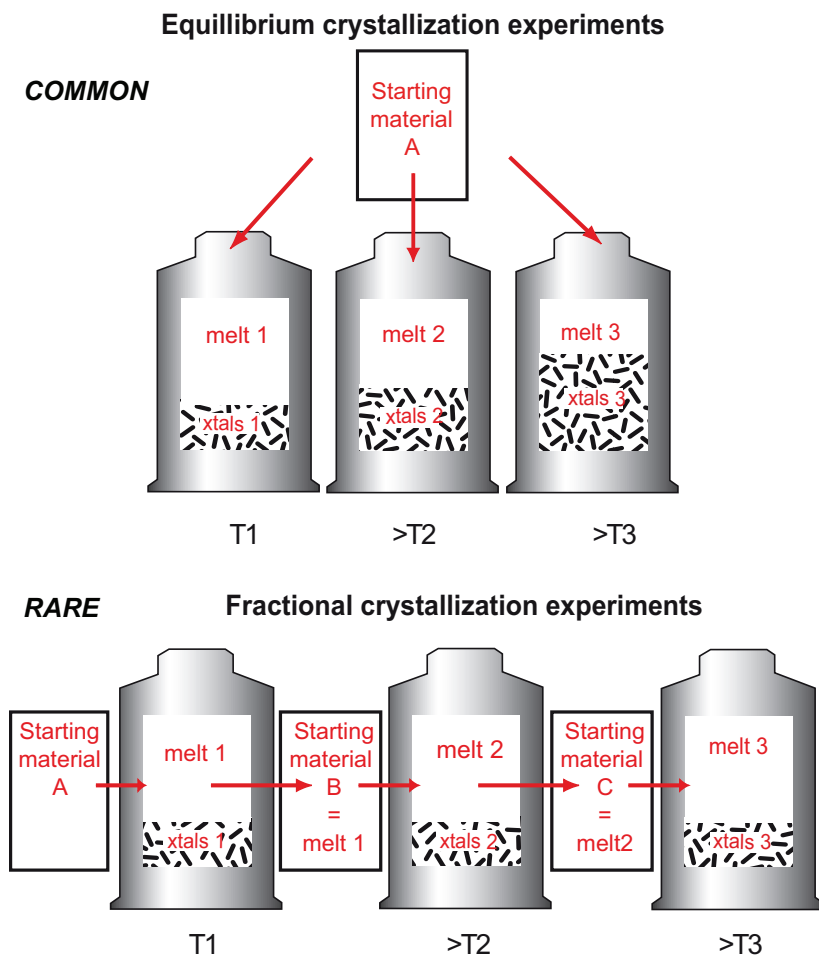


Fig. 2. Schematic illustration of the differences between ‘common’ closed system phase equilibrium experiments and ‘rare’ fractional crystallization experiments. Equilibrium crystallization experiments use one starting composition at a given pressure and vary temperature and/or composition to determine the phase relations and proportions of crystals and liquids. For the fractional crystallization experiments a step-wise approach was used. To simulate near-perfect fractional crystallization melting experiments were performed with temperature steps of 30° to 50° C and for each experiment a new starting composition corresponding to the measured liquid composition of the previous, higher temperature experiment was mixed. In this way the liquid line of descent of fractionating magmas can be approached. Starting compositions (Starting material A) for both equilibrium and fractional crystallization experiments vary from a high-MgO basalt and olivine tholeiite (Mg# 76 and 69) from the Adamello (Ulmer, 1986), to a basalt from partial melting experiments (HK#19, Mg# 76, (Hirose and Kushiro, 1993), to a basaltic andesite from Mt Shasta (85–44, Mg#: 0.71) and a high Mg# andesite (85–41c, Mg# 0.74; Baker and others, 1994). These starting materials correspond to mantle extraction depths between 2.7 and ~1.0 GP a, and cover the range of near primary liquids in intra-oceanic and continental arcs.

Mg# (molar Mg/Mg+Fe<sub>tot</sub> with all Fe as Fe<sup>2+</sup>). The rock types range from dunite, wehrlite, websterite, olivine clinopyroxenite to garnet-clinopyroxene hornblendite and hornblendite and amphibole gabbros for the hydrous experiments and dunite, lherzolite websterite, gabbronorite and gabbros for the dry experimental series. The differences in the chemical trends reflect differences in the underlying phase relations. The cumulates derived from equilibrium crystallization experiments display a

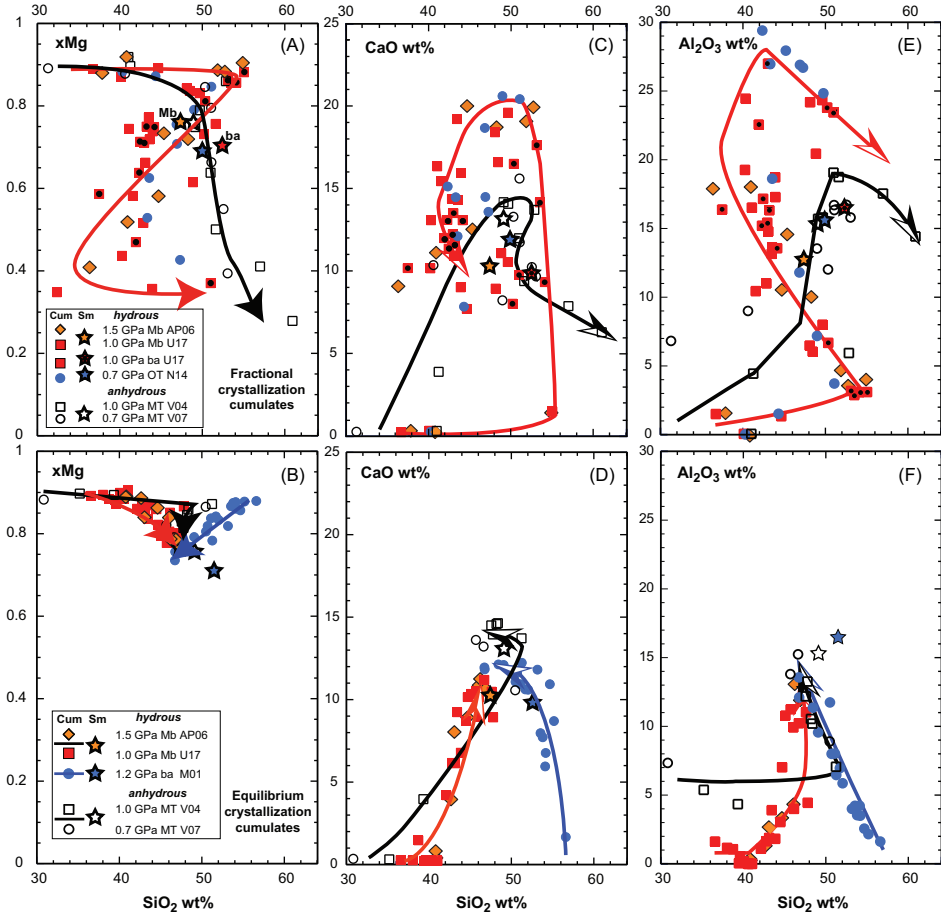


Fig. 3. Oxide concentrations and molar Mg# (molar Mg/Mg+Fe<sup>2+</sup>, with Fe<sup>2+</sup> as total Fe) of calculated bulk cumulate compositions for equilibrium and fractional crystallization experiments plotted versus wt% SiO<sub>2</sub>. Also shown are trajectories of cumulate evolution for the various experimental series during crystallization from the liquidus to ~900 °C. (A–B) Experimentally determined cumulate compositions in the Mg# vs SiO<sub>2</sub> reveal a “Z-shaped” evolution for hydrous fractionation experiments. Cumulates from equilibrium crystallization experiments display a limited range in Mg#; experiments on high-Mg basaltic starting materials at 0.7–1.5 GPa reveal decreasing Mg# and increasing SiO<sub>2</sub> along the CLD whereas the experiments on basaltic andesite at 1.2 GPa display the reverse trend related to extensive pyroxene crystallization followed by amphibole (plus garnet) at lower temperature. (C–D) CaO vs SiO<sub>2</sub> shows similar contrasting behavior for hydrous/anhydrous fractional and equilibrium crystallization. Hydrous fractional crystallization experiments reveal a counterclockwise evolution and reach highest CaO contents (nearly pure clinopyroxenites) whereas anhydrous fractional experiments show a clockwise evolutionary trend at lower CaO contents. (E–F) Al<sub>2</sub>O<sub>3</sub> vs SiO<sub>2</sub> reveals significant differences depending on crystallization mode, starting material employed and H<sub>2</sub>O-concentration: Hydrous fractionation experiments evolve along the CLD to highest Al<sub>2</sub>O<sub>3</sub> at lowest SiO<sub>2</sub> concentrations forming an S-shaped trend, whereas anhydrous fractionation experiments show a similar evolution at 1.0 GPa at lower maximum Al<sub>2</sub>O<sub>3</sub>-content. Lower Al<sub>2</sub>O<sub>3</sub> concentrations are mainly the result of considerably lower anorthite content of plagioclase and the absence of garnet in the crystallizing assemblage. Equilibrium crystallization experiments reach maximum Al<sub>2</sub>O<sub>3</sub> contents of 15 wt.%; the CLD evolves from low Al<sub>2</sub>O<sub>3</sub> to high Al<sub>2</sub>O<sub>3</sub> with increasing/decreasing SiO<sub>2</sub> contents for the Mg-basalt and basaltic andesite, respectively. The anhydrous equilibrium experiment evolve in two steps first increasing SiO<sub>2</sub> at low and near constant Al<sub>2</sub>O<sub>3</sub> (ol-cpx-opx) followed by decreasing SiO<sub>2</sub> and increasing Al<sub>2</sub>O<sub>3</sub> (cpx-spl-plag). Cum: cumulate composition; Sm: starting material; Mb: high Mg basalt; OT: olivine tholeiite; ba: basaltic andesite.



limited enrichment ( $\text{CaO}$ ,  $\text{Al}_2\text{O}_3$ ) and very limited range ( $\text{Mg}\#$ ) for all elements mainly because olivine and orthopyroxene build a substantial part of the calculated cumulates, while for the case of the fractional crystallization experiments the enrichment ( $\text{CaO}$ ,  $\text{Al}_2\text{O}_3$ ) and decrease ( $\text{Mg}\#$ ) is entirely different. The cumulates from the hydrous experiments define a z-shaped trend in the  $\text{Mg}\#$  vs  $\text{SiO}_2$  (fig. 3A), an s-shaped trend in the  $\text{Al}_2\text{O}_3$  vs  $\text{SiO}_2$  (fig. 3C), and an anticlockwise loop-like evolution in  $\text{CaO}$  vs  $\text{SiO}_2$  (fig. 3B). The cumulate line of descent (CLD) is composed of three segments: The ultramafic cumulates (dunites, wehrlites, olivine clinopyroxenites) are characterized by a substantial increase in  $\text{SiO}_2$  at high  $\text{Mg}\#$ , and low  $\text{Al}_2\text{O}_3$ , respectively. The second segment displays a decrease in  $\text{SiO}_2$  accompanied by a strong decrease in  $\text{Mg}\#$  and a strong increase in  $\text{Al}_2\text{O}_3$ , forming cumulates dominated by amphibole and garnet (and minor An-rich plagioclase and oxides), before increasing in  $\text{SiO}_2$  in the 3<sup>rd</sup> segment, forming mainly (garnet-bearing) amphibole gabbros (tables EA1 and EA2, <http://earth.geology.yale.edu/~7eajs/SupplementaryData/2018/Muntener>). Importantly, the  $\text{SiO}_2$  content is about 20 percent lower than the initial mantle melts at the end of the 2<sup>nd</sup> segment, a pre-requisite for Si-rich derivatives upon further differentiation. The counterclockwise evolution of  $\text{CaO}$  reflects the formation of clinopyroxene dominated cumulates up to >20 percent weight percent  $\text{CaO}$ , followed by increasing amounts of amphibole and garnet, which drives the bulk composition of these cumulates to lower  $\text{CaO}$  and  $\text{SiO}_2$ . Interestingly, the trends of hydrous CLD's are almost independent of the starting material used indicating that different primitive magmas in arcs form either Si-rich or Si-poor cumulate sequences at the base of the arc crust that modulate the derivative liquids to converge to amphibole-garnet-oxide  $\pm$  An-rich plagioclase compositions. In contrast to the hydrous experiments, the anhydrous evolution of cumulates is strikingly different and shows a continuous decrease of  $\text{Mg}\#$  with increasing  $\text{SiO}_2$ , a clockwise evolution at overall lower  $\text{CaO}$ , along the CLD, and a similar s-shaped trend of  $\text{Al}_2\text{O}_3$  with  $\text{SiO}_2$ , albeit at lower maximum  $\text{Al}_2\text{O}_3$ .

#### *Liquid Lines of Descent (LLD)*

Over the investigated temperature interval, the hydrous liquids evolve from basalt to andesite to dacite to compositions representing true granites and rhyolites (fig. 4). For each of the 3 fractionation series, the  $\text{Mg}\#$  decreases with increasing  $\text{SiO}_2$  (fig. 4A) up to about 68 weight percent  $\text{SiO}_2$ , before sharply increasing to higher  $\text{Mg}\#$  for rhyodacitic and rhyolitic compositions. The differences in the evolution of  $\text{Mg}\#$  between the 0.7 GPa and 1.0 GPa series reflect slight differences in controlling the Fe content in the experiments. Slight iron loss was not corrected for in the 0.7 GPa experiments (Nandedkar and others, 2014) resulting in minimum  $\text{Mg}\#$  in excess of 0.4 at about 68 weight percent  $\text{SiO}_2$ , whereas in the 1.0 GPa experiments  $\text{Mg}\#$  reach values as low as 0.2. In both series, however, the increase of  $\text{Mg}\#$  at  $\text{SiO}_2 > \sim 68$  weight percent is controlled by substantial crystallization of Fe-Ti oxides. The highly systematic evolution of  $\text{Mg}\#$  vs  $\text{SiO}_2$  is also observed in melting experiments under controlled  $f\text{O}_2$  (Sisson and others, 2005). They observed a significant increase in  $\text{Mg}\#$  under increasingly oxidized conditions, while experiments with  $f\text{O}_2 < \text{NNO}$  did not exceed  $\text{Mg}\#$  of 0.3 for granitic compositions. Since our experiments are not strictly buffered with respect to  $f\text{O}_2$ , it is remarkable that the trend of strongly increasing  $\text{Mg}\#$  closely matches the buffered experiments of Sisson and others (2005). This indicates that auto-oxidation might change the  $f\text{O}_2$  in experimental studies by several log-units (Ulmer and others, 2018) and implies that high  $\text{Mg}\#$  granitoids might be related to high  $f\text{O}_2$  during crystallization, while some other granites are characterized by low  $\text{Mg}\#$  (or high  $\text{FeO}_{\text{tot}}/\text{MgO}$ ) under rather reducing conditions (Turner and others, 1992; Sisson and others, 2005). Auto-oxidation or evolution towards increasing  $f\text{O}_2$  with

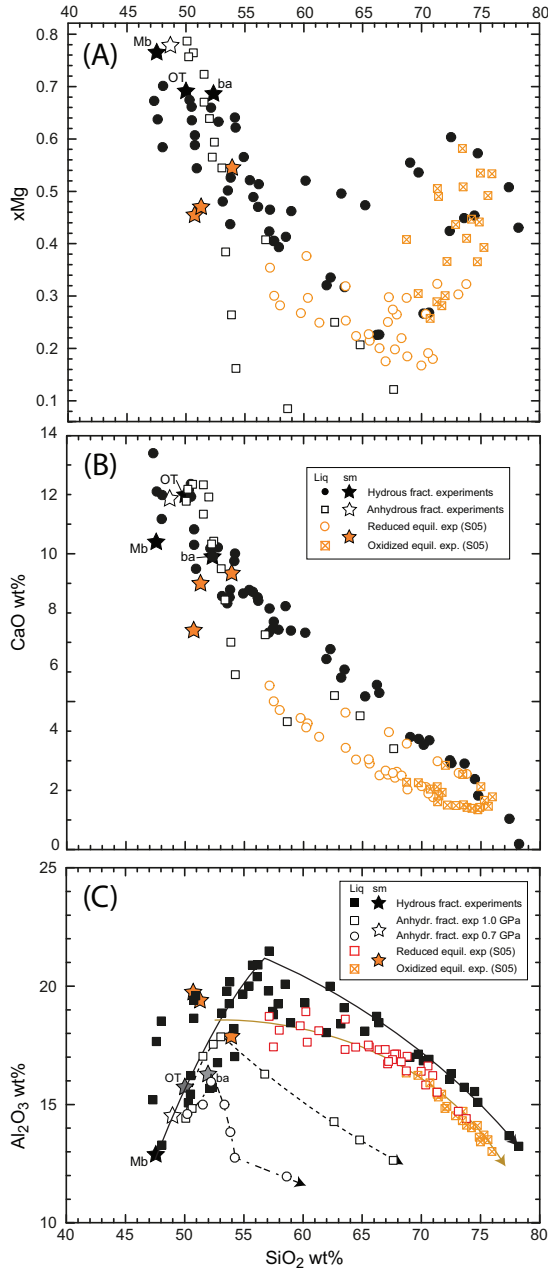


Fig. 4. Oxide concentrations and molar  $\text{Mg\#}$  of liquids (normalized anhydrous) derived from fractional crystallization experiments plotted versus wt%  $\text{SiO}_2$ . Black symbols represent hydrous fractional crystallization experiments, open symbols are nominally anhydrous fractional crystallization experiments. The orange circles are synthesized liquids at relatively reducing  $f\text{O}_2$  (QFM) while the orange squares are synthesized at oxidizing conditions (NNO-NNO+3), both from Sisson and others (2005). (A)  $\text{SiO}_2$  vs  $\text{Mg\#}$ : the hydrous fractional crystallization experiments produce granitic liquid compositions with a wide range of  $\text{Mg\#}$ , while anhydrous experiments show low  $\text{Mg\#}$  and limited  $\text{SiO}_2$  enrichment (up to 60wt%  $\text{SiO}_2$  at 0.7 GPa). (B)  $\text{SiO}_2$  vs  $\text{CaO}$ . All hydrous liquids plot along a straight line from andesitic to rhyolitic compositions, while anhydrous liquids and those of Sisson and others (2005) plot at lower  $\text{CaO}$  for a given  $\text{SiO}_2$ . (C) The respective fractionation lines are well distinguished in  $\text{SiO}_2$  vs  $\text{Al}_2\text{O}_3$ . Hydrous experiments reach higher maximum  $\text{Al}_2\text{O}_3$  concentrations than anhydrous liquids.

differentiation is related to high H<sub>2</sub>O-contents in the experimental liquids (Ulmer and others, 2018) resulting in much enlarged olivine stability that exclusively extracts ferrous iron. This might indeed be the reason for lower fO<sub>2</sub> in some A-type granites that are arguably derived from less hydrous systems.

Liquid CaO contents decrease systematically from 10 to 13 weight percent for basaltic compositions to < 3 weight percent for liquids exceeding 70 weight percent SiO<sub>2</sub>, without notable differences for different starting materials and different pressures (fig. 4B). This is the combined effect of plagioclase- and amphibole-dominated fractionation from andesitic to rhyolitic liquids. The dry fractionation series reach maximum CaO concentrations at slightly higher SiO<sub>2</sub> (~52 wt%). Earlier and more abundant plagioclase crystallization in the dry system probably accounts for the steeper slope compared to the hydrous liquids, especially at 0.7 GPa. Clearly the initial starting compositions influence liquid differentiation paths. As melt SiO<sub>2</sub> concentration increases CaO is systematically lower in the experimental series of Sisson and others (2005) at 0.7 GPa, independent of reducing or oxidizing conditions. Experimental data for olivine tholeiite and high MgO basalt at 0.7 and 1.0 GPa describe a clockwise trend (fig. 4C), with an increase in Al<sub>2</sub>O<sub>3</sub> between 47 and 57 weight percent SiO<sub>2</sub> up to 20 weight percent with a smooth decrease to about 13 weight percent Al<sub>2</sub>O<sub>3</sub> towards granitic liquids. Experiments on the basaltic andesite composition increase to a maximum of about 22 weight percent, mainly because of an extended crystallization of low Al websterite cumulates. Dry experiments saturate early in plagioclase and display a strong decrease in Al<sub>2</sub>O<sub>3</sub> during ongoing fractionation.

In terms of incompatible components, here represented by the enrichment of K<sub>2</sub>O and SiO<sub>2</sub> relative to the initial compositions, all liquids derived from hydrous fractional crystallization experiments display a moderate increase with respect to silica-enrichment (fig. 5). Over the entire interval from basalt to rhyolites the K<sub>2</sub>O concentration increases up to about 5 to 8 times the initial K<sub>2</sub>O content, the only exception being a K-rich starting composition from Sisson and others (2005), which is saturated in biotite over the entire crystallization interval and, thus, shows only a twofold increase in K<sub>2</sub>O over the investigated range. A similar evolution is evident for the equilibrium experiments on andesites at 0.8 to 1.2 GPa (Alonso Perez and others, 2009); this, however, is the result of the much higher melt fractions of these experiments for a given silica enrichment as the starting material is an andesite derived by fractional crystallization at 1.0 GPa (Mb series of Ulmer and others, 2018) saturated in the low-silica cumulate assemblage amphibole + cpx + Fe-Ti oxides, that is exactly at the onset of significant silica enrichment. In contrast to the hydrous series, the anhydrous experiments show more than an order of magnitude (28–40x) increase in K<sub>2</sub>O relative to the starting composition, at moderate silica-enrichment, the effect being most pronounced for the 0.7 GPa fractional crystallization experiments (Villiger and others, 2004; Villiger and others, 2007).

The highly systematic variations in melt compositions and restite phases with respect to the initial starting composition is particularly evident in *partial melting experiments* on basaltic to andesitic compositions (fig. 6). Each experimental series produces melts with distinct characteristics. Comparing different experimental series at 65 weight percent SiO<sub>2</sub>, melts from Ca or Mg# rich starting materials are higher in CaO and Mg# than melts from CaO or Mg# poor bulk compositions (figs. 6A and 6B). Most of the melts are saturated with plagioclase, amphibole and a Fe-Ti oxide and some of the highest temperature runs have clinopyroxene in addition, yet the cumulates cluster in a narrow compositional range and do not show the systematic variations observed for cumulates from fractional crystallization experiments. It is remarkable that fO<sub>2</sub> exerts a strong control on derivative granitic liquids in terms of Mg# (fig. 6A) but has a negligible influence on the CaO and Al<sub>2</sub>O<sub>3</sub> systematics of

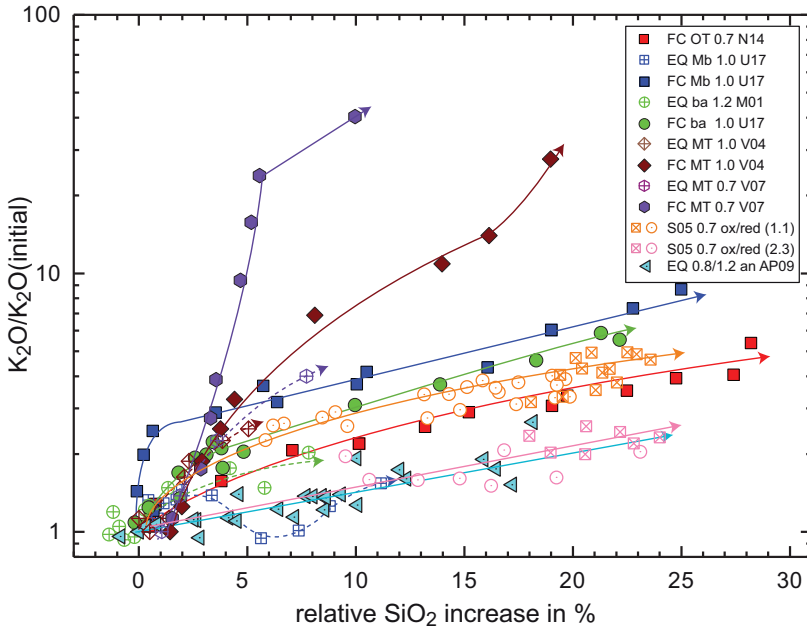


Fig. 5. Experimentally derived liquid lines of descent for fractional and equilibrium crystallization experiments illustrating the influence of the starting composition on the evolution of incompatible elements during differentiation, such as K. All data are normalized to the initial  $K_2O$  and  $SiO_2$  content. The experimental series follow near linear trends that are entirely determined by the initial  $K_2O$ , the only exception are the dry fractionation series with more than 10 times higher  $K_2O$  content in evolved compositions. The experimental series with elevated  $K_2O$  in the starting material (2.3 wt%  $K_2O$ ) shows very limited enrichment because significant amount of biotite is stable in almost all experimental runs.

derivative liquids. Although the experimental liquids closely resemble subduction zone magmas, the  $Al_2O_3$  concentrations are on the high end of some natural volcanic series from the Cascades (du Bray and others, 2006), while there is good overlap in terms of CaO and Mg#. This might be the reason why high pressure experimental studies (melting or crystallization) lead to peraluminous compositions between 61 and 62 weight percent  $SiO_2$  while the majority of natural samples are more  $SiO_2$ -rich (>65 wt%  $SiO_2$ ) when becoming peraluminous. This discrepancy between high pressure experimental studies and the volcanic rock record with respect to the formation of peraluminous granitoids has been discussed in detail by Blatter and others (2013) and imply that areas with active mafic arc volcanism do not predominantly differentiate at lower crustal pressures but show evidence for differentiation over the entire transcrustal magmatic system (for example fig. 14 of Grove and others, 2005). Therefore, some systematic discrepancies of liquid compositions produced in the lower crust and the arc volcanic record (fig. 6) such as the increased  $Al_2O_3$  content for a given silica content of the former can readily be explained by further differentiation, reaction, mingling and mixing processes during decompression in transcrustal magmatic systems.

#### DISCUSSION

##### *Silica Enrichment and Relationships to $SiO_2$ -Poor Cumulates in the Lower Crust*

A prominent result from the study of hydrous fractionation of arc-like magmas in the lower crust is the common production of cumulates that feature less than 50

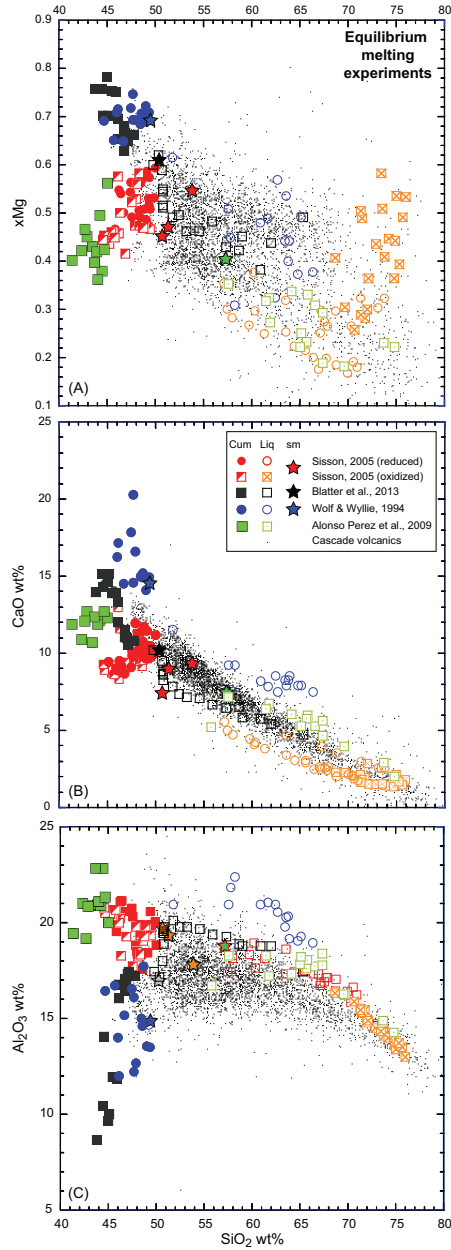


Fig. 6. Melting experiments on basalt to andesite and calculated anhydrous liquids and corresponding restites (data from Alonso Perez and others, 2009; Blatter and others, 2013; Sisson and others, 2005; Wolf and Wyllie, 1994) illustrated as  $\text{SiO}_2$  vs  $\text{Mg}\#$  (A),  $\text{CaO}$  (B) and  $\text{Al}_2\text{O}_3$  (C) in wt%. Liquids derived from partial melting experiments vary widely and define different trends related to the starting material (B and C) and/or  $f_{\text{O}_2}$  (A). Cascades data from du Bray and others (2006).

percent  $\text{SiO}_2$  (fig. 3). We illustrate this point by plotting the silica enrichment (absolute change in  $\text{SiO}_2$  concentration in wt%) of cumulates and melts relative to the starting material (fig. 7) as a function of residual melt fraction. The silica minimum of

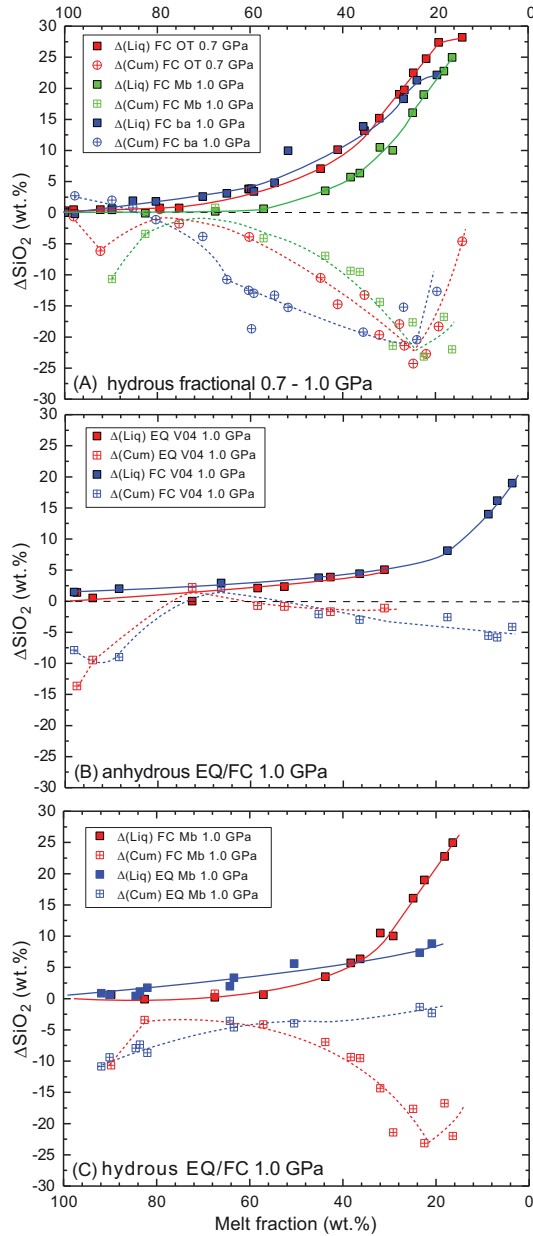


Fig. 7. Relative silica enrichment/depletion of synthesized liquids and cumulates (ISC) for fractional and equilibrium crystallization experiments, plotted against accumulated melt fraction. (A) Fractional crystallization experiments at 0.7 and 1.0 GPa. The relative silica enrichment in basaltic andesite at 1.0 GPa and olivine tholeiite at 0.7 GPa follows nearly identical trends down to melt fractions of ~30%. All 3 series form cumulates that are SiO<sub>2</sub>-depleted relative to the respective starting composition represented by the liquid composition of the previous, higher temperature experiment. This 'SiO<sub>2</sub> hole' is consistently found at melt fractions of 20–30%, suggesting that hydrous fractionation in the lower crust inevitably forms cumulates with 40–45 wt% SiO<sub>2</sub>, with liquids of andesitic composition (data from Nandedkar and others, 2014; Ulmer and others, 2018). (B) Fractional and equilibrium experiments at 1.0 GPa on an olivine tholeiite illustrating the monotonous evolution of dry magmas at lower crustal conditions in terms of silica enrichment/depletion (data sources: Villiger and others, 2007; Villiger and others, 2004). (C) Fractional and equilibrium experiments on a high-Mg basalt composition illustrating the more efficient production of silica depleted cumulates by fractional crystallization (data from Ulmer and others, 2018).

cumulate compositions in hydrous fractional crystallization experiments (fig. 7A) is represented by various proportions of amphibole-An-rich plagioclase-Fe-Ti-oxide  $\pm$  garnet  $\pm$  clinopyroxene in all 3 different starting compositions. Such amphibole-rich cumulates have a low SiO<sub>2</sub>-content irrespective of the coexisting liquid that becomes increasingly SiO<sub>2</sub>-rich. This has also been shown by Cawthorn and others (1973) for a calc-alkaline suite of Grenada. Our experiments permit to quantify the crystallization of such cumulates over a large interval of  $\sim$ 70 to 30 weight percent remaining melt fraction. The last steps characterized by increasing silica contents indicate the arrival on the feldspar-quartz cotectic and is also characterized by a decreasing slope of silica-enrichment of the coexisting liquids. The onset of SiO<sub>2</sub>-enrichment is located at different melt fractions for the various studies related to the fact that the high-Mg basalt crystallizes significant amounts of olivine and pyroxene-dominated cumulates prior to amphibole saturation (60%), whereas the basaltic andesite reaches the same paragenesis after only 35 percent of crystallization. The experimental series of Nandedkar and others (2014) is intermediate and uses an olivine tholeiite starting composition that is a derivative of the high-Mg basalt ( $-15\%$  olivine fractionation). Towards the end of the differentiation, the melt composition curves converge with 25 weight percent and less of rhyolitic liquids left relative to the initial liquid composition. Absolute enrichment differs somewhat but this is merely an expression of the SiO<sub>2</sub>-content of the initial starting material (figs. 3 and 5). The highest silica enrichment is shown by the fractionation series at 0.7 GPa, which is readily explained by the pressure dependence of the quartz – feldspar cotectic in the haplogranite system, which is shifted to higher normative SiO<sub>2</sub> at low pressure (for example, Holtz and others, 1992).

Anhydrous fractional crystallization reveals very moderate SiO<sub>2</sub> increase in the melts, in particular at 0.7 GPa (only in the last step) whereas at 1.0 GPa the SiO<sub>2</sub> increase is more pronounced and starts earlier due to delayed plag saturation (at 50 instead of 65 wt% liquid fraction) and crystallization of hercynitic spinel. Unlike hydrous fractionation experiments the SiO<sub>2</sub>-content of the complementary cumulates are only moderately SiO<sub>2</sub>-depleted (fig. 7B). Comparison of fractional and equilibrium crystallization experiments under identical conditions (*f*O<sub>2</sub> and pressure) emphasize the crucial role of fractional crystallization for silica enrichment: This is best illustrated by the experiments on the high MgO-basalt starting material (fig. 7C) where the two series evolve approximately in parallel up to a melt fraction of 40 weight percent where the equilibrium crystallization experiments start to deviate strongly not resulting in any further significant SiO<sub>2</sub>-increase. These contrasting evolution trends are controlled by the peritectic reactions involving both olivine + liquid = opx and cpx + opx + liquid = amphibole. If the latter is consuming the remaining andesitic liquid in the case of equilibrium crystallization, this will prevent any further evolution of the liquid towards true rhyolitic liquid compositions. If, however, pyroxenes are consumed by reaction with melt to form amphibole, continued crystallization of amphibole can lead to further increase in melt SiO<sub>2</sub>. The complementary cumulates remain rather silica-rich in the equilibrium case due to the controlling effects of opx (produced from olivine through the peritectic reaction). It should, however, be kept in mind that the cumulates of the equilibrium crystallization experiments always refer to the original starting composition and not to liquids of the last temperature step as this is not relevant when no fractionation is inferred. Similar relationships apply for the anhydrous experiments where the peritectic reaction ol + liq = opx suppressing significant SiO<sub>2</sub> enrichment. We conclude that if voluminous intermediate to granitic magmas are formed by crystallization differentiation at lower crustal pressures, these magmas require at least 2 to 4 weight percent H<sub>2</sub>O in their primitive parents, whereas dry or

damp magmas with less than 0.5 weight percent H<sub>2</sub>O produce negligible volumes of granitoids.

*The Role of Extraction Depth of Primitive Arc Magmas: Not Crucial*

The composition of the primitive melts from which arc cumulates are produced is highly debated and propositions range from picritic to high-Mg or high-Al basalts to high Mg# andesite and boninite (Kay and others, 1982; Grove and others, 2002; Kelemen and others, 2003). In general, primitive andesitic magmas are more abundant where the underlying crust is thin (<20 km) such as for example in the the Western Aleutian islands and the southern Izu-Bonin arc, while primitive basaltic magmas dominate in thick (>30 km) arc crust such as the eastern Aleutians and the Northern Izu-Bonin arc (for example, Kay and others, 1982; Tamura and others, 2016). For continental arcs, unequivocal primitive magmas are rare and range from olivine tholeiites to high Mg-basalts (for example, Grove and others, 2002; Hürlimann and others, 2016). Experimental studies on primary arc magmas have shown that the SiO<sub>2</sub> content increases with decreasing extraction depth (for example, Kushiro, 1975; Wood and Turner, 2009: for a review, see Ulmer, 2001), consistent with the findings from modern juvenile arcs. An important result of our fractional crystallization experiments is that the extraction depth of primary mantle-derived magmas for the case of hydrous fractionation does not exert a fundamental control on the LLD once the liquids are saturated in amphibole + plagioclase ± Fe-Ti oxides. With the exception of incompatible components such as K<sub>2</sub>O that are almost entirely controlled by the initial concentration of the primary melts and the remaining melt fraction (fig. 6), all other major element concentrations converge onto a similar LLD at 1.0 GPa (fig. 3) testifying that phase equilibria and accessory phase solubilities (for example apatite and Fe-Ti oxide on Ti and P) control the further evolution of derivative liquid compositions that form the bulk of lower to upper crustal compositions. The principal differences arise at the beginning of fractionation where different parageneses and modal proportions of ultramafic cumulates are formed: 40 to 60 weight percent of ultramafic cumulates, consisting of dunite, wehrlite and subsequently cpx dominated rocks (table EA1, <http://earth.geology.yale.edu/%7eajs/SupplementaryData/2018/Muntener>) for strongly ol-normative Mg-basalts and olivine-tholeiites that were extracted at greater depths (50–90 km, Ulmer, 1988; Hürlimann and others, 2016) as opposed to 35 to 40 weight percent opx-cpx dominated (websterite) cumulates for the basaltic-andesite that was in equilibrium with residual mantle just below the crust-mantle boundary (1.0–1.2 GPa, Baker and others, 1994; Müntener and others, 2001). The silica-poor vs. silica-rich nature of olivine and pyroxene-rich cumulates in the mantle section of the roots of island-arcs is, therefore, a potential indicator for the extraction depth of primitive magmas in arcs and/or a proxy for the thermal structure of the ultramafic parts of island arcs. Channelized extraction of primitive magmas originating at high pressure might be typical for relatively cold mantle roots, with the formation of dunite channels and generally cpx-rich cumulates. Primitive magmas recording shallow extraction depth are affected by extensive melt-rock reaction and almost continuous and complete equilibration with the surrounding mantle resulting in the formation of opx-rich pyroxenite cumulates (websterites) that are more typical for hot mantle roots beneath the arcs. This might result in different Moho temperatures as shown for some preserved juvenile arc crustal sections, such as the Kohistan and Talkeetna arcs (Ringuelette and others, 1999; Hacker and others, 2008) and to different compositions of ultramafic plutonic rocks. We elaborate on the consequences of the extraction depth below.



*Voluminous Granites from Amphibolite Melting: Comparison with Fractional Crystallization Experiments*

One of the most challenging questions faced in studies on the composition and refinement of the continental crust based on natural rock suites is related to the origins of voluminous tonalitic to granitic magmas that are exposed in oceanic and continental arcs. While it is generally accepted that strongly peraluminous tonalites to granites with crust-like isotopic compositions are formed by melting of metasedimentary protoliths (for example, White and Chappell, 1983), there is ongoing debate whether meta-aluminous to weakly peraluminous I-type granites originated by partial melting of preexisting lithologies, (for example, Clemens and others, 2011), by magma mixing and assimilation or by differentiation from basaltic parents (Bowen, 1928; Cawthorn and Brown, 1976; Sisson and others, 2005). A major objection to crystallization – differentiation is that mafic rocks in tonalitic to granodioritic batholiths such as the Sierra Nevada in the Western US are relatively rare. Although ‘downwelling’ or foundering has been invoked to explain the scarcity of mafic plutons in batholiths (Glazner, 1994) the most popular hypothesis is partial melting of fluid-absent mafic rocks (at high temperatures and pressures) to form granitoid rocks (Beard and Lofgren, 1991; Wolf and Wyllie, 1994) or to melt hydrous basaltic rocks to produce more evolved compositions (for example, Tatsumi and others, 2008) and especially Archean TTG (for example, Moyen, 2011). While we not dismiss the evidence for mixing and remelting, resorbed phenocrysts, reversed phenocryst compositions and disequilibrium assemblages in arc volcanic rocks, nor the evidence of assimilation of isotopically very different country rocks, nor that such secondary reworking processes are important at different times and places, they do not solve the fundamental problem of how silica-enriched compositions were produced in the first place. Here we restrict our discussion on partial melting experiments that closely correspond to the pressure conditions of our crystallization experiments between 0.7 and 1.0 GPa relevant for arc lower crust. We compiled partial melting experiments on amphibolite and hydrous equilibrium crystallization experiments (fig. 6, see figure caption for references). The restite compositions of all these experiments show a very restricted compositional range and trends in  $\text{SiO}_2$  vs Mg#,  $\text{Al}_2\text{O}_3$  and CaO are very different from those produced by fractional crystallization (fig. 3). One principal reason is that dehydration melting experiments on amphibolites are typically fluid absent (Beard and Lofgren, 1991; Wolf and Wyllie, 1994) thus for pure amphibolite melting the maximum bulk  $\text{H}_2\text{O}$  content available amounts to about 2 weight percent. This in contrast with numerous studies revealing that (primary?) arc magmas can have  $\text{H}_2\text{O}$  contents up to 8 weight percent (for example, Anderson, 1979; Wallace, 2005; Ruscitto and others, 2010). A second argument is that the partial melting experiments are restricted to temperatures  $<1100$  °C while our experimental cumulates extend all the way up to the liquidus of the primary magmas thereby generating compositions that are not encountered in restites of partial melting experiments. While the inferred cause for this apparent discrepancy might indeed be correct but has never been explored, there is little geological evidence for partial melting in excess of 1000 °C, and the major disadvantage of partial melting at high temperatures is the necessary heat and excess  $\text{H}_2\text{O}$  for efficient melting. Indeed, thermal models of repetitive injections of basaltic magmas into the lower crust can produce partial melts, yet this is energetically not a very efficient process (Annen and Sparks, 2002; Dufek and Bergantz, 2005) and the amount of felsic melt produced is small compared to fractional crystallization of hydrous magmas. In addition, to generate hornblende gabbro or garnet-bearing hornblendite restites the coexisting liquids require at least 4 weight percent  $\text{H}_2\text{O}$ , conditions that are not reached by partial dehydration melting experiments. On the other hand, if primary basaltic to andesitic melts from the mantle contain up to 8 weight percent  $\text{H}_2\text{O}$  they

carry their own heat and upon cooling, they crystallize  $\text{SiO}_2$  poor hornblende-rich cumulates. In essence, the question remains whether (a) there is convincing evidence from the arc lower crust that our experimentally produced cumulates have their natural analogues and (b) whether or not the experimentally produced LLD's produce derivative Si-rich liquids similar to batholiths worldwide.

*Comparison with Plutonic Rocks from Exposed Magmatic Arcs*

One of the major problems in recognizing lower crustal arc cumulates is that mafic rocks produced through fractionation-differentiation in the lower crust commonly underwent isobaric cooling thus obscuring their primary magmatic textures. Due to extensive sub-solidus re-equilibration, it is very difficult to identify and/or estimate the ratios of accumulated minerals to small or moderate amounts of trapped interstitial liquid or later modifications by percolating liquids. There is a vast literature on 'mafic granulites' (for example see review in, Rudnick and Fountain, 1995) and one common interpretation is that they are products of high grade metamorphism from near-surface mafic rocks. In juvenile arcs, there is substantial debate whether mafic rocks in the roots of island arcs are indeed igneous (for example, Ringuette and others, 1999) or formed as mafic granulites by prograde metamorphism of hornblende gabbros or amphibolites, either by post-magmatic heating and crustal thickening (Yamamoto and Yoshino, 1998; Hacker and others, 2008) or by dehydration melting (Garrido and others, 2006). From an experimental perspective dehydration melting produces restites that are similar to restites of equilibrium experiments, but they do not readily reproduce the cumulate trend generated from hydrous fractional crystallization experiments and they are also different from the plutonic rocks preserved in the Kohistan and Talkeetna lower crustal sections (fig. 8). In figure 8, we compare experimentally derived adcumulate compositions and their coexisting liquids with bulk rock compositions from the Kohistan (Pakistan) and Talkeetna (Alaska) island arcs, two of the most complete sections of juvenile island arc crust where thermobarometric estimates are similar to our experimental conditions. We restrict our discussion to the major components  $\text{CaO}$ ,  $\text{Al}_2\text{O}_3$  and  $\text{Mg\#}$ , all characterized by specific differentiation paths, z-shaped, s-shaped and inverse looped for hydrous experiments, while mostly linear for anhydrous experiments. The z-shaped trend for  $\text{SiO}_2$  vs  $\text{Mg\#}$  (figs. 8A and 8D) has previously been discussed for the southern plutonic complex of Kohistan and some of the Talkeetna rocks (Jagoutz and others, 2011): it is most instructive that the experimental cumulates closely reproduce natural rocks down to compositions with about 40 weight percent  $\text{SiO}_2$  and  $\text{Mg\#}$  of 0.45 to 0.55. There is a scarcity of calculated experimental cumulates at  $\sim 50$  weight percent  $\text{SiO}_2$  and  $\text{Mg\#}$  of 0.45 to 0.55, yet such compositions are common as garnet gabbros in lower crustal arc sections (figs. 8A and 8D). This merely indicates that natural garnet gabbros are not pure adcumulates but represent mesocumulates with small to moderate amounts (10–30 wt%) of trapped intermediate composition (andesitic) liquid, as further supported by their trace element compositions (for example, Garrido and others, 2006; Greene and others, 2006). Subtle differences are evident in the 3 experimental differentiation trends between 0.7 to 1.5 GPa, the lowest pressures producing the least pronounced z-shaped trend. Indeed, the 0.7 GPa fractionation trend might explain some of the Talkeetna data indicating that the CLD of Talkeetna has a lower pressure component, not observed (or present) in the Kohistan rocks. On the other hand the s-shaped trend in  $\text{SiO}_2$  vs  $\text{Al}_2\text{O}_3$  space expands with decreasing pressure approaching compositions of almost 30 weight percent  $\text{Al}_2\text{O}_3$  at  $\sim 42$  weight percent  $\text{SiO}_2$  at 0.7 GPa (fig. 8I). The compositions from Kohistan (fig. 8C) and Talkeetna (fig. 8F) document a similar variation in  $\text{Al}_2\text{O}_3$  vs  $\text{SiO}_2$ , but Talkeetna spreads to higher  $\text{Al}_2\text{O}_3$  consistent with slightly lower pressure of crystallization in Talkeetna. These differences in the principal depths of magmatic fractionation are further supported by counterclockwise loops

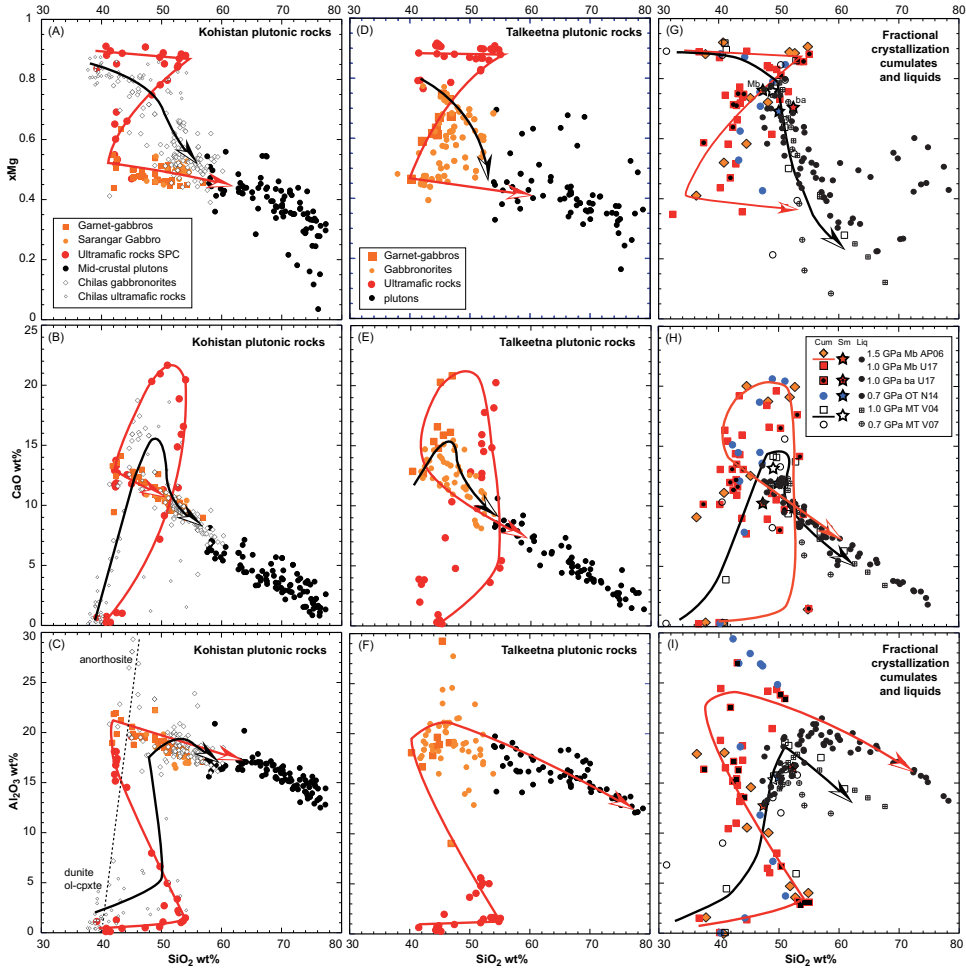


Fig. 8. Harker diagrams illustrating the bulk rock evolution of two of the most complete arc lower crustal sections (Kohistan; panels a–c; Talkeetna, panels d–f) compared to the experimental dataset (panels g–i) presented above. (A) Linear (black symbols) and Z-shaped trend in an SiO<sub>2</sub> vs Mg# diagram illustrate the dry and wet differentiation sequences of the Chilas complex and Southern Plutonic complex in Kohistan (Pakistan) (data from Jagoutz and others, 2006; Jagoutz and others, 2011) (B) Counterclockwise, wet (Southern Plutonic complex) and clockwise, dry (Chilas) loops in an SiO<sub>2</sub> vs CaO diagram illustrates the evolution of CaO in island arc crust. (C) S-shaped, wet trend (Southern Plutonic complex) and linear kink, dry (Chilas) trend in an SiO<sub>2</sub> vs Al<sub>2</sub>O<sub>3</sub> diagram, illustrating the different fractionation mechanisms in the deep crust with respect to dry versus wet Al-rich cumulates. The large variation of CaO and Al<sub>2</sub>O<sub>3</sub> between 40–45 wt% SiO<sub>2</sub> for the dry fractionation series of the Chilas complex represents dynamic sorting processes (layering between dunite - troctolite and anorthosite, (Jagoutz and others, 2006; Takahashi and others, 2007) in addition to fractional crystallization processes. (D) The z-shaped and linear trend is also expressed by the compiled data from the Talkeetna arc (from DeBari and Sleep, 1991; Greene and others, 2006; Hacker and others, 2008; Kelemen and others, 2003; Rioux and others, 2007; Rioux and others, 2010), with a predominance of the linear (dry/damp fractionation) trend. (E) Counterclockwise and inferred clockwise loops for CaO vs SiO<sub>2</sub>. (F) No obvious evolution is seen in an SiO<sub>2</sub> vs Al<sub>2</sub>O<sub>3</sub> diagram for the Talkeetna rocks.

observed in the SiO<sub>2</sub> versus CaO diagram. Experiments on the basaltic andesite primary magma generally results in cumulates with higher SiO<sub>2</sub> contents at a given CaO content (fig. 8H) due to substantially greater modal orthopyroxene. Very similar relations are found for the Talkeetna (fig. 8C) and Kohistan arcs (fig. 8B) respectively

inferring that the Talkeetna island arc might have fractionated from basaltic and basaltic andesitic primary liquids, consistent with a slightly shallower fractionation sequence than Kohistan.

In contrast, anhydrous fractional crystallization experiments display completely different fractionation paths well reproducing the rock compositions from the Chilas complex (see also Jagoutz and others, 2011) in Mg# vs SiO<sub>2</sub>, and CaO. For Al<sub>2</sub>O<sub>3</sub> vs SiO<sub>2</sub>, the Chilas rock compositions more closely follow the experimental data at 1.0 GPa, while crystallization pressures for the Chilas complex were estimated at 0.7 GPa (Jan and Howie, 1980). This difference can be reconciled by the anhydrous nature of the Villiger and others (2007) experiments, while the Chilas complex crystallized from a damp arc magma as indicated by rather An-rich plagioclase (Jagoutz and others, 2007). Increased H<sub>2</sub>O-contents decrease the plagioclase stability field relative to olivine and cpx (and opx) resulting in significantly higher Al<sub>2</sub>O<sub>3</sub> contents of the cumulates for a given SiO<sub>2</sub> content.

*Implications for Continental Crust Formation Via Arc-Magmatism: Is Relamination Required?*

Here we provide a quantitative calculation of ultramafic plutonic rocks based on the experimental record and compare them to the most complete arc crust (Kohistan arc: Jagoutz and others, 2007; Jagoutz, 2014) in figure 9. Experimentally-derived cumulate proportions are calculated for the temperature range between the liquidus and ~950 °C, where the corresponding liquid has acquired 60 weight percent SiO<sub>2</sub>. While experimentally-derived cumulates from 'wet' crystallization experiments at 0.7 and 1 GPa have a residual melt fraction between 28 and 41 weight percent, experimental cumulates of dry experiments have < 5 percent residual melt fractions. This clearly indicates that substantial proportions of derivative andesitic, dacitic and rhyolitic liquids can only be produced by mantle-derived magmas containing several weight percent H<sub>2</sub>O, while dry systems at pressures exceeding 0.7 GPa are volumetrically insignificant in terms of generating granitic compositions. Comparing natural and experimental rock columns from arcs also indicates that the 'dry' cumulates closely correspond to the rock proportions of the Chilas complex, yet the complementary hydrous fractionation series at 0.7 GPa produces too much (olivine-) pyroxenites that are not described from field studies (Jagoutz and others, 2006; Takahashi and others, 2007). The Chilas complex has been interpreted as related to intra-arc rifting and decompression melting. We conclude that the primary arc magma that produced the Chilas complex was nearly dry or damp and fractionated at ~0.7 GPa consistent with thermobarometry (Jan and Howie, 1980).

The wet fractionation series mimics the rock types of the Jijal complex with a few but important differences: (i) the modal proportions indicate significantly higher proportions of ultramafic (dunite to pyroxenite) cumulates than are observed in the field. This could be related to tectonic removal of deeper portions of the Jijal complex which at its base is in contact against the Indian plate (for example, review by Burg, 2011) and only a limited volume of the original cumulates are preserved and/or to the commonly accepted proposal that pyroxenites and garnet-rich lithologies were negatively buoyant and delaminated over geological time scales (for example, Bird, 1979; Kay and Kay, 1988; Jull and Kelemen, 2001; Jagoutz and Behn, 2013; for the discussion of the delamination potential of experimentally derived cumulates, see Müntener and Ulmer, 2006). (ii) The calculated experimental cumulates distinguish pyroxene-garnet hornblendite and garnet hornblende gabbros while in the Jijal section these two rock types are intermingled, in particular pyroxene-garnet hornblendite crops out in the ultramafic section and the garnet gabbro sections. This is important as it testifies that the Jijal complex was constructed by multiple batches of magma, with perhaps polybaric crystallization and/or variable H<sub>2</sub>O contents. It is therefore possible that assimilation, repeated partial melting and entrainment of evolved residual liquids was

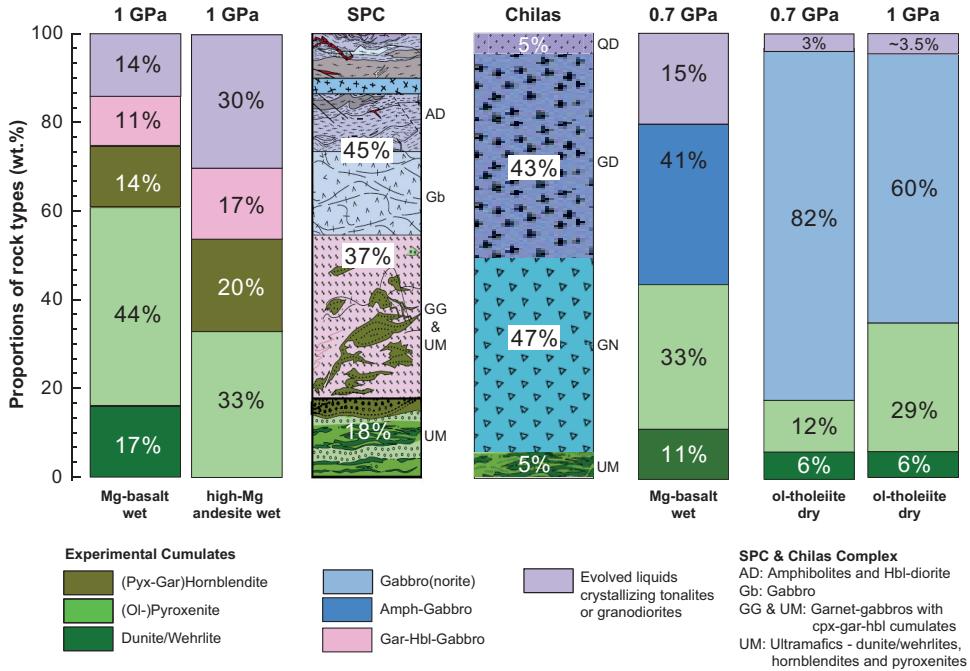


Fig. 9. Experimentally determined cumulate compositions between 0.7 and 1 GPa for moderately hydrous and dry/‘damp’ systems compared to the most complete rock record from juvenile arc crust (Kohistan arc: Jagoutz and others, 2007; Jagoutz, 2014). Experimentally derived cumulate proportions are calculated from fractional crystallization experiments listed in EA tables 1 and 2 (<http://earth.geology.yale.edu/~7eajs/SupplementaryData/2018/Muntener>) for the temperature range between the liquidus and  $\sim 900^\circ\text{C}$ , where the corresponding liquid has evolved compositions and would crystallize tonalites and granodiorites. Experimentally derived cumulates from ‘wet’ crystallization experiments at 0.7 and 1 GPa have a residual melt fraction between 14 and 30%. Experimental cumulates of nominally dry experiments have  $< 5\%$  residual melt fractions. Note that the experimentally derived ‘dry/damp’ cumulates closely correspond to the rock proportions of the Chilas complex. The wet fractionation series mimics the mineralogy of Southern Plutonic Complex (SPC) but has higher proportions of ultramafic (dunite to pyroxenite) cumulates than actually observed. Note that the natural rock association of the southern plutonic complex represents a polybaric section ranging from about 1.4 to 0.7 GPa. The difference between experimental cumulates and the southern plutonic complex might be explained by additional processes (for example, delamination and/or crystallization/interaction of ascending primary magmas with(in) the uppermost mantle). Proportions of experimental cumulates used in this figure are listed in EA table 1. Proportions of Chilas complex are quantified by intergrading the surface mapping in Jagoutz and others (2007). The Southern plutonic complex has been taken from Jagoutz (2014).

common in some places for some time in the Jijal complex, but interestingly, such open system processes do not result in a strong alteration of the CLD. Unless these cumulates can be distinguished isotopically, the CLD does not rule out open system processes. Some answers might be emerging from detailed investigations of evolved tonalites and granodiorites. Their isotopic variability or homogeneity might trace how important open system processes are in the roots of intra-oceanic arcs.

In recent years, alternative views to explain the composition of continental crust generated in arcs, collectively referred to as ‘relamination models’ have become popular (Hacker and others, 2011). Such models are mostly based on mass balancing the bulk continental crust against several subducted reservoirs to explain major and trace element concentrations and were inspired by numerical models that showed that many subducted rock types could detach from the subducting slab and ascent as diapirs to the base of the continental crust where they ‘relaminate’. In light of our

experimental data and the rather remarkable correspondence to arc cumulates as discussed in figure 8, we propose that relamination is not required to generate juvenile arc lower crust and that periodic removal of dense cumulates back into the mantle are sufficient for mass balances of the continental crust.

#### *Differences of Experimental Liquids and Transcrustal Magmatic Systems*

Discrepancies of liquid compositions produced in the lower crust and the arc volcanic record (fig. 6) can readily be explained by further differentiation, reaction and mingling and mixing processes during ascent and decompression in transcrustal magmatic systems, in particular in continental arcs, as shown by numerous studies on eruptive products of volcanic arcs (for example, Hildreth, 1983; Hildreth and Moorbath 1988). Such systems with active volcanoes on the surface crystallize on average at shallower pressures than the systems we studied here and will saturate earlier in plagioclase. This will produce liquid lines of descent that display lower  $\text{Al}_2\text{O}_3$  for a given  $\text{SiO}_2$  and that is indeed what the Cascades trend is showing (fig. 6C), relative to the experimental data discussed here. This has been explained by mixing of evolved high  $\text{SiO}_2$  magmas into recharging more primitive magmas (for example, Blatter and others, 2013). Another possibility is that our data does not cover decompression crystallization. Fractionated liquids leaving the lower crust likely will saturate in plagioclase early, and this might explain the difference in  $\text{Al}_2\text{O}_3$  between the Cascades data and our experimental liquids (fig. 6C). We speculate that the average composition of active arc volcanic systems might be slightly more mafic, perhaps related to 'mafic fingers' as inferred from geophysical data on Northeastern Japan (Tamura and others, 2002). What is required are comparisons with plutons representing a large crustal column from near surface conditions to middle to lower crustal depth and, ideally, investigated in detail as some of the well exposed arc lower crustal sections. Large batholiths such as the Sierra Nevada and Kohistan-Ladakh are dominated by tonalitic to granodioritic plutonic rocks and their average composition might be different than the average composition of active transcrustal magmatic systems, where the rock record is currently dominated by the volcanic edifice that, however, only represents 10 to 20 percent of the intermediate to acidic magmas present in the middle-upper crust forming voluminous granitoid plutonic series.

#### CONCLUSIONS

Experiments simulating fractional crystallization of hydrous arc magmas, quite remarkably, reproduce natural compositions of the roots of island arc cumulates in the pressure interval from 1.5 to 0.7 GPa, up to crystallization of about 50 percent of the initial liquid. The remainder of the liquid will crystallize similar compositions dominantly made of amphibole, An-rich plagioclase, Fe-Ti oxide followed by apatite and other accessory phases, nearly independent of the primary mantle derived liquid. This indicates that estimating the primary mantle magmas that gave rise to tonalitic to granitic plutonic rocks in batholiths is difficult to achieve and could vary from high Mg basalt to high Mg# andesite. Mantle extraction depth thus only exerts an important control on the nature of the cumulate compositions in the upper mantle and lowermost crust, but less so for the derivative arc crust where plagioclase is a dominant mineral.

While transcrustal magmatic systems show abundant evidence for open system processes such as AFC, mixing, reactive melt percolation, xenocryst transport and hybridization, this is more difficult to disentangle in exposed arc lower crustal sections. Despite the importance of open system processes for magmatic systems, it is remarkable that a simple mechanism such as fractional crystallization, more than hundred years after the original proposal by Bowen, is the most simple and most effective process in terms of energy considerations to explain the first order chemical variations of major oxides of the roots of island arcs, which are widely considered as the most important factory contributing to the net growth of Earth's continental crust.

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