

EUG XI



Theme VPP

Volcanic and Plutonic Processes and Products

EUG XI



Symposium VPP1

Thermodynamic, Structural and Physical
Properties of Melts and Element Fractionation
in Fluid-Magmatic Systems

Convenors

Matthias Gottschalk
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Sunday PM Session

VPP1 : SUPm26 : G4

The Composition of Brine Trapped in Diamonds

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Brine was detected along with silicate and carbonate minerals in micro-inclusions in cloudy regions within octahedral diamonds from the Koffiefontein mine in South Africa. The clouds are ~1 mm in size, consist of millions of sub-micrometer inclusions and commonly occupy the central zone of the diamonds. The brine micro-inclusions are closely associated with silicate minerals that may reside in neighboring inclusions or together with brine in individual inclusions. The silicate phases are either eclogitic garnet, clinopyroxene and Si-rich mica or peridotitic olivine, orthopyroxene, Cr-rich garnet and phlogopite. Inclusions carrying carbonate phases and solute-rich fluids broadly similar to those trapped in fibrous diamonds were also found.

Using IR and EPMA it is possible to constrain the major element composition of the brine. The proportions of oxides and chlorine were measured using EPMA. The relative proportions of water and carbonates were estimated from their IR absorption. We found correlation between the molar (Fe+Ca+Mg)/Cl ratio and the CO₂/(CO₂+H₂O) ratio. This correlation allows the combination of both data sets to yield the composition of the brine. When normalized to 10 Cl atoms, the average brine composition is: (K,Na)₈(Ca,Fe,Mg)₈SiO(CO₃)₃Cl₁₀(H₂O)₂₂₋₂₄. Average mass proportions are about 30-42% water, 28-33% sodium and potassium chloride, 22-25% Fe-Ca-Mg-carbonates, 4-7% of unassigned chlorine and 3-4% silica.

The silica content of the brine is much lower than that of fluids trapped in fibrous diamonds. This is not due to lower temperature of formation. Garnet-clinopyroxene thermometry of two eclogitic diamonds, olivine-garnet thermometry of a peridotitic diamond and Cr-diopside thermometry all indicate temperatures of 1000-1250°C (at 4.6 GPa, as calculated using a single Cr-diopside barometry). The low silica solubility is attributed to the 'salting out' effect of the high Cl and K in the brine. The close association of carbon-bearing brine, silicate minerals and diamonds suggests that such brines are important for diamond growth in both eclogitic and peridotitic environments. The similarity of fluids trapped in eclogitic and peridotitic diamonds and the short mantle residence time indicated by the nitrogen aggregation suggests that eclogitic and peridotitic diamonds were formed at the same time and that their host rocks reside in close proximity allowing similar fluids to percolate through both of them.

VPP1 : SUPm27 : G4

High Pressure Element Partitioning between Coexisting Supercritical Fluid and Crystalline Phases in a S-Type Granitic System

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Experiments were performed on a natural S-type granitic starting material at pressures of 20 to 45 kbar, temperatures of 675 to 900°C with 1.9 wt% to 9.9 wt% water content and a duration of 7 days. The run products of 240 experiments consist of quartz/coesite, phengite, jadeitic clinopyroxene, potassium feldspar / potassium feldspar-hydrate and the quench products of the coexisting fluid phases. Occasional accessories are garnet, epidote, titanite, rutile and apatite. The quench products of the supercritical fluids analyzed by electron microprobe have a alkalic (HT, LP) to foyaitic (HP, LT) chemistry. At low temperature and high pressure they are very hydrous. The potassium partition coefficient (calculated from atomic%) between fluid and coexisting phengite in the runs with 9.9 wt% H₂O added increases with increasing pressure and decreasing temperature from 0.5 to 3.0. The partitioning of silicon between super critical fluid and phengite generally ranges between 1.5 and 1.3. For the 45 kbar runs the partitioning coefficient clearly decreases from 1.4 at 900°C to 0.7 at

725°C. The aluminium partition coefficient decreases also with increasing pressure and decreasing temperature from 0.6 to 0.3. At 45 kbar and low temperature aluminium is too low for reliable values. Runs with lower H₂O contents added show similar element partitioning. On the basis of these results we expect that hydrous, supercritical, high-pressure, low-temperature fluids may transport large amounts of potassium from lower into higher levels of subduction zones

VPP1 : SUPm28 : G4

Fluids in the System Forsterite-Phlogopite-H₂O: Experimental Results at 6 GPa

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A series of high-pressure experiments in the KMASH-system (K₂O-MgO-Al₂O₃-SiO₂-H₂O) has been performed to constrain the compositions of potential K-rich metasomatic agents under upper mantle conditions in equilibrium with phlogopite. Two sets of experiments were conducted, one on the join phlogopite e-H₂O and a second one in the system forsterite - phlogopite e-H₂O. Forsterite and phlogopite were mixed from oxides on their respective composition and run at 0.15 GPa and 700°C in a hydrothermal vessel. The products consisted of phlogopite + glass and forsterite + talc + periclasite. These mixtures were sealed together with 15 wt% H₂O in Au-capsules. Experiments were performed at 6 GPa and 800 to 1200 °C in a Walker-type multi-anvil press. Fluids or melts were trapped in a diamond layer that was added to the experimental charge as a layer separate from the silicate phases. Traps were analysed by laser ablation - ICP - MS, and residues were inspected by micro-Raman spectroscopy.

Fluid compositions do not vary strongly with temperature and runs performed in the system forsterite-phlogopite-H₂O show similar results as runs conducted in the initially forsterite-free system. The quench-products of all fluids exhibit a composition, which is slightly more potassic and slightly poorer in silica than phlogopite; the Al-content closely matches that of phlogopite. Garnet, forsterite and phlogopite occur as residual phases in the system forsterite-phlogopite-H₂O. In runs, which initially did not contain forsterite, garnet and phlogopite form the residual phases; at temperatures in excess of 1100°C forsterite and minor amounts of enstatite were detected in addition.

The total amount of dissolved silicate was probably higher than 50 wt% under all run conditions. At the present stage of the study it cannot be answered unequivocally, whether the investigated systems behaves supercritical. However, in view of the high solubilities at low temperatures, this seems very probable.

Application of the experimental data includes modelling of K-metasomatism of upper mantle peridotite and the generation of ultrapotassic rocks and orangeites.

VPP1 : SUPm29 : G4

Element Fractionation during Fluid and Carbonate Melt Metasomatism in Suboceanic (S. Atlantic) and Subcontinental (Antarctica) Mantle

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Evidence of intense carbonate metasomatism was found in mantle material from different regions of the Globe. (Kogarko et al., 1995, Yaxley et al 1991, Ionov et al., 1993) Petrologic and geochemical study of lherzolitic and harzburgitic xenoliths of oceanic mantle from Montana Clara Volcano (Canary island archipelago) and Fernando de Naronha island (Brasil) revealed that mantle substrate of these regions has been affected by very strong carbonate metasomatism. Primary olivine, orthopyroxene, spinel and clinopyroxene are intersected by numerous veinlets of fine-grained material which replaces first generation minerals especially orthopyroxene. These fine grained zones contain second generation clinopyroxene, olivine, carbonate, glass, sulfides sometimes apatite, armalcolite, kirshtonite. The primary carbonate is Ca-rich with an

atomic Ca/(Mg+Ca+Fe+Na) ratio in the range of 0.85-0.96. The carbonate metasomatism led to wehrlitization of the primary mineral assemblage (ol, opx, sp). The wehrlitization was the result of interaction between a possibly ephemeral sodic dolomitic fluid or melt with the mantle peridotite according to the reactions: 4 MgSiO₃ + CaMg(CO₃)₂ = 2 Mg₂SiO₄ + CaMgSi₂O₆ + 2CO₂ 3CaMg(CO₃)₂ + CaMgSi₂O₆ = 4CaCO₃ + 2 Mg₂SiO₄ + CO₂. These results support a two stage model of Ca-rich carbonatite formation: 1st stage - metasomatic wehrlitization and carbonatization of mantle rocks; 2nd stage - partial melting of the carbonate-bearing wehrlitic substrate, resulting in generation of calcio-carbonatites. Petrological and geochemical evidence suggests that lithospheric mantle of Eastern Antarctic has been affected by very intense carbonate metasomatism resulting in the development of such minerals as Sr-rich apatite, Nb and REE-rich perovskite, Ba and Ti-rich mica, sulfides, carbonates and clinopyroxenes of second generation. The trace element signature of investigated mantle xenoliths and mineralogy of metasomatic zones indicate that metasomatized fluid contained S, Sr, P, Ba, LREE, Ti, Nb, Th, U, Zr and F. Experimental data imply that carbonate fluid is usually enriched by such elements as Ba, Sr, P, LREE but not Ti and other high field strength elements. The solubilities of these elements substantially increase in alkaline silicate melts. On the basis of obtained data we can conclude that the dominant metasomatic agent of investigated mantle material is highly alkaline silicate-carbonate melt or fluid.

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Ionov DA, Dupuy C, O'Reilly SY, Kopylova MG & Genshaft YS, *Earth Plan. Sci. Lett.* **119**, 283-297, (1993).

VPP1 : SUPm32 : G4

Bromine Behaviour in Synthetic and Natural Silicic Melts

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Bromine is one of volatile constituents produced by volcanoes which possibly interacts with the atmosphere. Bromine is injected into stratosphere during large volcanic eruptions of H₂O-rich silicic magmas because of its ability to partition into a H₂O-rich vapor phase and probably plays a key role in the stratospheric ozone depletion cycles (H. Bureau, H. Keppler and N. Métrich, EPSL 183, 51-60). In order to assess the effect of the melt composition on Br concentrations in magmas, we have investigated Br partitioning between bromide-rich aqueous fluids and hydrous iron-free silicic melts with variable Na+K/Al and Si/Al molar ratios (albite, haplogranite, rhyolite and pantellerite). The experiments were performed in TZM - rapid quench autoclaves (Bayerisches Geoinstitut, Bayreuth) over a range of pressure: 100, 150, 200 MPa, and temperature: 900, 1000, 1100°C. For each experiment, glass powder from one composition and an aqueous fluid containing NaBr in excess were equilibrated in Pt capsules. A series of natural volcanic glass samples and melt inclusions hosted in quartz and leucite were analysed together with the synthetic glasses by PIXE (Proton Induced X-rays Emission) with a nuclear microprobe (LPS- Saclay). The Br concentrations range from 5360 to 7850 ppm for albite, from 2800 to 3900 ppm for haplogranite, from 4200 to 5900 ppm for rhyolite, and from 9745 to 11250 ppm for pantellerite. Bromine concentrations are negatively correlated with pressure in H₂O-saturated silicic melts. They increase in peralkaline and peraluminous rhyolitic compositions and achieve a minimal value for (Na+K)/Al molar ratio close to one. Bromine behaves similarly to chlorine for these compositions. In natural obsidian samples and MI hosted in quartz and leucite which have been investigated, the Br content varies from < 2 ppm to 28 ppm, with the highest concentrations in pantelleritic melts (Na+K/Al=2). Br behaves as an highly incompatible and hydro-magmaphile element that implies that magmas are far to be saturated with respect to Br prior to eruption. Br behaviour during magma crystallisation is controlled by its partitioning into the H₂O-rich fluid phase when occurs. On the contrary, its potential high solubility in silicate melts makes it a very sensitive chemical tracer of magma contamination by external sea water and Br-rich material.

VPP1 : SUPm33 : G4

Boron and Carbon in Highly Evolved Magmatic Systems: Accumulation in Residues and Fractionation between Immiscible Melts and FluidsIlya V. Veksler (veksler@gfz-potsdam.de)¹,
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Boron and carbon are neighbours in the periodic table and they share similar fate during magma evolution. With the exception of mantle-derived carbonate and kimberlite melts, primary magmas contain very small amounts of B and C. Throughout initial stages of fractional crystallisation both of them are incompatible. Thus, provided special conditions are met, concentrations of B₂O₃ and CO₂ in residual systems may reach weight percent levels (e.g., CO₂ in secondary carbonates and B₂O₃ in granitic pegmatites). Here the role of B and C becomes crucial for the whole trend of magma evolution. Three main scenarios are: (1) stabilisation of liquidus B₂O₃- and CO₂-rich minerals; (2) separation of immiscible melts, or (3) fractionation to low-density H₂O-CO₂ fluids. At this stage different capacity of B and C for polymerization with aluminosilicate network comes into play and the further development becomes increasingly complex. However different secondary carbonates and granitic pegmatites appear to be, their interpretation poses similar problems and require similar approaches.

Bulk analyses of highly evolved rocks may not reflect the abundances and the role of B and C in parental melts and fluids. Hence the significance of constraints that come from experimental petrology and melt and fluid inclusion studies. Without them it is hardly possible to distinguish between liquidus crystallisation, metasomatic reactions and liquid immiscibility.

Recent studies of synthetic systems and natural inclusions seem to change conventional views on the genesis of natrocarbonatite lavas. Following Bailey (1993) we suggest that natrocarbonatite eruptions at the Oldoinyo Lengai volcano (Tanzania) are likely to result from fluid fractionation rather than liquid immiscibility (Nielsen et al., 1997). This conclusion is based on the analysis of experimental data on two-melt (Hamilton et al., 1989; Jones et al., 1995; Veksler et al., 1998) and fluid-melt (Veksler & Keppler, 2000) element partitioning, other experimental constraints (e.g., thermal divides) and melt inclusion studies.

In granitic pegmatites we find evidence for generation of B-rich melts at the final stages of magma evolution. This process is recorded in melt inclusions and was reproduced experimentally with synthetic compositions. On-going studies are aimed on sorting out the importance of liquid immiscibility and fractional crystallisation for the record-high boron enrichment.

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VPP1 : SUPm34 : G4

Neodymium and Strontium Decoupling in the Infiltration Process: Implication for Sr-Nd Isotope Systematic of Metasomatism Related RocksValeri Savatenkov (lev@ad.iggp.ras.spb.ru) &
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The Sr-Nd isotope systematic in Archean gneisses (2.7 Ga) in fenitization zone of Devonian (370 Ma) ultrabasic-alkaline intrusion Ozernaya Varaka (Kola peninsula, Russia) was studied as an example of natural geological model illustrating material exchange between two components: the alkaline magmatic melt bearing the isotope signature of depleted mantle ($\epsilon\text{Nd}_{(370)} = +3.4$, $^{87}\text{Sr}/^{86}\text{Sr}_{(370)} = 0.7034$) and

the country gneisses of granitic composition with Archean crust isotope characteristic ($\epsilon\text{Nd}_{(370)} = -33.3$ - -29.4 , $^{87}\text{Sr}/^{86}\text{Sr}_{(370)} = 0.7322$ - 0.7210). Metasomatic influence of the intrusion on the gneisses was associated with CO₂ enriched fluid, that play role of transport for chemical and isotope exchange between the intrusion and the gneisses. The studied gneisses were sampled on the different distances from the contact and show different intensity of metasomatic reworking.

The shift of the Sr isotope composition comparatively to the Nd one in the metasomatically changed rocks from the initial composition depends on the abundance of Nd and Sr in two components (intrusion and gneisses) and on the mobility of those elements in the fluid flow. On the short distance from contact with the intrusion the extensively metasomatized gneisses maximally response to chemical equilibrium with fluid. The figurative points for those gneisses on the $\epsilon\text{Nd}_{(370)}$ - $^{87}\text{Sr}/^{86}\text{Sr}_{(370)}$ diagram closely fit to the bulk mixing line 'intrusion-gneisses'. For the gneisses more distant away the contact the figurative points deviate significantly from the bulk mixing line. Contribution of the Sr increment from intrusion into the gneisses decreases with increasing of distance more strongly than Nd increment decreases. The specifics of behavior of Rb-Sr and Sm-Nd isotope systems in the metasomatic process can be considered in terms of 2-D model of advective isotopes transport in fluid flow (Bickle, 1992): infiltration transport through aquifers with transverse diffusion into wall rock. According to this model metasomatic contamination trend for the gneisses on the $\epsilon\text{Nd}_{(370)}$ - $^{87}\text{Sr}/^{86}\text{Sr}_{(370)}$ diagram is determined by difference in kinetic dispersion of isotope fronts for Nd and Sr. Extent of isotope front dispersion is characterized by Damkohler number ND: dispersion of the front increases with decreasing of ND. In turn Damkohler number directly depends on diffusion coefficient of an isotope in pore space. Character of Rb-Sr and Sm-Nd isotope systems behavior on the mineral scale in the studied gneisses reveals more fast diffusion of strontium along grain boundaries than neodymium diffusion. Thus Sr transported into in fluid flow from intrusion more rapidly attains isotope equilibrium with wall rocks and form more sharp isotope front in comparison to Nd. On the other hand neodymium migrating in fluid flow from intrusion through aquifers owing to high kinetic dispersion is able to spread on the more long distance without complete isotope equilibration with wall rocks.

VPP1 : SUPm35 : G4

Orthomagmatic Copper Origin in Zaldivar Cu-Porphry Deposit, ChileEduardo Campos (came@geo.vu.nl),
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Remnants of melt inclusions (MI) occur in igneous quartz phenocrysts from the Llamo porphyry unit, Zaldivar deposit northern Chile. They are rare, found on clean areas of the host, either isolated or in clusters, small (5 to 30 μm), rounded to ellipsoidal, and filled with a partly or fully micro-crystalline aggregate, with no visible gaseous or liquid phase at room temperature. This type of inclusions, which do not show any obvious relation with the numerous fluid inclusions found in the same phenocrysts, is identified by its behaviour upon high temperature heating.

Melting of micro-crystals begins around 800°C, with a sudden appearance of a large gas bubble (about 20% of the total inclusion volume) and some liquid. This provokes distinct movements of the remaining solids, which disappear progressively upon further heating. Total homogenization, always by bubble disappearance, occurs between 950° and 1050°C. In one type of inclusion (M1), micron-size black dots remain in the homogeneous melt. Some disappear around 1100°C, others shows no size change even after heating to 1100°C for 30 min. Quenching results in the formation of a clear semi-transparent glass with several micron-size black dots randomly distributed in the inclusion, which show no changes in size or shape upon cooling to room temperature. This makes the differences with another type (M2), in which no black dot were observed during heating.

Composition of the quenched glasses in both types show a high-silica content (SiO₂:75.43; Al₂O₃:13.99; Na₂O:1.46; K₂O:6.71; CaO:0.59) and a well-defined magmatic differentiation series, corresponding to a rhyolite-granite trend. The same range of rock composition and evolution is found in the regional magmatism.

EPMA analyses on M1 and M2 glasses, made in areas free of black dots, indicate high copper concentration (0.03 to 0.57 wt% with a mean value of 0.10 wt%), at least one order of magnitude higher than what should be expected for calc-alkaline plutons in general. Bulk analyses including black dots give extremely high Cu-content, up to 1.23%. The black dots have been found by EDS and SEM analysis to consist of Cu-chloride.

In conclusion, copper was already present in the magma at the origin of the porphyry copper system (orthomagmatic model). It separated by magmatic immiscibility at an early stage of the magma crystallization in the form of chlorine solutions, not sulfides as commonly assumed.

VPP1 : SUPm36 : G4

Exsolution of Salt-Rich Melts: Observations in Highly Fractionated Melts from Hornblende CumulatesAxel D. Renno (axel.renno@mineral.tu-freiberg.de),
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TUBAF seamount is located south of Lihir island (north-east of Papua New Guinea). During cruise SO-133 with the research vessel R/V Sonne, numerous xenoliths (mantle peridotites, gabbros, basalts and cumulates) were sampled by TV-guided grab (Herzig et al. 1998). Host rock of these xenoliths is a Quaternary trachybasalt. One sample - a hornblende cumulate - shows manifold exsolution phenomena of melts. The hornblende cumulate is composed of about 92 modal-% of irregularly zoned magnesiohastingsite, which carries magmatic inclusions of phlogopite and magnetite. The mesocumulate is mainly composed of glass. Only the mineral apatite crystallized from this highly fractionated melt. We found four different types of exsolution phenomena with the probable chronology: - exsolution of Cu-Fe-rich sulfide melt - exsolution of a second silicate melt - exsolution of a salt melt in the system Cu-Cl-H₂O - exsolution of a gas phase. The glass shows no signs of hydrothermal alteration. Exsolved Cu- and Fe-rich melt drops have shapes typical for separated sulfidic melts. Some of them are deformed by the movement of the amphibols in the cumulate. The sulfidic melt crystallized as an isocubanite solid solution proving a temperature of formation of > 557°C (Craig & Scott, 1974). The isocubanite shows exsolution lamellae, which all belong to the compositional field of isocubanite solid solution. Additional minerals from the systems Cu-Fe-S or Fe-S were not found. Intense greenish-blue coloured droplet-shaped objects with a diameter of up to 2 mm were identified as mineral phase in the system Cu-Cl-H₂O (e.g. atacamite). The observed microstructures (e.g., intergrowth with apatite crystallized from the melt) and the complete absence of hydrothermal alteration support the hypothesis, that these copper-minerals are the products of an exsolution of a salt melt from the highly fractionated silicate magma. Due to the very special conditions of formation, most of these very salt rich melts (solutions) were not able to escape from the system and remained as large melt-inclusions in the glass. The freezing of this state was made possible by the rapid uplift of the hornblende cumulate xenolith to the seafloor by a later pulse of trachybasalt. We exclude a formation as a pseudomorph after Cu-Fe-sulfides. Electron microprobe investigations of the exsolved salts showed no signs of other metals (e.g., Zn or Fe). Detailed examinations of the silicate melt and gas exsolutions together with a study of stable isotopes and of different trace elements in the glasses as well as dating of the amphibole is in progress. The shape, the state of preservation and the deformation of the exsolved melt phases show, that the formation of the four types of exsolution was nearly contemporaneous. The rapid freezing allows a very rare insight into magmatic unmixing processes, which may lead to the formation of ore deposits. The ongoing studies will allow us to quantify the fractionation processes, particularly of the elements Cu, Fe and Cl in mafic magmatic systems.

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VPP1 : SUPm37 : G4
Diamond Formation during Source
Carbonation- Unusual Inclusion Parageneses
in Diamonds from Namibia

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Metasomatic processes occurring in the mantle have significant consequences for the depth and temperature of magma formation as well as for their composition. Studies of mineral inclusions trapped in diamonds can provide information on the composition of melts and fluids present in the mantle. In Namibian diamonds, unusual mineral compositions and associations observed in nine out of the 109 diamonds studied suggest that either a source rock was sampled which has not been recognised before, or that melt-rock interactions occurred during diamond crystallization. Typical for this new paragenesis are intimate lamellar intergrowths of clinopyroxene and orthopyroxene. The Cpx has compositions ranging from eclogitic and lherzolitic (mg#: 76-94; Al₂O₃: 1.4-6.7 wt%; CaO: 11.5-20.2 wt%, Cr₂O₃: 0.1-3.2 wt%, and Na: 1.2-5.2 wt%). In two diamonds the cpx inclusions (mg# of 89 and 85) have a small kosmochlor component (NaCr). In one diamond MgCO₃ is enclosed by cpx. Other minerals form touching inclusions with the pyroxenes: a SiO₂ phase is observed in three diamonds and two diamonds contain pyroxenes of lherzolitic composition together with CaCO₃ in one case accompanied by SiO₂, Phlogopite and a Cr-rich titanate (together with lherzolitic garnet) are attached to pyroxenes in two other diamonds.

These mineral inclusions may be witnesses of a multi-stage history occurring in part after diamond formation: (1) Touching inclusions can re-equilibrate after their trapping in diamonds and thus exsolution of opx from cpx (and vice versa) could have happened during cooling after diamond crystallization. (2) Phlogopite and Cr-rich titanates are stable in the diamond stability field but they are frequently interpreted as epigenetic minerals in diamonds. However careful examination before breakage did not reveal any cracks in the case of Namibian diamonds. Therefore we favour the hypothesis that the unusual textural relationships shared by the pyroxenes and other minerals result from a metasomatic event during diamond crystallization. The bleb of magnesite inside clinopyroxene may result from the interaction between opx and a dolomitic melt. Extreme carbonation of peridotite leads to the crystallization of SiO₂. Variations in the Mg/Ca ratio in relation to variations in the pressure and/or temperature conditions during the interactions would explain (1) that orthopyroxene was not completely consumed and (2) that either magnesite or aragonite crystallized. This melt could also have been rich in Na₂O, K₂O and H₂O. Such carbonate-rich melt may originate during incipient melting of peridotite or during slab melting and be responsible for the formation of lherzolite from previously depleted harzburgite within the cratonic lithosphere.

VPP1 : SUPm38 : G4
Trace Element Distribution in Melilite-Bearing
and Melilite-Free Series- Derivatives from
Ca-Rich and Ca-Poor Mantle
Alkaline-Ultrabasic Melts

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Rare element contents and compositions in carbonatites are essentially different dependent on their association with ring alkaline-ultrabasic complexes, containing or not containing melilite-bearing rocks (Rass, 1998). This may be explained with different trace-element fractionation features during differentiation of two alkaline-ultrabasic primary magmas - Ca-rich and Ca-poor, melted at different levels, more or less respectively (Kravchenko, Rass, 1985; Kravchenko et al., 1992). The distribution of trace elements, REE, and P in melilite-bearing and melilite-free rocks of alkaline-ultrabasic massifs Kugda, Odikhincha and

Kara-Meni (Maimecha-Kotui province, NW Siberian Platform) was investigated. The principally different Sr-, REE-, Zr-, and P-contents in the above rocks, comparable in differentiation degree, are found out.

Sr contents in the rocks of melilite-bearing, high in Ca, series is an order of magnitude higher than those in melilite-free series. Sr partition coefficient melilite/melt is near 1 (Kuehner et al., 1989), and just the availability of melilite as a rock-forming mineral in all sequential derivatives controls the Sr contents and zoning in other minerals. Rims of apatite grains in melilite-free rocks are impoverished in Sr, since apatite is the mineral, mainly concentrating this element; perovskite from melilite-free rocks is also enriched in Sr than perovskite from melilite-bearing rocks (Rass, Laputina, 1996).

Based on the analysis of La/Sm logarithmic relations in sequential Derivatives, their fractionation during differentiation of parentmagmas for the two series - Ca-rich and Ca-poor - was established to be in accordance with Rayleigh model, with different parameters. The data points for rocks with or without melilite are approximated by the following linear equations: lgSm=1,40lgLa+0,053 and lgSm=0,70lgLa+0,43 (Kugda); lgSm=1,40lgLa+0,053 and lgSm=0,675lgLa+0,40 (Odikhincha); lgSm=0,924lgLa+0,07 and lgSm=0,711lgLa+0,51 (Kara-Meni). Clinopyroxenes from melilite-bearing rocks in any massif are enriched by REE, as compared with clinopyroxenes from melilite-free rocks in the same massif. When taken into account the REE concentrations in melilites themselves are of the similar magnitude, it may inferred the parental magma for melilite-bearing serie is enriched by REE.

Ulvospinel-contents in investigated magnetites are usually lower than 30%, but magnetites in Kugda pyroxenites contain near 50% of Fe₂TiO₄, that may show the higher crystallization temperature (as compared with investigated magnetites from the other rocks - from jacupirangite to syenites and from kugdite to mica okaite - in the three massifs). Magnesioferrite-contents in magnetites from melilite-bearing series are higher than that from melilite-free series. That is the indicator of the higher oxygen fugacity (Speidel, 1970) during Ca-rich derivative crystallizations. Since melilite stability field is bounded with 14 kbar, the necessary condition for possible differentiation of Ca-rich primary magma, parent for melilite-bearing serie is the existence of an intermediate magma chamber, whereas Ca-poor primary magma fractionation could take place during magma uplift.

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Sunday PO Session

VPP1 : SUPo01 : PO
Volcanites of the Ichetju Diamind Area
(Middle Timan, Russia)

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In recent years, new data on lamproite-like volcanites in the Middle Timan have revived a lively discussion. Taking into account, that there are diamond localities in the Middle Timan and the original source of diamonds is still unknown, presence or absence of such geological objects as lamproites is of great importance. The geological structure of the Middle Timan consists of late Precambrian sedimentary-metamorphic rocks forming the folded basement, and Paleozoic sedimentary and volcanic rock (sandstones, argillites, tuffs and basalts) forming the platform cover. Magmatic rocks are not wide spread in the Middle Timan, being represented by the middle timan gabbro-diorite complex and chertass of the alkaline-ultrabasic complex. Petrographic study of basalts from the Ichetju diamond area revealed the following rock-forming minerals: andesine and pyroxene, olivine, fragments of brown volcanic glass and accessory minerals (titanomagnetite, magnetite, hematite). As impregnations, large crystals of labrador predominate, andesine and pyroxene phenocrysts are rare. Volcanic glass is replaced by calcite, chlorite and leucocoxene. The structure of the rocks is amygdaloidal, intersertal. Chemical analysis allowed us to determine petrochemical features of the rocks. The first group of the rocks includes normal basalts with the typical alkali content (Na₂O-1.9%, K₂O/Na₂O=0.07) and medium alumina content (Al₂O₃ about 13.5%). Another group is represented by alkaline volcanites - light bluish-grey dense rocks with zonal black microaoutoliths about 0.5-2 mm in diameter. Potassium content is quite high (K₂O-8.54%, Na₂O-0.043, K₂O/Na₂O=199). By SiO₂ content these rocks are attributed to ultrabasic rocks or to basic rocks with a low SiO₂ content. On petrochemical diagram, part of points is located in the field of alkaline basic rocks associating with diamond-bearing rocks of Siberia. Another group of points is located in the field of poorly studied rocks, that makes their diagnostics difficult. Two samples are in the field of alkaline basalts with accessory diamonds. Thus, our research of volcanites from the Ichetju diamond area allows the region of peralpotassium basalts occurrence to be considered as a perspective diamond area.

VPP1 : SUPo02 : PO
Pre-Eruptive Storage Conditions of the
Highly Differentiated Phonolitic Laacher See
Magma (East Eifel, Germany)

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Experimental studies on phase equilibria in erupted magmas have greatly improved the knowledge about the P, T and X(H₂O) conditions just prior to eruption. (Rutherford et al., 1985; Rutherford and Devine, 1996). To investigate storage conditions that prevailed in the Laacher See magma reservoir, hydrothermal experiments have been performed on a natural pumice sample from LLST (Lower Laacher See Tephra, sub-unit IV). Phase stabilities and compositions of natural phenocrysts and glasses were compared with those produced during the experiments to infer the most likely P-T conditions of the roofward parts of the Laacher See magma reservoir. Experiments were performed at pressures between 50 and 150 MPa and temperatures between 725 and 800°C. The most important phase transition is the breakdown of sanidine and the appearance of amphibole, because sanidine and amphibole are the dominant phenocryst phases in the LLST. Sanidine is stable at water pressures less than ca. 140 MPa and temperatures less than ca. 775°C. Amphiboles are stable at pressures higher than ca. 60 MPa and temperatures less than 770°C. Melt only exists at pressures higher than 60 MPa and temperatures higher than ca. 765°C. This clearly shows, that the magmatic temperature in the cupola region was lower than 765°C. Our study suggests that the highly differentiated Laacher See magma last equilibrated at temperatures between 750 and 760°C and water pressures between 90 and 120 MPa. This implies that the

magma would have contained about 6 wt% H₂O. Glass inclusions in various phenocrysts contain up to 5.7(±0.4) wt% H₂O (Harms and Schmincke, 2000), confirming our pressure estimate. We suggest that the magma was close to water saturated at depth, which implies that the uppermost levels of the Laacher See magma reservoir was at a depth of about 4 to 5 km.

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VPP1 : SUPo03 : PO Diffusion of Cations in Hydrous Melts of Rhyolitic to Andesitic Compositions

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Diffusion of elements in molten silicate systems is a fundamental process during crystal growth and dissolution, magma mixing and interaction of ascending magmas with their neighbour rocks. Of particular interest are systematics and differences in diffusion behaviours of the elements. This is a first step for a better understanding of transport mechanisms and the relation between diffusion and melt viscosity. To quantify the enhancing effect of water on diffusion we investigated anhydrous and hydrous systems. We determined diffusion coefficients of trace elements like Rb, Sr, Ba, Cu, Co, Ni, Cr, La, Ce, Nd, Sm, Eu, Gd, Er, Y, V, Zr, Nb, Hf, Ta, Sc, Ge, Sn and major components SiO₂ and Al₂O₃. Experiments were performed with rhyolitic to andesitic melts at 800-1400°C and 500-1000 MPa. For rhyolitic compositions we tested a modified diffusion couple technique, where three glass cylinders are coupled together to study the diffusion of trace and major elements independently in a single experiment under identical conditions. The glass cylinder in the middle is of rhyolitic composition, the one on bottom is doped with trace elements and the one on top is enriched with SiO₂. The concentration profiles of major elements were determined by electron microprobe and profiles for trace elements by synchrotron radiation x-ray fluorescence microanalysis (SYXRF) and secondary ion mass spectrometry (SIMS). Water distribution along the diffusion profiles was analysed with infrared microscopy. At the same temperature and water content, the tracer diffusion coefficients for rhyolitic melts are generally lower than for andesitic melts (e.g., by 0.2 log units for Rb and Ba and 1.0 log units for Zr and Nb at 1200°C for a melt containing 5 wt % water). Diffusion coefficients decrease systematically from the LFSE to the HFSE for each melt, e.g., for rhyolitic melts with 2 wt % water diffusivities decrease from log D_{Rb} = -11.09 over log D_{Ba} = -12.16 and log D_{Zr} = -13.24 to log D_{Nb} = -14.16 (D in m²/s) at 1100°C, 500 MPa. At the same conditions, diffusivities are systematically higher for a melt containing 5 wt % water (log D_{Rb} = -10.87, log D_{Ba} = -11.34, log D_{Zr} = -13.15 and log D_{Nb} = -12.86). Tracer diffusivities for the HFSE are similar to chemical diffusivities of the major elements Si and Al. Diffusivities of the REE (La-Yb) are almost identical except of Eu, which is mobiler than the others especially in rhyolitic melts. This difference compared to its neighbour elements Sm and Gd decreases with increasing temperature in rhyolite.

VPP1 : SUPo04 : PO Diffusion of CO₂ in Silicate Melts: The Influence of Bulk Composition

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Experimental studies on the mobility of volatiles in magmatic systems can provide important constraints on degassing and the volatile speciation. However, there are only few data on CO₂ diffusion, although CO₂ may effectively control the degassing of magmas. During the ascent of hydrous magma some water may partition into the CO₂ gas phase even at depth where water is nominally undersaturated.

To investigate the diffusion mechanism of bulk CO₂ we performed a systematic study on CO₂ diffusion in a range of iron free compositions. The compositions studied were (in the order of decreasing polymerization) haplo-rhyolite, haplo-dacite, haplo-andesite, haplo-tholeiite to haplo-hawaiite. Furthermore we investigated the system NaAlSi₃O₈ + n Na₂O (n=0-7, in wt%). With these synthetic glasses we performed diffusion couple experiments in an internally heated pressure vessel with argon gas pressure up to P_{MAX}=0.5 GPa and temperatures up to T_{MAX}=1623 K.

Evaluation of the one dimensional diffusion profiles with micro IR spectroscopy shows a distinct dependence of the bulk CO₂ diffusion coefficient on melt composition. In the simple natural system we observe a decrease in the diffusivity of CO₂ in haplo-rhyolitic to haplo-andesitic melt from log D_{CO2} = -11.08 to -11.55. The degree of polymerization changes from NBO/T = 0.05 to 0.37. With increasing depolymerization of the melt up to NBO/T = 0.76 (haplo-hawaiite) the diffusivity increases up to log D_{CO2} = -11.16. This dependence of the CO₂ diffusion on the melt composition can be explained by two different diffusion mechanisms: In fully polymerized rhyolitic melt all CO₂ is dissolved and transported as the neutral molecular CO₂. In highly depolymerized melts like haplo-tholeiite and haplo-hawaiite, CO₂²⁻ is the diffusing species which is suggested to be coupled to the network. The minimum CO₂ diffusivity in melt containing both, molecular CO₂ and CO₃²⁻ can be explained by diffusion of molecular CO₂ and fixation of molecular CO₂ in the network by the interconversion reaction: CO₂ + O²⁻ = CO₃²⁻ which decreases the diffusivity of bulk CO₂.

In the case of aluminosilicate melts we observe an increase in diffusivity of CO₂ with progressive depolymerization. NBO/T increases from 0 to 0.16 by adding up to 7 wt% Na₂O to the albite melt. This is consistent with the trend observed in the more complex system, if one considers that the speciation of CO₂ in albite is comparable to that in haplo-andesite. With continuous depolymerization of this composition the diffusivity trend of CO₂ is similar to that in aluminosilicate melts.

VPP1 : SUPo05 : PO Raman and Microthermometric Studies of Fluid and Melt Inclusions in Magmatic Xenoliths from the Sabatini Volcanic District (Roman Comagmatic Province, Italy): Evidence for Sulphate-Rich Melts

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Study xenoliths are high silica magmatic rocks collected from three distinct hydromagmatic units from the Sabatini Volcanic District (Roman Comagmatic Province, northern Latium, Italy). They consist of 60-70% of sanidine with clinopyroxene, mica, feldspatoids (cancrinite (S-rich variety), hauyne, nepheline), melanitic garnet, plagioclase, plus accessory phases such as Fe-oxides, S-rich apatite, titanite and rare fluorite. A complex association of melt and fluid inclusions is particularly evident in K-feldspar. Primary silicate-melt inclusions (10-100 μ) consist of colourless transparent glass, one or more H₂O bubbles

±daughter phases. In many inclusions the silicate glass (M1) contains several small globules (1-5 μ) of an immiscible yellowish phase (M2), that might be birefringent at crossed light. The relative M1 - M2 ratios vary in the different inclusions. Raman spectroscopy in the silicate glass (M1) reveals that Sulphur is present in the glass mainly as S⁶⁺ (SO₄²⁻; 995 cm⁻¹) and further indicates that glass is rich in CO₃²⁻ (1075 cm⁻¹). Spectra in the immiscible M2 phase reveals only the presence of SO₄²⁻ vibrations, with minor CO₃²⁻, and allow the identification of M2 phase as an original sulphate melt. Raman investigations in fluid inclusions indicate that liquid water contains high concentrations of HSO₄⁻ and SO₄²⁻ ions in solution (peaks at 890 and 980 cm⁻¹). High sulphate concentration in the fluids is substantiated by the identification of cesanite and anhydrite as daughter minerals. Carbonates (mainly calcite) were also identified. Fluid inclusions occur as clusters associated to melt inclusions or as short intragranular trails. They may contain one or two high birefringent daughter minerals. Low density CO₂ has been detected in a few vapour bubbles. Preliminary data of final melting temperatures up to +28°C confirm the presence of SO₄²⁻ in the solution; homogenisation temperatures occur to the vapour phase (Th L+V > V) between 365 and 410°C. We consider these data as evidence that study xenoliths crystallised from a sulphate-rich magma, that at late stage exsolved immiscible sulphate-rich melts and aqueous fluids. High-sulphur fugacities during crystallisation are supported by the presence of hauyne, cancrinite and SO₃-rich apatite in the mineral assemblage. Since most of the sulphur is present as S⁶⁺ these results further indicate very oxidising conditions during crystallisation. The initial volatile saturation of a partially crystallised magma resulted in the exsolution of a free sulphate-rich aqueous fluid that may have migrated and interacted with the wall rocks providing a highly mobile metasomatic agent.

VPP1 : SUPo06 : PO In-Situ XAS Study of the Effect of Water on Local Structure Around Nickel in Silicate Melts

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The study of water and transition metals in silicate melts is important to understand the explosive character of subduction fields volcanoes, and the occurrence of metallic layers in relation to this type of volcanism.

XAS spectroscopy constitutes a method of choice to probe structural environment around given atomic species. In this study, we were concerned with the effect of water on the local environment of nickel in silicate materials. We will present results obtained at different temperatures (between 25 to 1100°C) on crystalline model compounds, and on various glasses containing from 1000 ppm to 2 wt.% of nickel and from 0 to 8.2 wt.% of water. Results for three compositions will be presented : albite, sodium disilicate and trisilicate. Those glasses were quenched isobarically from a melt equilibrated during 24 to 64 hours at a temperature in the range 1100-1350°C and a pressure in the range 1 bar – 5 kbars.

This study is based on high resolution X-ray Absorption Spectroscopy (XAS) pre-edge analysis. The spectra were collected on ID26 beamline at ESRF. Height and energy variations in the pre-edge features can be related to coordination changes. To interpret glass spectra, the variations have been calibrated using the model compounds in which the Nickel coordination is clearly known.

Anhydrous albite glass (2000 ppm of Ni) has been heated at 1100°C. The pre-edge feature does not present any structural modification in the local environment of nickel which remains 5-fold coordinated. By contrast, in the anhydrous sodium disilicate glass (4000 ppm of Ni), nickel is 5-coordinated at ambient temperature, and 4-coordinated in the melt (1030°C). Around T_g, the coordination number is 6. Water saturated sodium trisilicate (2 wt% of Ni), presents a nepouite crystal structure at ambient temperature in which nickel is 6-fold-coordinated. At 900°C, nickel is 5-fold-coordinated. Transmission Electron Microscopy analysis reveals the presence of nanocrystallites of nepouite in all hydrous glasses.

Experiments clearly show a phenomenon of nanocrystallisation of a nickel-bearing phasenduring the hardening of hydrous glasses. These results imply that hydrous glass

VPP1

Melts and Element Fractionation in Fluid-Magmatic Systems

structure is not a good picture of melt structure and that work has to be done in a diamond anvil cell in order to characterise the effects of pressure, temperature and water contents on the atomic environment of transition metals in volcanic magmas.

VPP1 : SUPO07 : PO H₂O and CO₂ Solubilities in Rhyolitic to Basaltic Melts – An IR Spectroscopic Study

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Dissolved volatiles in silicate melts influence degassing processes in volcanic eruptions. H₂O and CO₂ solubilities in natural rhyodacitic, dacitic and andesitic melts (Unzen volcano, Japan) in equilibrium with H₂O/CO₂ fluids were determined at pressures of 50 to 500 MPa, and at temperatures of 1200 and 1250°C. Water contents of the glasses were analyzed by IR spectroscopy using OH and H₂O combination bands at 4500 and 5200 cm⁻¹, respectively. The molar absorption coefficients for these bands determined by Ohlhorst et al. (in press) decrease strongly from rhyolitic to basaltic glass compositions.

In the pressure range of 50 to 200 MPa (at X_{H₂O}=1) we observe a maximum solubility of water (X_{H₂O} is the mole fraction of water in the fluid phase) in dacitic melts and lower solubilities in both rhyolitic and basaltic melts (at 200 MPa 5.7 wt.% H₂O (18.8 mol% on one oxygen basis) in dacite, 5.2 and 4.8 wt.% (16.8 and 16.6 mol%) in rhyolite and basalt, respectively). In contrast, at 500 MPa the solubility of water is higher in rhyolitic (11.0 wt.%) than in dacitic (10.6 wt.%) melts. At 100 and 200 MPa, the water solubility turns over from a square root dependence on X_{H₂O} at low X_{H₂O} to a linear dependence for X_{H₂O}>0.1 (100 MPa) and >0.2 (200 MPa), respectively. For water contents up to about 6 wt.% (corresponding to X_{H₂O}~0.5) a similar behavior is observed at 500 MPa. At higher X_{H₂O} however, the dependence of water solubility on X_{H₂O} is more pronounced than at 200 MPa. At 200 MPa the solubility of water is always higher in the rhyodacitic melt than in a rhyolitic (Tamic et al., 2000) melt (up to 10%). Whereas, at 500 MPa the water solubility is higher in dacitic melts for X_{H₂O}<~0.7 (up to 50%) but lower for X_{H₂O}>~0.7.

C-species are dissolved both as carbonate and molecular CO₂ (CO_{2,mol}) in dacitic and rhyodacitic glasses. The quantitative determination of the dissolved C-species by IR spectroscopy is complicated because (1) the intensity ratio of both bands varies with water content and cooling rate and (2) the weak carbonate band is difficult to separate from background features in the IR spectra. Our data indicate that either (1) the ratio of CO_{2,mol}/carbonate decreases with increasing water content in the glass, or (2) the absorption coefficient of the CO_{2,mol} band increases with increasing water content in the glass. The solubility of total C-species (expressed as CO₂) for x_{CO₂}=1 at 500 MPa measured with CO₂ - titration is similar for dacitic and rhyodacitic melts (2530 ±100 ppm and 2580 ±100 ppm, respectively).

Tamic N, Behrens H & Holtz F, *Chemical Geology*, (in press).
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VPP1 : SUPO08 : PO Partial Melting of Mafic Rocks from Electrical Impedance Spectroscopy Measurements

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Electrical properties of partially molten mafic rocks have been studied at high temperatures and under pressure by measuring electrical resistance as a function of frequency and time in a natural pyroxene gabbro (Oman) and a synthetic mantle rock (olivine + 5% basalt). Gabbro samples were chosen because of the uniform grain size 250-300 μm. The gabbro rock consists of 50-55 vol% Plg (An₂₀), 35% Cpx (CaFe_{0.25}Mg_{0.75}Si₂O₆) and 15% Opx (Fe_{0.5}Mg_{1.5}Si₂O₆). The electrical impedance experiments

have been carried out in a piston-cylinder apparatus at 5, 10 kbar and temperature up to 1300°C. For each temperature, the electrical impedance in a frequency range from 10⁻² to 10² Hz has been measured. The bulk electrical resistance of samples has been estimated from Argand plots. At each temperature, samples were kept for several days to reach thermal and textural equilibration. At subsolidus temperatures the electrical conductivity of gabbro follows Arrhenian dependence with an activation energy 1.15 ±0.05 eV. At temperature close to the liquidus (1215°C at 10 kbar, 1175°C at 5 kbar), the conductivity increases drastically with the time reaching a steady-state value in few days: at 5 kbar from 3.6x10⁻³ S/m (at 1151°C) to 2.6x10⁻² S/m (1187°C); at 10 kbar from 8.6x10⁻³ S/m (at 1215°C) to 4x10⁻¹ S/m (at 1240°C). The time constant τ in the time dependence of the specific electrical resistance ΔR=[1-exp(-t/τ)] is 3x10³ sec at 5 kbar and 1188°C and 1.4x10³ sec at 10 kbar and 1240°C. This relaxation effect is perhaps due to a slow developing of a better melt interconnection or caused by chemical reactions between crystals and melt. In a partially molten state with about 15 vol% of melt gabbro is characterised by electrical conductivity ca. 0.3-0.4 S/m.

Monday AM Session

VPP1 : MOam01 : G4 Fractionation of Platinum Group Elements and the Formation of Platinum Group Minerals in the Konder Massif, the Aldan Shield

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Isoferroplatinum (n=216) and iridium-osmium alloy (n=66) new analyses from placers, related to dunite stock of the Konder massif, the Aldan Shield (Kravchenko et al., 2000), as well as chromspinele from dunite (Nekrasov et al., 1994) are discussed. It is shown that distribution of platinum group element in isoferroplatinum, and iridium-osmium alloys and Cr and Mg in chromspinele are in good agreement with Rayleigh model of fractional crystallization. According to these results dunites form the comagmatic serie. Equations are: lg Ir= -0.91lg Pd+2.25 and lg CrO=0.624lg MgO+1.24, (for platinum group element in Konder dunites and in other chromspinele ores, and Cr and Mg for chromspineles in dunite correspondingly).

A single-mode statistic distribution of Ir/Pd ratio in isoferroplatinum is established. Compositions of chromspinele, which form inclusions in olivine, and those of interstitial chromspinele are differ markedly. That permits to suppose that microfractionation in dunite melt took place.

The results obtained unambiguously proved the orthomagmatic formation of Konder ores, and are in contradiction with the hypothesis of their postmagmatic genesis (Ukhanov et al., 1997). Nevertheless it is possible that the dunite from upper part of dunite stock, enriched in platinum group elements, is formed by fluid-magma interaction. This dunite also contains zoned chromspineles.

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VPP1 : MOam02 : G4 Fractional Distillation and Critical Phenomena in Earth's Interior

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Magmatic distillation plays important role in the processes of geochemical differentiation. Degassing of granitic magmas may result in the appearance of gas with the elevated contents of HCl and other acid components which after condensation would be agents of acid wall-rock alteration (greisenitization etc). Concentrated saline brines may separate from crystallising granitic melts simultaneously with acid vapour (Ryabchikov, 1975). Subsequent reaction of these brines with the altered by interaction with vapour wall rocks may result in the fixation of alkalis - typical sequence of postmagmatic metasomatic processes (Korzhinsky, 1953). Separation of gas from carbonatites and alkaline magmas results in the mobilisation of alkaline components (Veksler and Keppler, 2000) which subsequently participate in the fenitisation of country rocks. Action of magmatic distillation is restricted by critical phenomena in the systems silicate - volatiles at high pressures (Wyllie and Ryabchikov, 2000). Positions of critical end points in the systems aluminosilicate - water was recently determined experimentally (Bureau and Keppler, 1999; Shen and Keppler, 1997; Stalder et al., 2000). Critical end point on the solidus of lherzolite-water system was approximately placed between 3 and 4 Gpa on the basis of the data on the solubilities of silicates in compressed aqueous vapours in the systems MgO-SiO₂-H₂O, phlogopite-forsterite-H₂O, diopside-jadeite-H₂O (Ryabchikov, 1993). Critical phenomena may be strongly affected by the presence of the additional volatile components. It is widely believed that in the presence of CO₂ melt+vapour field expands considerably by comparison with silicate-H₂O systems. However, at high pressures near solidus melts in lherzolite-H₂O-CO₂ system become

carbonate-rich with fairly high Na_2CO_3 content (Wallace and Green, 1988; Wyllie, 1995). Since in the system $\text{Na}_2\text{CO}_3\text{-H}_2\text{O}$ second critical end point is situated at rather low pressure, the pressure of critical end point in the system lherzolite- $\text{H}_2\text{O-CO}_2$ may not significantly differ from the system lherzolite- H_2O .

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VPP1 : MOam03 : G4

Thermodynamics of Solutes in Aqueous Hydrothermal Solutions at Extreme Conditions: Tough Problems and Potential Solutions

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Traditionally, the thermodynamic properties of solutes in hydrothermal fluids have been described with solution thermodynamics and standard state concepts developed for solutions at ambient conditions. The currently available concepts such as the HKF model for standard state properties, however, have severe shortcomings at extreme conditions (e.g., high T, low P) and are prone to failure in the critical and supercritical regimes. In addition, the HKF concept is questionable for neutral species. The problems of HKF result from the usage of the Born model, using the dielectric constant of water as a measure for the solvent contribution to solute thermodynamics. More recent theoretical work (e.g., by Levelt-Sengers, Chialvo and Cummings, and others) has shown that the solute's properties can strictly be divided into a short-range and a long-range part, which can largely be attributed to the solvent's compressibility. The latter is responsible for the solute's diverging partial molar properties at the solvent's critical point. New models based on these theoretical results may circumvent most of HKF's problems and probably can equally well be applied to neutral and charged species. A brief review of the current state of development of such models is given. The question of calculating standard state properties, however, is only one problem. More severe is the problem of how to handle the thermodynamics in real world systems with finite, sometimes very high concentrations. The conventional choice of standard state for the solute is dangerous since the infinite dilution reference state and the actual state of an ionic solute in high temperature brines with essentially complete ion association are extremely different, making it difficult to develop satisfying activity models.

VPP1 : MOam04 : G4

Thermodynamic of Fluid Mixtures at High Temperature and Pressures

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Water is one of the main components of magmatic and metamorphic fluids. Other important fluid species are CO_2 , CH_4 , N_2 , and various salts. The mixing behavior of these components in water-rich fluids is substantially non ideal. For non-polar gases the deviation from ideality is usually positive. This means that the activities of fluid components in such mixtures are higher than predicted by ideal mixing. With the increase of temperature the non-ideality of mixing decreases. In contrast with increasing pressure higher deviations from ideality are observed. At some P-T conditions these fluid mixtures show such strong a non-ideal behavior that phase separations occur. For example, at 773 K in the system water-nitrogen ($X_{\text{N}_2} = 0.25$) fluid separates into two phases above 10 kbar (Costantino et al. 1991). These immiscible fluids have different composition, densities and viscosities. Therefore, it can be expected that the mechanism of mass transport in the poly-phase fluids differ substantially from that of single phase fluid. Trace elements are fractionated not equally between coexisting fluid phases which can lead to spatial redistribution by the fluid filtration. Coexisting fluids put constraints on the thermodynamic equilibria so that composition of fluid can be used

for thermometry or barometry. The recently developed equation of state for multicomponent fluids (Churakov et al. 2000) has been used to analyze non-ideality of mixing of various non-polar gases with H_2O at different pressures and temperatures relevant to the metamorphic and magmatic processes in the earth crust. Based on the results of thermodynamic equilibria calculations we discuss possibility of the phase immiscibility in the natural fluids at high pressures and temperatures.

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VPP1 : MOam05 : G4

Mathematical Model for Thermodynamic and Transport Properties of the $\text{NaCl-H}_2\text{O}$ System from 0-750°C, 0.1 to 500 MPa, and $X_{\text{NaCl}}=0-1$

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Realistic modeling of fluid flow in magmatic-hydrothermal and other high temperature geological environments is currently mainly limited by the availability of accurate but simple mathematical descriptions of fluid compositions other than pure water. The NaCl -water system is commonly believed to be the best proxy for real fluids in such environments. However, to date only the formulation by Palliser & McKibbin (1998a,b,c) gives a comprehensive mathematical description of this system. While implementing their correlations to integrate them into the CSP software package (Matthai & Roberts, 1999), it was found that their formulations have serious problems at certain conditions that lead to totally unphysical behavior of the model fluid.

In the present contribution, an alternative mathematical description of the phase boundaries in P-T-X space, the fluid density, enthalpy, isobaric heat capacity and dynamic viscosity is presented. In general, accurate mathematical formulations were constructed for the properties along the phase boundaries and properties between these are calculated from appropriate interpolation schemes. Strong emphasis has been put onto an accurate description of the near-critical region including the "bird's beak" near the critical temperature of water. This was done in order to be able to later extend the model to include thermochemical properties as well. A detailed comparison with experimental data demonstrates the accuracy of the model.

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VPP1 : MOam08 : G4

Magmatic Volatile Phase Evolution – Torres del Paine Laccolith (Patagonia/Chile)

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The 12 Ma old Cordillera del Paine laccolith (CPL) is located in the southernmost Andes of Chile. The CPL is composed of a basal layer of gabbroic rocks. A fine to medium grained biotite granite (I type) forms the majority of the pluton. The laccolith intruded mudstones, sandstones and conglomerates of the Cretaceous Cerro Torre and Punta Barrosa formation [1]. The emplacement of the CPL is thought to be related to the subduction of the Chile Ridge [1]. The CPL contains abundant textural evidence of fluid exsolution and eutectic crystallisation, and hence represents a good example for the transport of large quantities of magmatic aqueous fluids to the uppermost level of the crust. Textural observations, oxygen isotope and fluid inclusion data are presented to discuss mechanism of magmatic volatile phase exsolution. The CPL is remarkable for its abundance of miarolitic cavities. Locally, at the margins of the pluton, a microgranitic phase is found with up to 15% of cavities. While some miaroles are isolated,

others are interconnected e.g., forming tube-like structures. Open miaroles contain euhedral crystals of quartz and feldspar. Other important phases are biotite, tourmaline, fayalite and late chlorite and carbonate. Individual crystals are typically between < 1 cm up to a few cm in length. Some miaroles are completely filled with coarse quartz forming pods of up to one meter resembling pegmatites. Miaroles are surrounded by a fine-grained groundmass characterized by an eutectic (micrographic) quartz-feldspar intergrowth. Oxygen isotope data for granites range from 9.1-9.9 ‰ for quartz and 5.1-6.0 ‰ for biotite. The miaroles show a wider range in their quartz values of 8.8-11.1 ‰. Yet, the similarity between miarolitic and granitic quartzes suggests the formation from a common fluid. This is supported by oxygen isotope thermometry of quartz-fayalite pairs from miaroles which yields a temperature of 750 ± 30°C. Analyses of individual hand specimen reveal slight differences between the miaroles and their hosting granite: miarolitic quartzes are between 0.2-0.8 ‰ lighter than the granitic quartzes. At present, these variations are best explained by Rayleigh fractionation effects upon exsolution of the fluid from the granite or modal compositional variations. Comparison of fluid inclusions in phenocryst and miarolitic quartz reinforce the suggestion that the two occurrences of quartz trapped the same magmatic fluid. Low-pressure contact-metamorphic assemblages (e.g. prehnite-bearing) and fluid inclusion data suggest a shallow intrusion level corresponding to pressures of < 1-2 kbar.

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VPP1 : MOam09 : G4

Metals Speciation in the High Temperature Volcanic Gases of the Kudrjaviy Volcano (Iturup Island)

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High temperature gas-jets of Kudrjaviy volcano on the Iturup island (Kuril Islands, Russia) associated with recent near surface magmatic activities. It is well established that the main components of high temperature fumaroles consist of H_2O , CO_2 , SO_2 , H_2S , HCl and HF . The surface temperature of volcanic gases, vary significantly from about 373K up to 1223K. These gases are produced by boiling of shallow level magmas and then cooled down by mixing with cold meteoric water (Tatan et al., 1995). The volcanic gases with highest temperatures bear direct information on the composition of magmas, which they released from, especially volatility contents. Concentration of ore- and rock forming elements in fumaroles vary in the range of 1-100 ppm. This seems to be a very small number but taking into account the bulk emission of magmatic gases evaluated as 200 metric tones per day (Shmulovich & Churakov 1998), total mass of ore-forming elements which could be released in few years of degassing becomes enormous. The chemistry of volcanic gas and associated sublimates possess particular feature different in many aspects to the typical hydrothermal mineralisation. The solid phases sublimated from high temperature gases form monomineral clusters of fine grained tiny crystals, mainly chlorides and sulfides of ore metals, corresponding to the end members of solid solutions. Even the elements with extremely small concentration of about 1-10 ppb, form own solid phases (Tkachenko, et al. 1999). Some of these phases have not been observed as substantive minerals, because usually they enter other phases as isomorphic mixtures (Korzhinskii et al., 1994). Detailed SEM studies and microprobe analyses of sublimates samples indicate presence of native aluminum and silica (Korzhinskii et al., 1995)., despite of oxygen fugacities being too high for this phases to be stable. To explain field observations detailed thermodynamic equilibria in gas-sublimates are needed. The precipitation of minerals from volcanic gases is controlled by the temperature and the form of the chemical transport in the fluid phase. Results from thermodynamic modeling of isobaric cooling of volcanic gases (Churakov et al., 2000) are compared with the field-observation. The calculations shows a drastic change of the main transport species of the metals in gas phase with the cooling. The changes in the chemistry of the gas phase explain the formation of observed sublimates.

VPP1

Melts and Element Fractionation in Fluid-Magmatic Systems

Taran Yu.A, Hedenquist JF, Korzhinskii MA, Tkachenko SI & Shmulovich KI, *Geochim. Cosmochim. Acta*, **59**, 1749-1761, (1995).

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VPP1 : MOam10 : G4 Experiments in Conjugate Basalt-Water Systems – Clue to Podiform Chromite Formation and PGE Fractionation

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Globular textures are observed in many geological settings, such as ovoidal apatite ores, ocelli in various Archean volcanics, and nodular chromites in so-called leopard ore in podiform chromitite deposits. The textures are presumably magmatic in origin. Wherever globular textures occur in magmatic rocks, the globules are characterized by sharp interfaces and often show plastic deformation against each other and against the matrix. The origin of globular textures is not well understood. Here we report experiments to understand the formation of chromite nodules in podiform chromite ore of supra-subduction zone ophiolites. It is shown that in conjugate basalt-water systems, chromite and refractory PGE alloys concentrate exclusively in the exsolved H₂O-rich fluid.

A picritic composition was doped with 5 wt. % chromite component and 1 wt. percent of Ru as metal powder. Experiments were performed under water-saturated conditions at 1150°C and 0.5 GPa in a piston cylinder. Run products quench to devitrified glass and an exsolved aqueous fluid that decomposes upon quenching to sodic silicate flakes and water. Chromite crystals and metallic Ru nuggets are found suspended exclusively in the quenched fluid pools. Time series experiments show that fractionation of chromite and PGE metal nuggets into the fluid is extremely fast. After 15 min, chromite crystals and water bubbles are evenly distributed in glass. After one hour, the fluid has already formed an interconnected channel network, and after about 10 hours, separation of silicate melt and fluid is complete. All chromite grains and all Ru nuggets are exclusively found suspended in the fluid phase. Silicate melt and exsolved fluid are separated by stable, sharp interfaces. In experiments with controlled quench rates of 1°/min, the fluid pools crystallize to euhedral pargasitic amphibole coexisting with chromite and Ru nuggets.

The experimental results compare well with natural observations. Primitive mantle melts in supra-subduction zone settings - the most likely location where podiform chromite concentrations form - are known to be water-rich. Judging from water contents of primitive boninites, fluid exsolution should occur at pressures around 0.2 GPa. The exsolved fluid, initially present as bubbles, collects suspended liquidus chromite grains and refractory PGE alloys, presumably because interfacial energy differences between chromite and alloy are smaller in contact with fluid than with silicate melt. The collection mechanism is equivalent to flotation.

Podiform chromitites are commonly found enriched in Os, Ir, Ru, and Rh (i.e. the refractory PGEs), relative to Pt and Pd. The basaltic melts associated with podiform chromitites show a complementary depletion pattern in refractory PGEs. In the past, this has been attributed to Os, Ir, and Ru entering to some extent the chromite lattice. Our results imply that PGE fractionation may have physical reasons. If podiform chromite ores form in conjugate basalt-water systems and if the basalt is saturated with refractory PGE nuggets, these nuggets will concentrate, along with chromite, in the fluid phase.

VPP1 : MOam11 : G4 Thermodynamical and Rheological Properties of a Hydrous Basaltic Melt

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Volcanological processes are governed by many physical and chemical parameters (such as pressure, temperature, chemical composition...) well known for exerting a strong influence on the physical properties of molten silicates. Amongst compositional variables, the volatile content plays an important role on fragmentation, degassing and transport of magmas. Recent work in our lab has for instance shown the tremendous effect of dissolved water on the density and viscosity of silicate melts of certain compositions (Whittington et al. 2000). Basalt is the most common magmatic composition on the Earth's surface, occurring mostly as oceanic crust and continental floods, and yet, the effects of the volatile content - and especially that of water- on its physical properties are poorly known. Part of the reason for the lack of experimental data is the high pressures required to dissolve significant amounts of water in synthetic glassy samples (analogous of melts). Our goal is to bring out the role of dissolved water on the properties of basaltic glass samples bearing from 0 to 3 wt% H₂O, synthesized in a high pressure vessel at 5 kbar and 1200°C. Several experiments are performed on the samples, we measure the density with the Archimedes method, the viscosity near the glass transition range, i.e. at temperatures < 1000°C, with a creep apparatus, the thermal expansion of the melt with a dilatometer and the compressibility from Brillouin scattering. We will finally investigate, for the first time experimentally, the enthalpy of exsolution (evaporation) of water from the basaltic melt, using Differential Scanning Calorimetry (DSC), in order to study temperatures of exsolution of water in the future. First viscosity results already attest the strong influence of water: the addition of 1.5 wt% reduces the viscosity of the basaltic melt by over a factor of 3.

Whittington A, Richet P, Holtz F, *Geochim Cosmochim Acta*, **64**, 3725-3736, (2000).

VPP1 : MOam12 : G4 The Effect of Fe₂O₃ on the Viscosity of Melts in the Na₂O-Al₂O₃-SiO₂ System

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As many magmas have a composition close to the subaluminous join it is important to understand the changes in viscosity as a function of Na:Al ratio in this compositional range. It has been previously shown in the Na₂O-Al₂O₃-SiO₂ system that there is a maximum in viscosity in the vicinity of the peraluminous join. This maximum occurs within the peraluminous field, suggesting the presence of triclusters consisting of one aluminate and two silicate tetrahedra.

In order to take these observations further we have determined the effect on viscosity of the addition of Fe₂O₃ to melts in the Na₂O-Al₂O₃-SiO₂ system with 50 mol% SiO₂, and Na:Al ratios from 0.4 to 0.6. The micropenetration technique has been used to measure viscosities in the range 10⁹ to 10¹² Pa s.