

Experimental model for alkalic chloride-rich liquids in the upper mantle

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ARTICLE INFO

Article history:

Received 24 September 2008

Accepted 14 March 2009

Available online 28 March 2009

Keywords:

High-pressure experiment
Chloride–carbonate–silicate melts
Liquid immiscibility
Upper mantle
Inclusions in diamonds
Kimberlites

ABSTRACT

In order to expand an applicability of a model for evolution of the alkalic chloride-rich liquids in the upper mantle additional runs in chloride–carbonate–silicate systems involving model silicate end-members were performed at 5 GPa. Results of experiments can be evaluated using the single scheme of phase relations. They show that interaction of the chloride–carbonate liquids with silicates produces a wide range of the chloride-rich carbonate–silicate melts, whose composition is determined by the chloride/carbonate ratio in the chloride–carbonate liquid and composition of the interacting silicate assemblage. The carbonate–silicate melts are immiscible with Si–Al-saturated chloride–carbonate melts. With decreasing temperature, evolution of both melts proceeds toward the homogeneous chloride–carbonate or carbonatitic liquid coexisting with silicates. Trends for melt evolution in the model systems closely reproduce the evolution of the carbonate–silicate and saline inclusions in kimberlitic diamonds. These trends suggest a diamond growth during cooling. In addition, experiments showed stronger effect of interaction of eclogite-related phases with the alkalic chloride–carbonate melts, in contrast to peridotite-related phases.

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1. Introduction

Findings of alkalic chloride-rich (saline) inclusions in kimberlitic diamonds worldwide (Chen et al., 1992; Johnson et al., 2000; Izraeli et al., 2001, 2004; Klein-BenDavid et al., 2004, 2006, 2007; Tomlinson, 2006; Tomlinson et al., 2006; Zedgenizov et al., 2007a) suggest their direct relation to kimberlite magmatism. The saline inclusions are mixtures of carbonates and alkali chlorides (predominately, KCl), containing 10–30 wt.% of H₂O, and 3–6 wt.% of silicate and phosphate components (Fig. 1). Associations of these inclusions with mineral inclusions of both eclogitic and peridotitic assemblages show that the fluids were trapped by diamonds coevally with silicate minerals at pressures around 4–5 GPa (Izraeli et al., 2004; Klein-BenDavid et al., 2006, 2007; Tomlinson, 2006; Tomlinson et al., 2006). Recent findings of the chloride–carbonate inclusions in a diamond from an eclogitic xenolith (Zedgenizov et al., 2007a) and in minerals of a sheared lherzolite xenolith (Golovin et al., 2008) of the Udachnaya pipe imply that the saline liquids could be encapsulated by mantle minerals prior or during the incipience of the kimberlite magma. Moreover, the

chloride–carbonate inclusions in groundmass olivines from kimberlites (Fig. 1; Kamenetsky et al., 2004, 2007, this issue) suggested that these liquids accompanied further evolution of the kimberlitic magmas up to their latest stages.

The saline liquids are compositionally linked to alkalic silica-poor carbonatitic Cl-bearing liquids, which are also found in fibrous diamonds world wide (Fig. 1). In turn, carbonatitic liquids are connected to carbonate–silicate and silicate (silicic) liquids (Fig. 1). However, a lack of links between carbonate–silicate and saline liquids suggests a wide miscibility gap between them at the upper mantle *P–T* (Navon et al., 2003; Safonov et al., 2007a). The immiscibility in the alkalic chloride–carbonate–silicate melts has been invoked as a basis for the model of evolution of these liquids (fluids/melts) in the upper mantle (Perchuk et al., 2002; Navon et al., 2003; Safonov et al., 2007a; Klein-BenDavid et al., 2007, this issue). Navon et al. (2003, 2008) and Klein-BenDavid et al. (2007) suggested that the carbonatitic high-density fluid (HDF) could be parental for all the other types of liquids, while its evolution toward the miscibility gap proceeds by initial fractionation of carbonates, followed by separation into the saline and silicic portions. Experiments in the system CaMgSi₂O₆–(Na₂CO₃ ± CaCO₃)–KCl at 5 GPa (Safonov et al., 2007a) supported an existence of a wide miscibility gap between Cl-bearing carbonate–silicate (*L*_{CS}) and Si-bearing chloride–carbonate (*L*_{CC}) melts. It was found that precipitation of silicate resulted in displacement of the *L*_{CS} composition along the gap boundaries toward the low-temperature carbonate region, where it mixes with *L*_{CC} to form homogeneous liquids saturated with silicate components. Thus, in contrast to models by Navon et al. (2003, 2008) and Klein-BenDavid et al. (2007), our

Abbreviations: Cpx, clinopyroxene; Di, diopside; En, enstatite; Fo, forsterite; Grt, garnet; Grs, grossular; Jd, jadeite; *L*_{CC}, chloride–carbonate melt; *L*_{CS}, carbonate–silicate melt; Mc, mica; Mrw, merwinite; Phl, phlogopite; Prp, pyrope; Spl, spinel.

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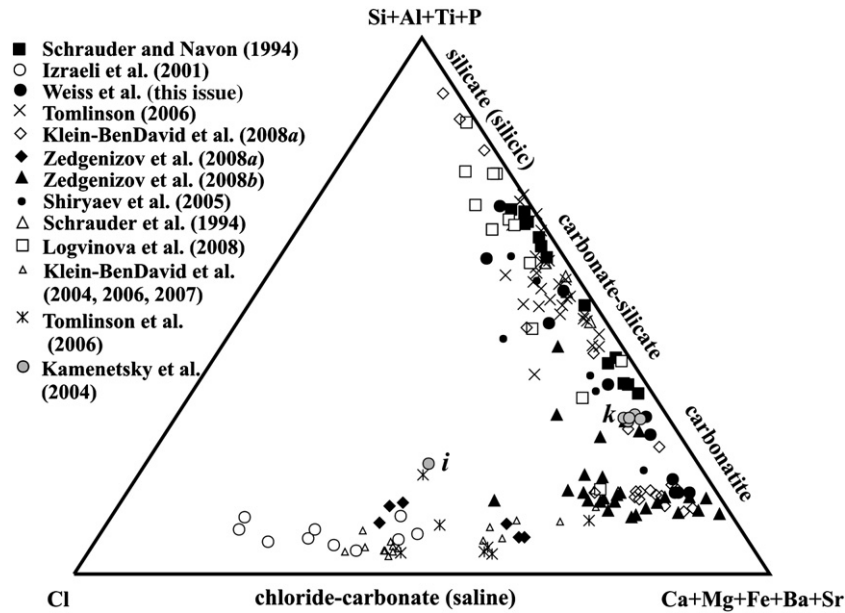


Fig. 1. Compositions of the melt/fluid inclusions in diamonds and olivines from kimberlites of *African pipes*: 1. Jwaneng (Schrauder and Navon, 1994), 2. Koffiefontein (Izraeli et al., 2001), 3. Kankan (Weiss et al., this issue), 4. Mbuji Mayi (Tomlinson, 2006); *Yakutian pipes*: 5. Udachnaya, Aykhal, Yubileynaya, Zarnitsa, Komsomolskaya (Klein-BenDavid et al., 2008), 6 and 7. Udachnaya (Zedgenizov et al., 2007a,b); 8. Brazil (Shiryaev et al., 2005); 9. India (Schrauder et al., 1994); *Canadian pipes*: 10. Snap Lake (Logvinova et al., 2008), 11. Diavik (Klein-BenDavid et al., 2004, 2006, 2007), 12. Panda (Tomlinson, 2006; Tomlinson et al., 2006). 13. Inclusions (i) in groundmass olivine from salty kimberlites (k) of the Udachnaya pipe (Kamenetsky et al., 2004). Alkalis (K and Na) are excluded from the diagram, since they mask behavior of chlorine (Safonov et al., 2007a).

experiments (Safonov et al., 2007a) demonstrate that the chloride-rich carbonatite melt is unable to produce immiscible liquids by fractionation. An alternative model (Safonov et al., 2007a) considered the formation of carbonate–silicate Cl-bearing melts via interaction of mantle silicate rocks with the external saline liquids. In order to further demonstrate an applicability of the model additional runs involving model silicate end-members were performed.

2. Starting materials, experimental technique, and analytical procedure

Mixtures of crystalline CaCO_3 , Na_2CO_3 , and KCl were used as components of starting chloride–carbonate melts, since the data on the melt inclusions in diamonds show that KCl is the major Cl-contributing component in them, while Na and Ca are mostly bound to carbonates (Izraeli et al., 2004; Klein-BenDavid et al., 2006; Shiryaev et al., 2005; Kopylova et al., this issue). Two chloride–carbonate mixtures were prepared for the experiments in the ternary system (Table 1 of the supplementary data): KCl-rich mix. 1 and carbonate-rich mix. 2. They were proportionally mixed with synthetic forsterite (Mg_2SiO_4), MgSiO_3 gel, $\text{Mg}_3\text{Al}_2\text{Si}_3\text{O}_{12}$, $\text{Ca}_3\text{Al}_2\text{Si}_3\text{O}_{12}$, $\text{CaMgSi}_2\text{O}_6$, and $\text{NaAlSi}_2\text{O}_6$ glasses (Table 1 of the supplementary data). The chloride–carbonate material was predominant in the starting chloride–carbonate–silicate mixtures (i.e. 71 wt.%) in order to clearly see intensity of reactions. About of 20 mg of each mixture were placed into Pt or $\text{Pt}_{60}\text{Rh}_{40}$ capsules and subsequently dried at 110 °C. No additional water was added into the starting mixtures. However, despite drying, the starting mixtures contained some uncontrolled amount of water because of hygroscopic nature of KCl.

All experiments (Table 1 of the supplementary data) were performed at 5 GPa with the high-pressure toroidal “anvil-with-hole” apparatus following to the same procedure as in our previous study (Safonov et al., 2007a). The microscopic features of run products (Fig. 2a–i) were examined in polished samples by means

of BSE with a CamScan MV2300 (VEGA TS 5130MM) electron microscope equipped with the EDS electron microprobe Link INCA Energy. In order to avoid a loss of soluble phases, after polishing with diamond paste surfaces of the samples were cleaned by compressed air, ethanol or acetone. Microprobe analyses were performed at 20 kV accelerating potential, 10 nA of beam current, and beam diameter of 3 μm . Products of melt quenching were analyzed with rastered beam over areas 500–20 μm^2 . The ZAF matrix correction was applied. In order to compare quenched carbonate–silicate and chloride–carbonate liquids, their compositions in Table 2 of the supplementary data are shown as wt.% of elements, since the oxygen basis for the chloride–carbonate liquids is not fully justified. No special attempt to analyze carbon in the quenched melts was made. It is responsible for low total oxide content (below 95 wt.%) in analyses of the quenched melts (Table 2 of the supplementary data). We have adopted an assumption (Safonov et al., 2007a) that the difference between the total oxide content and 100% can be tentatively used as a marker for variation of carbonate component with respect to other components in the melts.

3. Results of experiments

Fig. 3 shows phase relations in the system Mg_2SiO_4 – CaCO_3 – Na_2CO_3 –KCl exemplified by mixture [mix. 2] $_{71}\text{Fo}_{29}$. Quenching products of the homogeneous chloride–carbonate melt ($L_{\text{CC}}^{(1)}$) coexisting with euhedral grains of Fo and Mrw were identified in the experiment at 1360 °C (Fig. 2a). Increase of temperature to 1450 °C results in the incipience of the immiscible carbonate–silicate melt ($L_{\text{CS}}^{(2)}$, Figs. 2b and 3), which forms by dissolution of Fo and Mrw in the chloride–carbonate melt. Coexisting $L_{\text{CC}}^{(2)}$ slightly enriched in Si and Mg with respect to the melts at lower temperatures (Fig. 3). Two immiscible liquids ($L_{\text{CC}}^{(3)}$ and $L_{\text{CS}}^{(3)}$ in Fig. 3) only coexist above 1500 °C. Because of consumption of Fo, compositions of $L_{\text{CS}}^{(3)}$ are Si-enriched, while composition of $L_{\text{CC}}^{(3)}$ is much close to the K+Cl apex. Thus,

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