

Carbonatite to kimberlite link in the chloride-carbonate-silicate systems

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In addition to conceptual studies on crustal carbonatites, Peter Wyllie and his colleagues firstly predicted that kimberlitic magmas could be produced via gradual transition from Mg-rich carbonatitic melts formed during partial melting of carbonatized mantle peridotites (Wyllie, Huang, 1975, 1976). New experimental and natural data show that the introduction of alkali chlorides in the kimberlitic melts, i.e. in the carbonate-silicate (CS) systems, results in immiscible separation of chloride-carbonate (CC) melts. Available experimental data on the peridotite-related CS-CC systems at pressures above 4 GPa (Safonov *et al.*, 2007, 2009; Litasov, Ohtani, 2009) allow testing the role of chlorides in the formation of kimberlitic melts with participation of the CC liquids. In all systems studied, the homogeneous CC melt coexists with crystalline silicates (+oxides) at low temperature, while intensive reactions of silicates with CC liquids at increasing temperature result in saturation of these liquids in Si and Al. The major agents of these reactions, however, are carbonate components of the CC melts, while no clear reactions between chloride components of the CC melts and peridotitic minerals are observed. In most cases, "dissolution" of pyroxenes and garnets results in formation of olivine and enrichment of the melts. The Si-Al-saturation of the CC melt at higher temperature causes CC-CS immiscibility. Re-distribution of Ca-carbonate component from CS melt to coexisting CC leads to a shift of its composition toward kimberlite at 1400-1600°C performing the enrichment of the melt in alkalis and chlorine. Similar liquids occur in the Udachnaya-East pipe as the Cl-rich kimberlites (Kamenetsky *et al.*, 2004; Maas *et al.*, 2005), as well as preserved as Cl-rich carbonatitic inclusions in diamonds and olivines worldwide (Navon *et al.*, 2008; Kamenetsky *et al.*, 2008).

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An Atomic Force Microscopy study of calcite growth, as a function of the Ca²⁺:CO₃²⁻ ratio in solution at constant supersaturation

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The dissolution and growth of calcite play an important role in geochemical processes and so factors influencing these processes are of broad interest. Calcite growth in natural systems often occurs where the Ca²⁺ and CO₃²⁻ ion ratio is not 1, such as seawater with a ratio of approximately 5:1 [1]. Also, non-equivalent [Ca²⁺] : [CO₃²⁻] ratios have been shown to play a definitive role in biomineralizing systems. In industrial calcite precipitation, it has been shown that the Ca²⁺ : CO₃²⁻ ratio plays an important role in determining the morphology, providing a controlled synthesis route without the need for expensive additives [2]. Calcite growth experiments using Atomic Force Microscopy (AFM) were conducted at two constant supersaturations, while varying the ion [Ca²⁺] to [CO₃²⁻] activity ratio. Measurements of the growth rates of calcite surfaces, calculated at an atomic scale showed a direct relationship between the [Ca²⁺] : [CO₃²⁻] ratio and the rate at which existing etch pits can close and overlapping steps can form. The growth rate was maximum for a solution with equal amounts of Ca²⁺ and CO₃²⁻ concentrations ($r = [Ca^{2+}] : [CO_3^{2-}] = 1$), while for $r < 1$ and $r > 1$ the rates decrease almost symmetrically. A constant and equal supply of [Ca²⁺] and [CO₃²⁻] would be needed for an effective and fast growth of a calcite substrate. It is also evident that a lower supersaturation leads to a lower growth rate. Different fitting functions were needed on each side of the $r = 1$ composition plot, suggesting a different growth mechanism. Different nuclei develop when [CO₃²⁻] \gg [Ca²⁺] or [CO₃²⁻] \ll [Ca²⁺]. In the first case the nuclei are elliptical while in the second case the nuclei have jagged irregular edges. These shapes are also consistent with the shapes that the etch pits developed upon contact with the growth solution and were retained until the closing was complete.

[1] Holland *et al.* (1986) *Nature* **320**, 27-33. [2] Carmona *et al.* (2003) *J. Coll. Interf. Sc.* **261**, 434-440.