

ON POSSIBLE MECHANISM OF PERICLASE ASSOCIATED WITH DIAMOND AND FORSTERITE FORMATION UNDER UPPER MANTLE CONDITIONS

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During the last decade carbonate-carbon systems have been attracted a considerable interest in the field of experimental studies on diamond crystallization. Recently it has been established that carbonates are able to provide diamond crystallization within a wide range of temperatures and pressures [1-5]. In this case alkaline carbonates have the lowest melting temperatures and provide diamond crystallization at the parameters of diamond formation in nature [5]. With the intent of adequate modeling of natural processes a systematic study of more complicated systems can be initiated basing on the whole set of available data.

In this connection to model diamond formation processes we choose the following systems: K_2CO_3 - SiO_2 -C, K_2CO_3 - Mg_2SiO_4 -C и K_2CO_3 -MgO-C. All the experiments were carried out at 6,3 GPa, 1650°C and 40 hours using a multi-anvil high-pressure apparatus of "split-sphere" type. The silica/carbonate and forsterite/carbonate ratios were chosen as a variable parameter. The experiments were performed using a high-pressure cell in the form of tetragonal prism made of refractory oxides. Pressure in the cell was determined by correlation of the results of diamond crystallization in the nickel-carbon system and P-T parameters of the graphite-diamond equilibrium line. The temperature in each experiment was controlled by platinum-rhodium thermocouple calibrated at high pressure according to the gold and nickel melting. Phase composition of the samples was studied using a DRON-3 diffractometer ($Cu_{K\alpha}$ -radiation). Chemical composition was determined using a "Camebax-Micro" X-ray microprobe-analyzer.

The features of the phase formation in the studied systems were so that as the content of silicate increases in the systems than a carbonate-silicate melt no longer provides spontaneous diamond nucleation and at high concentrations of silicate diamond growth only on seeds and metastable graphite re-crystallization occur. When the melt is saturated by silica in K_2CO_3 - SiO_2 -C system than at first potassium wadeite and later coesite crystallize. In system K_2CO_3 - Mg_2SiO_4 -C when the silicate content increases, the melt first becomes supersaturated relative to forsterite and later to forsterite and periclase.

The ceasing of spontaneous diamond nucleation in the studied systems coincides with the beginning of crystallization of metastable graphite. The joint crystallization of metastable graphite and stable diamond has been established previously in C-O-H fluid and in alkaline carbonate-fluid-carbon systems. It is probably caused by kinetic peculiarities of diamond and graphite producing processes. Following from this we assume that crystallization of metastable graphite in our experiments is related to the expansion of induction period of diamond nucleation in the area of high concentrations of silicate.

Forsterite and periclase crystallize as the silicate concentration in K_2CO_3 - Mg_2SiO_4 -C system increases from 40% Mg_2SiO_4 (table 1). This process is accompanied by the increase of SiO_2 /MgO ratio in the melt from the values close to forsterite stoichiometry (0,75) up to the values, which are essentially greater than enstatite stoichiometry (4,6). The simultaneous formation of forsterite and periclase in a similar $K_2Mg_2Si_2O_7$ - K_2CO_3 system at 3,7 GPa and 1300-1650°C has been described earlier in [2]. The formation of periclase is probably related to the higher than that of MgO solubility of SiO_2 in K_2CO_3 melt. Thus, according to our estimations based on the results of the experiments in K_2CO_3 - SiO_2 -C и K_2CO_3 -MgO-C systems (table 1), SiO_2 solubility in K_2CO_3 melt is at least 5 times greater than MgO solubility. An indirect evidence for this are the experimental data of [4] where at 7,0 GPa and 1700-1750°C in K_2CO_3 - SiO_2 -MgO- Al_2O_3 -C system with oxides ration corresponding to garnet stoichiometry ($Mg_2Al_3Si_3O_{12}$) spinel ($MgAl_2SiO_4$) was found to crystallize whereas silica probably dissolved completely in carbonate melt. There are two possible variants accounting for the periclase occurrence in the mantle medium of diamond crystallization. According to the first variant, periclase in association with MgSi perovskite was formed through olivine decay under the conditions

of low mantle [6]. According to the second, periclase could be formed as a result of magnesite reduction in carbonized dunite [7]. Basing on the obtained experimental data the third variant can be proposed. According to this variant, periclase could be a product of metasomatic influence of high-potassium mantle melts-fluids on ultra-basic rocks.

Table 1.

Experimental results on crystallization of diamond in the K_2CO_3 - SiO_2 -C, K_2CO_3 - Mg_2SiO_4 -C, and K_2CO_3 - MgO -C, systems at 6,3 ГПа, 1650°C and 40 hours duration.

Run №	Initial composition	C, wt. %	Products	Diamond crystallization	
				nucleation	growth
1	$K_2CO_3+SiO_2+Gr$	5	Dm+Q(KC+K-wad)	+	+
2	$K_2CO_3+SiO_2+Gr$	10	Dm+Q(KC+K-wad)	+	+
3	$K_2CO_3+SiO_2+Gr$	25	Dm+Gr+Q(KC+K-wad)	+	+
4	$K_2CO_3+SiO_2+Gr$	50	K-wad+Dm+Gr+Q(KC+K-wad)	–	+
5	$K_2CO_3+SiO_2+Gr$	75	Co+Dm+Gr+Q(KC+K-wad)	–	+
6	$K_2CO_3+Fo+Gr$	5	Dm+Q(KC)	+	+
7	$K_2CO_3+Fo+Gr$	10	Fo+Dm+Q(KC)	+	+
8	$K_2CO_3+Fo+Gr$	10	Fo+Dm+Q(KC)	+	+
9	$K_2CO_3+Fo+Gr$	25	Fo+Dm+Q(KC)	+	+
10	$K_2CO_3+Fo+Gr$	25	Fo+Dm+Q(KC)	+	+
11	$K_2CO_3+Fo+Gr$	30	Fo+Dm+Q(KC)	+	+
12	$K_2CO_3+Fo+Gr$	40	Fo+Pc+Dm+Q(KC)	+	+
13	$K_2CO_3+Fo+Gr$	40	Fo+Pc+Dm+Q(KC)	+	+
14	$K_2CO_3+Fo+Gr$	50	Fo+Pc+Dm+Q(KC)	+	+
15	$K_2CO_3+Fo+Gr$	50	Fo+Pc+Dm+Q(KC)	+	+
16	$K_2CO_3+Fo+Gr$	75	Fo+Pc+Dm+Gr+Q(KC)	–	+
17	$K_2CO_3+Fo+Gr$	75	Fo+Pc+Dm+Gr+Q(KC)	–	+
18	$K_2CO_3+Fo+Gr$	75	Fo+Pc+Dm+Gr+Q(KC)	–	+
19	$K_2CO_3+Fo+Gr$	75	Fo+Pc+Dm+Gr+Q(KC)	–	+
20	$K_2CO_3+Fo+Gr$	90	Fo+Pc+Dm+Gr+Q(KC)	–	+
21	$K_2CO_3+Fo+Gr$	90	Fo+Pc+Dm+Gr+Q(KC)	–	+
22	$K_2CO_3+Fo+Gr$	95	Fo+Pc+Gr+Q(KC)	–	–
23	$K_2CO_3+Fo+Gr$	95	Fo+Pc+Gr+Q(KC)	–	–
24	$K_2CO_3+MgO+Gr$	1	Dm+Q(KC+KMC)	+	+
25	$K_2CO_3+MgO+Gr$	5	Pc+Dm+Q(KC+KMC)	+	+

C – initial concentration of SiO_2 or forsterite in systems, Fo – forsterite, Pc – periclase, Co – coesite, K-wad – potassium wadeite ($K_2SiSi_3O_9$), KC – K_2CO_3 , KMC – $K_2Mg(CO_3)_2$, Dm – diamond, Gr – graphite, Q – quench phases.

It should be noted that in both K_2CO_3 - Mg_2SiO_4 -C and K_2CO_3 - MgO -C systems periclase crystals are represented by octahedra. At the same time in [3] the formation of cubic periclase crystals under crystallization from the sodium carbonate melt was found. Probably the variation of periclase crystal morphology is related to the change of the composition of crystallization medium and incorporation of sodium impurity in periclase structure. Thus, in [8] it was shown that the increase of Na activity in crystallization medium could cause Na entering (up to 2 wt. Na_2O) into periclase structure at 3-5 GPa and 1300-1600°C.

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