EXPERIMENTAL MODELING OF "SUPERDENSE" FLUID INCLUSIONS FORMATION

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The experiments of increasing fluid inclusions' density were done with quartz samples where the inclusions of H_2O-CO_2 composition were preliminarily synthesized at 700°C and 3 kbar. Oxalic acid has been used as a fluid source. The experiments were performed in platinum capsules at 400°C, 3 kbar and 700°C, 5 kbar during 7 days. Oxygen fugacity was supported by NNO buffer. Before and after the experiments the inclusions were studied under the microscope and analyzed using digital camera, video- and computer technique. Temperatures of homogenization (Th) and melting (Tm) of carbon dioxide were measured using Linkam heating-cooling stage (THMSG 600) with the working temperature range from -195 to 600°C. The accuracy of measurements was ±0.1ºC. Synthesized inclusions are mostly flat isometric, sometimes faceted to some extent, sometimes more volumetric and elongated. All of the obtained inclusions have decreased CO_2 melting temperatures comparing to pure carbon dioxide (-56.6°C) (Table 1,2), what indicates a possible methane impurity. It is confirmed by the Raman spectroscopy data (IM&P SB RAS, Novosibirsk) in accordance to which methane content in carbon dioxide varies from 4.2 to 5.4 mol.%. Volume relationships of fluid phases (CO_2/H_2O) in the inclusions are 0.9/0.1 on the average.

Table 1. Modeling of isobaric cooling, thermocriometric data.

№ incl	(-)Tm		Th	
	before the run	after the run	before the run	after the run
1	62.2	60.8	10.3	10.0
2	62.2	60.4	12.0	11.0
3	61.2	59.6	13.6	13.4
4	60.4	60.0	15.7	16.9
5	60.1	56.9	16.4	-1.5
6	59.5	59.5	14.6	14.7
7	59.2	59.0	17.6	17.7
8	60.4	59.0	18.6	18.3
9	64.1	61.0	8.3	8.3
10	62.6	61.2	7.7	7.7
11	61.1	59.6	14.9	14.6
12	59.3	59.0	15.1	14.9
13	60.0	59.8	14.2	14.1
14	60.8	60.6	12	11.8
15	60.5	59.9	13.2	13.0
16	60.9	60.7	10.6	10.6
17	61.1	60.7	11.6	11.5
18	57.9	60.4	12.8	-3.5

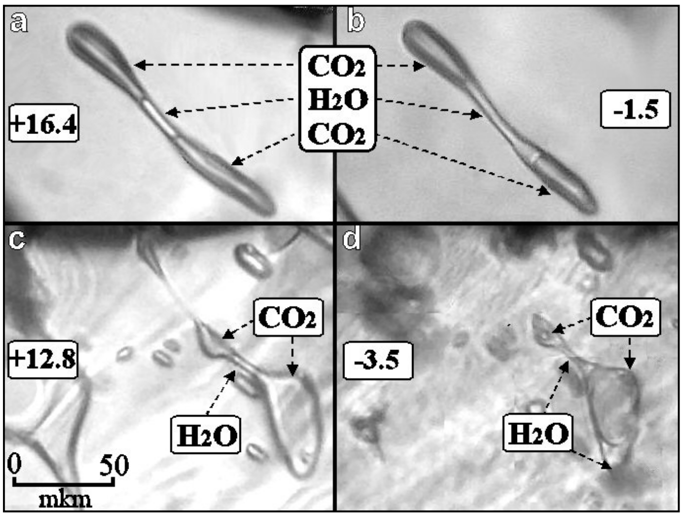

Fig.1. Two most changed inclusions: a,c – before the experiment; b,d – after the experiment. Carbon dioxide homogenization temperatures are denoted by figures.

Under the conditions of isobaric cooling (400°C, 3 kbar) the most noticeable changes took place in two inclusions, which were closer than others to the surface of the sample. The volume of the aqueous phase increased in them with corresponding decreasing of the CO_2-rich phase volume (Fig.1); the temperature of homogenization sharply decreased, the temperatures of melting of carbon dioxide changed (№№5 and 18, Table 1). No changes of the inclusions' shape occurred in this case. All of these indicate the formation of such dense inclusions not due to the "overdensity" of the primary less dense varieties, but as a result of penetration of a more dense external fluid through microdislocations. Less marked changes took place in the inclusions, more remote from the fluid surface. The volume relationship of phases did not change visually. A change of the vacuole form is observed in some inclusions: faces appeared, as well as sharp angles, sometimes uneven "eaten" boundaries. If before the experiment an aqueous phase was usually uniformly distributed over the walls of the vacuole, then after the experiment in many inclusions it was concentrated in small "apophyses". Temperatures of homogenization did not change on the whole (Table 1), but the temperatures of melting of carbon dioxide in all inclusions increased, what definitely indicates the decrease of methane impurity that could take place either in the process of its selective loss or as a result of new depleted by methane fluid portion leakage in the inclusions. In this case the absence of a marked increase of Th can show a great probability of the second case – "sucking" of the external fluid through the microdislocations. Probably, a more remote position of these inclusions from the sample surface did not allow reaching full equilibrium between the fluid in the inclusions and the external one.

At 700°C and 5 kbar all the inclusions were necked down with the formation of a great number of small varieties (Fig.2). These inclusions are characterized by notably lower temperatures of homogenization and slightly decreased temperatures of melting of carbon dioxide comparing to the initial sample (Table 2). The largest varieties (relatively more dense and with lower Tm) were formed at necking down of the inclusions filling the large channels (Fig.2).

Table 2. 700°C and 5 kbar. Thermocriometric data. Figures in brackets – a number of measurements.

group of incl.	before the experiment		after the experiment	
	(-)Tm	Th	(-)Tm	Th
1	58.6-61.6 (8)	+6.2-+13.4 (13)	61.1-61.6 (5)	-8.8-+9 (11)
2	58.1-61.6 (19)	+4.4-+16.9 (23)	59.8-61.2 (10)	-8.1-+7.7 (38)
3	59.5-61.6 (8)	+4.9-+12.1 (8)	57.4-61.4 (8)	-3-+4.3 (15)
4	59.7-61.5 (7)	+6.2-+15.0 (11)	61.0 (1)	+0.7-+3.1 (9)
5	58.8 (2)	+11.9-+13.1 (10)	59.6 (4)	-6.6-+10.6 (12)
6	59.1 (1)	+12.9-+13.2 (3)	59.5 (1)	+10.8-+12.0 (3)
7	60.7-60.8 (4)	+7.9-+9 (6)	61.3-61.6 (5)	-1.8-0 (4)
8	58.4-59.7 (8)	+11.8-+13.6 (8)	57.5-62.1 (12)	-2.7-+11.5 (16)
9	58.7-59.7 (2)	+11.8-+12.5 (2)	59.3-61.3 (4)	-4.8-+12.1 (16)
10	58.6-58.8 (4)	+13.2-+14.3 (4)	59.3 (1)	+9.9-+10.4 (3)

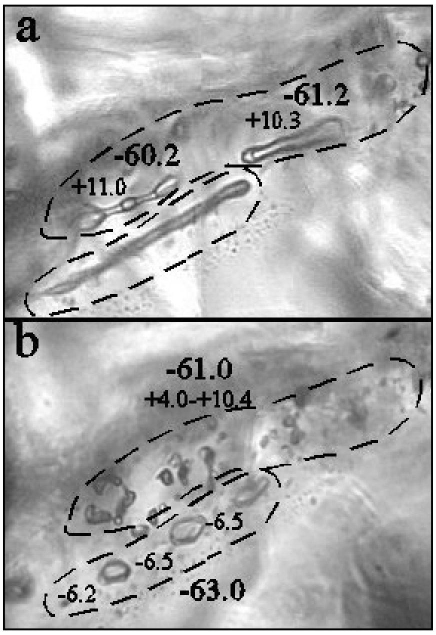

Fig.2. Necking down of fluid inclusions at T=700°C and P=5kbar; a – inclusions before the experiment; b – after the experiment. The sections, corresponding the same groups of inclusions are distinguished by dashed lines. Carbon dioxide homogenization and melting temperatures are denoted by figures.

Theoretically the increasing fluid inclusions' density can take place because of two reasons: either as a result of the decrease of the vacuole volume [1,2] or due to the leakage of a more dense fluid in the inclusions [3]. The experiments described above were done under hydrostatic conditions in the absence of stress tensions. Under such conditions the decrease of the vacuole volume due to deformations was very unlikely. At least, at a rather detailed study of the inclusions there no evidence of such a change of their volume has been discovered. A change of temperatures of melting of carbon dioxide is also in favour of the hypothesis of "sucking" of the external fluid into the inclusions. Probably, a more dense external fluid, differing in composition from the internal one, penetrated into the inclusions through microdislocations and microcracks, causing the processes of dissolution and substance redeposit, what resulted in necking down and increasing fluid inclusions' density. At that the easier access of the external fluid to large channels caused their more intensive change.

Thus, an experimental modeling confirmed a possibility of formation of dense ("superdense") fluid inclusions as a result of entrapment of the additional fluid directly from the external medium. The example of such "overdensity" of the inclusions was found while investigating of charnockite formation and evolution of metamorphism of the Kerala Khondalite Belt, S. India [3].

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