

CARBON ISOTOPE FRACTIONATION IN THE MODEL SYSTEM Fe-Ni-C AT THE HIGH PT-PARAMETERS

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The carbon is forming a lot of compounds at the wide scale of PT-parameters. Isotope fractionation of carbon at the normal temperature and pressure is well known from theoretical and experimental studies. The experimental data about carbon isotope fractionation at the high P and T are very limited. This fact makes troubles with using theoretical calculations for the interpretation of geochemical characteristics of nature samples that originate at the high temperature and pressure. However, this question is very important for number of geochemical problems, such as diamond genesis, early evolution of the Earth, genesis of meteoritic carbon etc. In this connection the obtaining of new experimental data about fractionation of carbon isotopes at the high PT-conditions is very actual. One of the way to solve this question is the investigation of isotope distribution in the model systems that using for synthetic diamond crystallization.

The metal-carbon systems are most studied from the number of solvent-carbon type systems. The first investigation of longtime carbon crystallization in Fe-Ni-C system shown that the fractionation of isotopes ^{13}C and ^{12}C between carbon phases can up to 4.5 ‰ [1].

At the present work a number of experiments was carried in the system Fe-Ni-C for the investigation of carbon isotope fractionation. Diamond was crystallized from the graphite MG-1 by TGG method ($P=5.5-6$ GPa; $T=1400^\circ\text{C}$; $\tau=40$ hrs) [2]. The metal composition was 70%-Ni and 30%-Fe. The reaction cell was hermetically soldered in the Pt ampoule for keeping gas component. MgO and ZrO bushes and washers isolated the platinum at the both sides (from the reaction volume and graphite heater).

According to chromatographic analysis [3] the gas component is represented by CH_4 . After dissolution of metal the following phases was founded: 1) the rest of the carbon source (graphite or diamond aggregates in different cases); 2) fine-crystal carbon (FCC); 3) diamond crystals; 4) graphite crystals (not for all experiments). The FCC is representing the carbon that was dissolved in the melt and isolated like phase after hardening of the system. The isotope analysis was made using technique and methodic described before [4] and results are shown in the table.

According to isotopic characteristics and relations to the initial carbon isotopic ratio all investigated phases may be divided to the two groups: 1) the phases enriched by ^{13}C (rest of carbon source – if diamond aggregate, diamond crystals and graphite crystals) and 2) the phases depleted by ^{13}C (FCC and methane). It is interesting to note that in case of multiphase crystallization (see column 1 in the table) the

Table
Carbon isotopic composition of investigated phases

Sample	$\delta^{13}\text{C}$, ‰ PDB	
	1 [1]	2
Carbon source	-28.0±0.2	-27.2±0.2
Gas component	?	-30.3±1.0
FCC	-31.3±0.3	-30.5±0.2
Graphite crystals	-26.6±0.2	—
Diamond crystals	-28.1±0.2	-27.1±0.2
Initial graphite MG-1	-27.8 ±0.4	

graphite crystals have highest ^{13}C contents and diamond have the same or slightly lighter value $\delta^{13}\text{C}$ relatively initial carbon. If the diamond is crystallized alone it have slightly higher ^{13}C content relatively initial ratio (see table).

Following processes are proceeding in the reaction volume during experiment. When the temperature and pressure are established source graphite going to the diamond aggregate. Because the touches of H_2 and O_2 are present in the initial components the Mg-wustite (interaction of melt with MgO bushes [2]) and methane will be generated in the cell. The temperature gradient is stimulated the solution of the carbon source and transport of the carbon to the crystallization zone. The crystallization either diamond or graphite or both will occur depends on the real PT-parameters in this zone. No isotope fractionation during catalitical synthesis of diamond from graphite was found [5, 6]. It is well known that the formation of methane is accompanied by enriching of CH_4 by ^{12}C [7]. The crystal phases that were grown from the melt in general case must be enriched by ^{13}C [7].

The calculations of the fractionation coefficient (α) between diamond and FCC are give the value 0.9965 (the same in all experiments). This stability seems to be evidence of the real isotopic relationship between growing diamond and Fe-Ni melt in the equilibrium with the methane at the $P=5.5-6$ GPa and $T=1400^\circ\text{C}$. According to Polyakov and Kharlashina [8] the β -factor of diamond at this PT-conditions is 1.0083. From this value the α -factor of FCC is 1.0048. It may be assumed that the carbon dissolved in the melt will have the same characteristic. An important conclusion from these calculations is that at thermodynamic condition of diamond stability (at least in the case of Fe-Ni-C system) the carbon exists in the melt in not an atom-dispersed form (β -factor this form is 1) like it was supposed by some authors [9, 10].

The nature systems are much more complicated and number of parameters is leave out of account in the experiment. However our data are show real possibility of significant carbon isotope frac-

tionation in the field of diamond stability. Value $\delta^{13}\text{C}$ of diamond is depends on the initial carbon isotope ratio and characteristics of coexisting carbon phases.

Besides, according to the presented results we can conclude:

1) The scale of carbon isotope fractionation in the Fe-Ni-C system at the PT-parameters of diamond stability is depends of coexisting carbon phases that take part in the reactions;

2) During carbon crystallization from the Fe-Ni melt the crystal phases are enriching by heavy isotope but melt and equilibrium methane are depleted by ^{13}C .

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