Carbon recycled into deep Earth: Evidence from dolomite dissociation in subduction-zone rocks

Yongfeng Zhu* Institute of Geochemistry, School of Earth and Space Sciences, Peking University, Beijing 100871, China Yoshihide Ogasawara Department of Earth Sciences, Waseda University, Tokyo 169-8050, Japan

ABSTRACT

The dolomite-dissociation textures documented here in rocks from the Kokchetav ultrahigh-pressure massif suggest that the experimentally expected dolomite dissociation happened in the subducted slabs represented by these rocks. Two reactions, magnesite = $C + MgO + O_2$, and majoritic garnet + $MgO + H_2O$ = garnet + clinochlore, recorded in carbonate inclusions and the host majoritic garnet are responsible for generation of graphite and clinochlore during the exhumation. The dolomite dissociation indicates that carbonate materials were subducted to depths of >250 km below Earth's surface. Such deep subduction evidently brings abundant carbon and carbonate into deep Earth.

Keywords: dolomite, aragonite, clinochlore, diamond, graphite, carbon recycling, subduction.

INTRODUCTION

Earth is an active planet, as evidenced by carbon recycling. Carbon in Earth's interior mainly occurs as diamond or graphite, as carbonate (magnesite, aragonite, dolomite), and as C-O-H fluid trapped in silicate minerals. Hydrocarbons devolatilized from the mantle are interpreted to be abiogenic (organically synthesized by a reaction of Fischer-Tropsch type in the mantle or delivered by meteorites and comets to early Earth) and/or biogenic (recycled by subduction, Sugisaki and Mimura, 1994). Unlike the situation on Venus, carbon on Earth's surface is returned to Earth's interior by subduction, which probably is the main reason for the generation of life, as subduction allowed carbon and other light elements as well as water to be present in Earth's mantle (Ernst et al., 1998; Kerrick and Connolly, 2001; Lecuyer et al., 2000; Molina and Poli, 2000). Subducted carbon corresponds to $\sim 10 \text{ wt\%}$ (Javoy, 1997; Lecuyer et al., 2000) or 30 wt% (Canil et al., 1994) of the carbon reservoir in the primitive mantle. Geochemistry data (Nishio et al., 1998; Ray et al., 1999) demonstrated that ancient carbonate rocks have been transported into the mantle through subduction zones. Lindsay and Brasierb (2002) explained the coincidence of carbon isotopic excursions and tectonism observed in West Australian basins by the sequestration of carbon during ocean closure with organic-rich passive margin sediments containing isotopically light carbon subducted into and stored in the mantle. They argued that the evolution of both the atmosphere and the biosphere may have been driven forward by planetary evolution, implying that biospheric evolution has largely been driven by the dynamo of Earth's tectonism.

The aragonite + magnesite assemblage, the major carbon-bearing phase in deep Earth, is stable to 28 GPa at 1000 °C (Liu and Lin, 1995). However, no aragonite + magnesite assemblages have been reported in the geologic literature; this limits the subduction of carbonate materials to depths <240 km (<7.85 GPa at 1000 °C), based on the phase boundary of the dolomite-dissociation reaction determined experimentally by Sato and Katsura (2001). This paper reports the discovery of dolomite-dissociation textures in the Kokchetav dolomite marble, which suggests that carbon and/or carbonate from Earth's surface has been subducted to depths >250 km (possibly to 300 km) and has returned to the surface.

SAMPLE DESCRIPTIONS

Rocks from the Kokchetav ultrahigh-pressure (P) massif have been extensively studied (e.g., Dobrzhinetskaya et al., 1994; Shatsky et al., 1995; Zhang et al., 1997). The Kokchetav ultrahigh-P massif consists of several metamorphosed lithotectonic units with Precambrian protolith ages, overlain by Devonian volcaniclastic rocks and Carboniferous-Triassic shallow-water and lacustrine deposits, and intruded by Ordovician granites and gabbros of an island-arc series (Dobretsov et al., 1995; Maruyama and Parkinson, 2000). Metamorphic diamonds have been found as inclusions in many kinds of minerals, including garnet and clinopyroxene from gneiss, eclogite, and dolomite marble (e.g., Sobolev and Shatsky, 1990; Ogasawara et al., 2000). Dolomite marble crops out in the Kumdy-kol region of the central part of the Kokchetav ultrahigh-P massif, where the marble coexists with eclogites in gneiss (Zhang et al., 1997; Ogasawara et al., 2000). These rocks were subducted to the mantle from continental crust levels, underwent ultrahigh-P metamorphism, and were exhumed to Earth's surface.

Dolomite (>50 vol%) is the major phase in dolomite marble; other carbonate minerals (aragonite and calcite) and carbon phases (diamond and graphite) occur only in trace amounts (<0.1%). Garnet and clinopyroxene account for >40 vol% of the dolomite marble. Most garnet and some clinopyroxene contain microdiamond inclusions. Garnet coronas, formed during retrograde metamorphism, consist chiefly of zoisite, hornblende, and phlogopite.

RESULTS

All textural relationships described here were investigated employing a JEOL Superprobe 8900 by using the backscattered-electron (BSE) image mode, combined with wavelength-dispersive X-ray spectroscopy (WDS). Garnet contains abundant carbonate inclusions of various shapes and sizes. Radial fractures generally formed in garnet around the carbonate inclusions (Fig. 1). Dolomite is the most common mineral phase in these inclusions, and it is typically replaced by a calcite + clinochlore in the rim or along boundaries between dolomite and the host garnet (Fig. 1A). Relict dolomite contains diamond inclusions, whereas the calcite + clinochlore assemblages coexist with graphite (Fig. 1B). In some samples, euhedral carbonate inclusions consisting of calcite with minor amounts of relict dolomite are found in garnet, and clinochlore occurs on the boundary between the inclusion and garnet (Fig. 1C). Where the clinochlore, coexisting with calcite and graphite, was in contact with garnet, a kind of sieve texture formed in the garnet (Fig. 1D). In rare cases, a transformation texture from aragonite to calcite is preserved in a carbonate inclusion (Fig. 1E); in this texture, calcite grows around aragonite and coexists with clinochlore. Aragonite obviously differs from calcite in the BSE image (Fig. 1E), owing to aragonite's high density (2.97 g/cm^3) compared to calcite $(2.73 \text{ g/cm}^3).$

Minerals were analyzed using a JEOL Superprobe 8900 with WDS. The analysis conditions were 15 kV, 1 nA, a spot diameter of 1 μ m, and a counting time of 40 s for every element. Representative compositions for all phases in the studied system are listed in Table 1. Garnet contains majoritic components (0.16–1.39 mol% majorite; average = 0.71%; n = 81), as shown by constant Si concentrations higher than 3.0 p.f.u. (per formula unit) (3.005–3.042 p.f.u.; average = 3.022 p.f.u.). However, garnet in the reaction zone (~10 μ m in width)

^{*}Corresponding author. E-mail: yongfeng@eyou.com.

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Figure 1. Backscattered-electron images show dolomitedissociation textures and radial fractures in garnet. A-B: Dolomite inclusion decomposed into calcite and clinochlore in rim. Diamonds occur in garnet; one euhedral diamond grain occurs in relict dolomite. Irregular graphite grains are distributed in garnet fractures (as shown in A) and coexist with clinochlore and calcite. C-D: Euhedral calcite inclusion (with trace amounts of relict dolomite) in garnet with well-developed radial fractures; clinochlore occurs in boundary between calcite and garnet. Sieve texture shown in D developed in garnet in contact with clinochlore, which suggests chemical reaction between them (see text for details). E: Aragonite is replaced by calcite in rim; calcite coexists with clinochlore. Phases: cc-calcite, CCH-clinochlore, dia-diamond, dol-dolomite, G-graphite, grt-garnet. Scale bars in A and C represent 100 µm; those in B and D represent 10 µm, and that in E represents 50 µm.

has relatively lower Si content (2.977–3.003 p.f.u.; average = 2.992 p.f.u.; n = 28) but higher Mg content (1.194–1.242 p.f.u.; average = 1.221 p.f.u.) relative to the majoritic garnet (Mg = 1.107-1.218 p.f.u.; average = 1.158; Fig. 2A, Table 1).

Aragonite is almost pure $CaCO_3$ in composition. Calcite, however, contains MgO (to 3 wt%) as well as trace amounts of FeO (Fig. 2B; Table 1). Calcite exhibits a relatively large compositional variation and differs from aragonite by higher Mg and Fe concentrations.

DISCUSSION

Most subducted slabs contain carbonate material, which could be introduced into the deep mantle. In fact, this process has happened, as indicated by the occurrence of diamond-bearing dolomite marble coexisting with eclogites in the Kokchetav ultrahigh-P massif. Except for the high-P requirement for formation of aragonite + magnesite as the product of dolomite dissociation, another possible reason for the rare occurrence of the aragonite + magnesite assemblage in natural rocks is the relative instability of magnesite, which can be easily transformed to OH-bearing silicates when the magnesite is raised from the deep mantle into shallow-level, water-bearing environments. Clinochlore coexists with calcite (Fig. 1, D and E). Calcite is the low-P (high volume) variant of aragonite, and the transformation from aragonite to calcite apparently is responsible for the radial fractures developed around the carbonate inclusions in garnet. The evidence for this transformation implies that aragonite coexisted with the precursor of clinochlore.

The products of dolomite dissociation should be aragonite and magnesite (Liu and Lin, 1995). In the mantle environment, magnesite can decompose to graphite or diamond + magnesium oxide according to the reaction (Wang et al., 1996)

$$MgCO_3 = C + MgO + O_2.$$
(1)

The occurrence of graphite in garnet fractures (Fig. 1A) and its coexistence with clinochlore (Fig. 1, B and D) suggest that reaction 1 is responsible for conversion of some magnesite to form graphite in dolomite marble and that some magnesite has been changed to clinochlore. The following reaction of majoritic garnet combining with certain components to form garnet and clinochlore is appropriate to the studied system and may be responsible:

$$\begin{aligned} & f[(Ca, Fe)_{3-n}Mg_n][Al_{2-m}Si_{3+m}]O_{12} + MgO \\ &+ 8yH_2O + 0.5(xm + 4y)Al_2O_3 \\ &+ (6y - xm)SiO_2 \\ &= x[(Ca, Fe)_{3-n-z}Mg_{n+z}][Al_2Si_3]O_{12} \\ &+ y(Mg_{10}Al_2)[Al_2Si_6]O_{20}(OH)_{16}, \end{aligned}$$
(2)

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where y = 0.1(1 - xz), m = 0.005-0.042, n = 1.107-1.242, and z = 0.024-0.087 (based on the composition of the studied garnet; Fig. 2A; Table 1). For 1 mol of MgO and majoritic garnet with a constant x value, equation 2 is fixed. For example, with m = 0.04 and z = 0.05, 1 mol MgO (corresponding to 1 mol magnesite) reacts with 2 mol majoritic garnet (x = 2) to produce 2 mol new garnet and 0.09 mol clinochlore and consumes 0.72 mol H₂O, 0.22 mol Al₂O₃, and 0.46 mol SiO₂ from fluid (supplied from outside the system). These reactions only involve the garnet in contact with clinochlore, as evidenced by the sieve texture developed in garnet in the reaction zone (Fig. 1D) and the compositional difference of garnet in the reaction zone compared to that elsewhere in the rock (Fig. 2A). The majoritic garnet (away from the reaction zone) is Si rich but Mg poor relative to the new-formed garnet in the reaction zone.

The characteristics of fluid inclusions in metamorphic diamonds from Kokchetav eclogites (De Corte et al., 2000) and two nanocrystalline magnesite inclusions in diamond from Kokchetav gneiss suggest that diamond grows in a hydrous fluid (Dobrzhinetskaya et al., 2001). Si- and Al-bearing C-O-H fluid at a depth of ~150 km played a significant role in diamond origin, as shown by the fact that diamond formed together with Si- and Al-bearing phases such as phengite and phlogopite (Stockhert et al., 2001). Such a fluid would provide H₂O as well as Al₂O₃ and SiO₂ to the studied system in which reactions 1 and 2 occurred.

Diamond coexists with dolomite, whereas graphite occurs in the calcite + clinochlore assemblage (Fig. 1, A and B). These distinct associations rule out any relationship to the diamond transformation. Textural relationships shown in Figure 1 therefore indicate that reaction 1 was responsible for the formation of graphite coexisting with clinochlore in the dolomite marble.

TABLE 1. REPRESENTATIVE COMPOSITIONS OF THE CARBONATE AND SILICATE MINERALS

	Dol	dol	arag	arag	CC	СС	CCH	CCH	grt-M	grt-M	grt	grt
Oxides												
SiO ₂	0	0	0.01	0.03	0.07	0.20	31.96	32.61	41.68	41.70	41.29	41.45
TiO ₂	0	0	0	0	0	0	0	0	0.25	0.21	0.31	0.25
Al ₂ O ₃	0.02	0.05	0	0.02	0	0.03	16.93	17.61	22.62	22.75	23.10	23.51
MgO	19.65	19.37	0.64	0.23	2.64	3.09	29.54	29.70	11.04	10.90	11.29	11.36
MnO	0.18	0.29	0.32	0.09	0.19	0.10	0.11	0.16	0.64	0.77	0.64	0.69
FeO	1.41	1.94	0.25	0	0.51	0.44	4.78	5.84	5.99	5.83	5.81	6.08
CaO	32.13	30.96	55.42	55.49	52.34	52.24	0.40	0.21	17.82	17.51	17.58	17.47
Na ₂ O	0	0.01	0	0	0	0	0.01	0.02	0	0.02	0.03	0.06
Total	53.39	52.62	56.64	55.86	55.75	56.10	83.73	86.15	100.04	99.69	100.05	100.87
O cation	6	6	6	6	6	6	28	28	12	12	12	12
Si	0	0	0	0.002	0.003	0.010	6.284	6.259	3.034	3.042	3.003	2.992
Ti	0	0	0	0	0	0	0	0	0.014	0.012	0.017	0.014
AI	0.001	0.003	0	0.001	0	0.002	3.922	3.983	1.941	1.956	1.980	2.000
Mg	1.350	1.353	0.047	0.017	0.194	0.225	8.658	8.498	1.198	1.185	1.224	1.222
Mn	0.007	0.012	0.013	0.004	0.008	0.004	0.018	0.026	0.039	0.048	0.039	0.042
Fe	0.054	0.076	0.010	0	0.021	0.018	0.786	0.937	0.365	0.356	0.353	0.367
Ca	1.587	1.554	2.928	2.974	2.770	2.731	0.084	0.043	1.390	1.368	1.370	1.351
Na	0	0.001	0	0	0	0	0.004	0.007	0	0.003	0.004	0.008
Total	2.999	2.999	3.000	2.998	2.997	2.989	19.757	19.754	7.979	7.969	7.992	7.997
Note: do	I = dolomite,	arag =arago	nite, cc =cal	cite, CCH =	clinochlore, gi	rt-M = majori	itic garnet, grt	= garnet in t	he reaction zoi	ne.		

Majoritic garnet is stable in a higher pressure environment relative to normal garnet, according to experimental studies (Irifune et al., 1986; Ono and Yasuda, 1996) as well as geologic observations (e.g., van Roermund et al., 2001). The new-formed garnet contains lower Si (average 2.99 p.f.u.) relative to the majoritic garnet (average 3.02 p.f.u., Fig. 2A). This difference implies that the reaction between the magnesite and majoritic garnet happened as the rocks moved into a low-*P* environment (retrograde metamorphism). This pressure-temperature (*P*-*T*) history is also evidenced by the coexistence of garnet, clinochlore, and graphite in the reaction zone (Fig. 1D). Thus, reaction 2 happened in the graphite-stability field. Reaction 1 probably happened at the same *P*-*T* conditions, as shown by the coexistence of graphite and clinochlore. This agreement therefore implies that magnesite decomposition occurred in the graphite-stability field and that the aragonite + magnesite assemblage was stable during the exhumation until it reached a depth of <90 km (field B in Fig. 3), corresponding to the *P*-*T* space where diamond partly transformed to graphite (Dobrzhinet-skaya et al., 2001).

The exact depth to which the carbon and/or carbonate has been brought down by the subducted slab depends on the temperature, as shown in Figure 3, and relates to the geothermal gradient in which the slab moved. The coldest geothermal gradient in the continental lithosphere mantle is found in the African Kaapvaal craton (Finnerty and Boyd, 1987). This geothermal gradient is \sim 50 mW/m, and fails to pass through the dolomite-dissociation curve. The reference geothermal line



Figure 2. Compositional variations for garnet (region A) and aragonite and calcite (region B) (pfu—per formula unit).



Figure 3. Pressure-temperature (*P*-*T*) space for subducted dolomite marble. Dolomite-dissociation reaction is based on Sato and Katsura (2001); diamond-graphite boundary is based on Kennedy and Kennedy (1976). African Kaapvaal craton reference is based on Finnerty and Boyd (1987). Field A represents *P*-*T* space where dolomite-dissociation reaction happened, whereas field B corresponds to *P*-*T* conditions for graphite and clinochlore generation at exhumation.

with 40 mW/m passes through the dolomite breakdown at \sim 9.62 GPa (1290 °C, Fig. 3), according to the dolomite-dissociation reaction determined experimentally by Sato and Katsura (2001). This temperature is close to the extended wet solidus for mantle rock and therefore represents the highest temperature to which the subducted slab could have been subjected. Progressive metamorphism of the Kokchetav dolomite marble should have happened at a temperature lower than 1290 °C, as evidenced by the complete absence of partial-melting phenomena during exhumation. Therefore, the geothermal gradient where the Kokchetav dolomite marble was subducted was lower than 40 mW/m. During subduction, the dolomite marble reached pressures exceeding the dolomite-dissociation boundary. The P-T space for dolomite dissociation shown as A in Figure 3 therefore represents the possible P-T space that the carbonate materials have reached. Therefore, subduction brought abundant carbon and carbonate to depths of >250 km. As a result, the surface carbon and/or carbonate were recycled into deep Earth. This process is essential for Earth dynamics as well as for life generation, because such deep subduction keeps the carbon preserved in Earth's system and makes the planet Earth different from Venus, where most of the carbon has been expelled into the atmosphere.

CONCLUSIONS

The Kokchetav dolomite + aragonite (+ calcite + clinochlore) + diamond (+ graphite) assemblage as inclusions in majoritic garnet with well-developed radial fractures indicates prograde metamorphism at P > 7.85 GPa. Textural relations preserved in the Kokchetav dolomite marble suggest that the subducted carbonate rocks passed through the dolomite-dissociation reaction boundary and reached depths of >250 km below Earth's surface. Such a deep subduction process has significant implications for carbon recycling, in which abundant carbon was brought deep into Earth, a key factor to make planet Earth suitable for life.

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