



Chemical compositions of carbonate minerals from Bayan Obo, Inner Mongolia, China: implications for petrogenesis

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Received 24 September 2002; accepted 9 September 2003

Abstract

Carbonate rocks at the Bayan Obo giant rare earth element (REE)–Nb–Fe ore deposit, Inner Mongolia, China are divided into four categories: sedimentary limestone and dolostone (H8_s), deformed mineralized coarse-grained dolomite marble (H8_c) and fine-grained dolomite marble (H8_f), and carbonatite dikes (D), based on their texture, mineral assemblage and geological occurrence. Although the sedimentary carbonate rocks (H8_s) show weak deformation north of the Kuangou fault zone, this unit is not metamorphosed, which occurs together with quartz sandstone, sandstone and shale, comprising the Middle Proterozoic Bayan Obo Group. In contrast, the mineralized fine-to-coarse-grained dolomitic marbles (H8_f and H8_c) that occur to the south of the Kuangou fault zone are sheared, deformed and metamorphosed. The H8_c consist mainly of dolomite associated with apatite, magnetite, minor pyrochlore, and sodic amphibole. Dolomite in the main ore-bearing unit (H8_f) occurs with magnetite, monazite, bastnaesite, and parisite. Fine-grained monazite occurs as fracture fillings in dolomite. Electron microprobe data show that dolomite and/or ankerite in the ore-bearing rocks (H8_c and H8_f) are similar to carbonatite dikes (D) occurring within 3.5 km northeast of the deposit. These carbonates contain high MnO (>0.50 wt.%) and SrO (>0.15 wt.%) as is typical of carbonatites; this contrasts with very low MnO and SrO (<0.1 wt.%) in typical sedimentary limestone and dolostone (H8_s). The variation of MnO and SrO content in dolomite from the ore-bearing dolomite marbles may be interpreted by fractional crystallization of a carbonatitic magma, which results in REE enrichment in the evolved residual magma. This interpretation is consistent with their field relations, trace element geochemistry, stable isotopes, and ⁸⁷Sr/⁸⁶Sr ratios. The MnO and SrO contents of carbonate minerals may be used as important chemical characteristics to identify their genesis. MnO and SrO contents in carbonate minerals higher than 0.15 wt.% are used as indicators to distinguish carbonatite from sedimentary carbonate rocks. These criteria together with fenitization may be taken as critical signatures to separate carbonatites from sedimentary carbonate rocks, even when both were metamorphosed to marbles.

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Keywords: Carbonate minerals; Dolomite; Dolomite marble; Carbonatite; Bayan Obo

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1. Introduction

The Bayan Obo giant REE–Nb–Fe ore deposit of Inner Mongolia, China (110°E, 41°48' N) is well known as the largest rare earth element (REE) mineral deposit in the world (Fig. 1). Its origin has been a

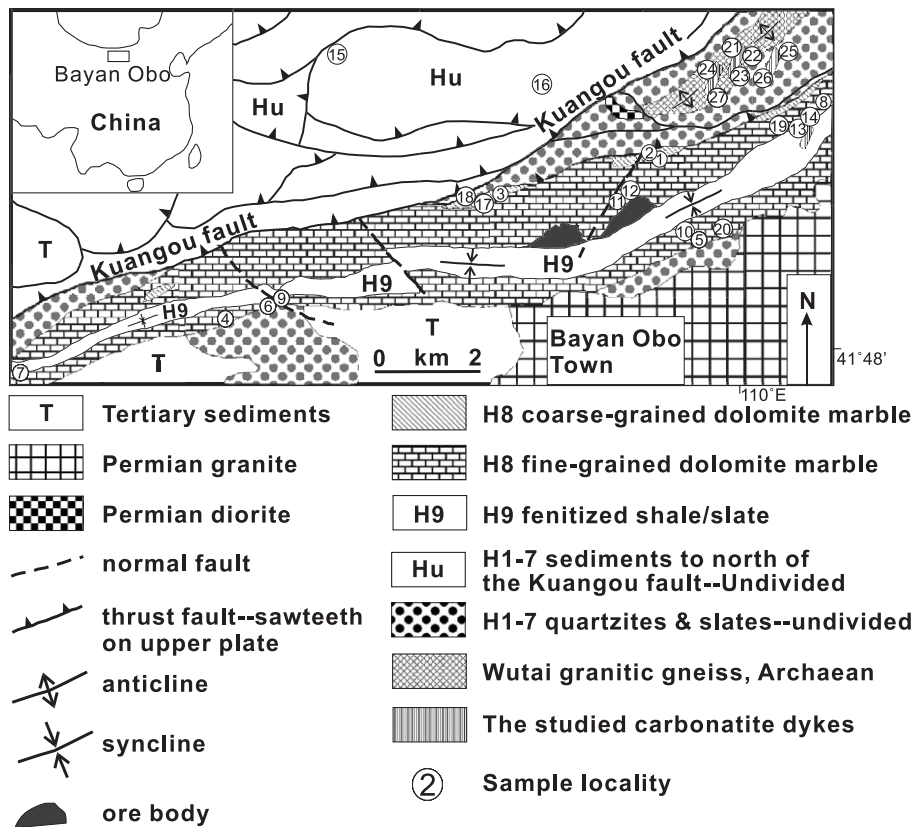


Fig. 1. Simplified geological map of the Bayan Obo area, Inner Mongolia, China (modified after Drew et al., 1990; Le Bas et al., 1997). The Bayan Obo area can be geologically divided into two parts: (1) north of the Kuangou fault zone, the lithology is represented by an unmineralized and weakly deformed limestone, dolostone (H8), quartz sandstone (H1, H2, H4, H6 and H7) and shale (H3, H5) sequence undivided and shown as Hu in the map, belonging to the Middle Proterozoic Bayan Obo Group (Institute of Geochemistry, Chinese Academy of Sciences, 1988), where no mineralization occurs; (2) south of the Kuangou fault zone, the lithology in the mine region is represented by the recrystallized marbles (H8_c and H8_f) and slate (H9), which is deformed, sheared and locally mineralized. Although the ore body shown on the map was defined in terms of grade of Fe ore, at 35 wt.% iron oxide, the ore-bearing dolomite marbles (H8_c and H8_f) constitute the REE ore with average REE₂O₃ grade at 1.5 wt.% (Institute of Geochemistry, Chinese Academy of Sciences, 1988). Sample locality (see Table 1 for the chemical data of carbonate minerals): (1) 88/150 (field number); (2) 88/152; (3) B241; (4) 90/62; (5) B222; (6) 90/63; (7) 88/157; (8) 90/58; (9) 90/64; (10) 88/117; (11) 88/148; (12) 88/147; (13) 88/142; (14) 88/141; (15) 90/65; (16) Y8/230; (17) B239; (18) B240; (19) B253; (20) B256; (21) CD7(2); (22) CD9(1); (23) CD14(1); (24) CD11(1); (25) 93/149; (26) 90/42; (27) CD15(2); (28) LCS-05 (this sample is taken from Lachaoshan, 50 km south of the Bayan Obo deposit, beyond the scope of map).

disputed topic since it was discovered in 1927 (Li, 1959; Zhou et al., 1980; Meng, 1982; Bai and Yuan, 1985; Institute of Geochemistry, Chinese Academy of Sciences, 1988; Drew et al., 1990; Chao et al., 1992, 1993, 1997; Le Bas et al., 1992, 1996, 1997; Yuan et al., 1992; Zhang et al., 1995; Wu et al., 1996; Smith et al., 1999, 2000; Smith and Henderson, 2000). The geology of the deposit has been described in detail in previous papers or books (Institute of Geochemistry, Chinese Academy of Sciences, 1988; Drew et al.,

1990; Zhang et al., 1995; Bai et al., 1996; Chao et al., 1997; Le Bas et al., 1997). The most prominent feature is that the ore-bearing dolomite marbles, which extend about 16 km long from west to east and are 1–2 km wide (Fig. 1), contain remarkably high REE. The dolomite marble body contains huge mineral reserves: 48 million tonnes of REE oxides (average grade 6 wt.%) plus 1 million tonnes of Nb (average grade 0.13 wt.%) and 1500 million tonnes of Fe (average grade 35 wt.%) (Drew et al., 1990). The

REE in the deposit mainly occur as REE fluorocarbonate series minerals, (e.g. bastnaesite [(Ce,La)(CO₃)F], parisite [(Ce,La)₂Ca(CO₃)₃F₂], cordylite [CaBa(Ce,La)₂(CO₃)₄F₂], huanghoite [BaCe(CO₃)₂F], and cebaite [Ba₃Ce₂(CO₃)₅F₂]) and monazite [(Ce,La,Nd,Th)PO₄] hosted in dolomite marbles (Institute of Geochemistry, Chinese Academy of Sciences, 1988; Yuan et al., 1992; Zhang et al., 1995, 2002; Yang et al., 1999a, 2000a). The sedimentary versus igneous origin of the ore-bearing dolomite marbles has become an essential issue in the study of genesis of the deposit.

Many researchers considered the mineralized dolomitic marbles to be sedimentary (Li, 1959; Meng, 1982; Tu et al., 1985; Chao et al., 1992, 1993, 1997; Wang et al., 1994), whereas others suggested them to be of carbonatitic origin (Zhou et al., 1980; Meng, 1981; Bai and Yuan, 1985; Liu, 1986; Le Bas et al., 1992, 1996, 1997; Yuan et al., 1992; Bai et al., 1996; Yang et al., 1999b, 2000a,b, 2003). Zhou et al. (1980) suggested that the dolomite marble was a dolomite carbonatite intrusion based on intrusive relations, mineralogy, and petrochemistry. Bai and Yuan (1985) considered it formed through exhalative carbonatitic sedimentation in marine environment in Middle Proterozoic, then was metamorphosed to marble in terms of various geological and geochemical evidence. In light of systematic mineralogical and geochemical studies on a coarse-grained calcite carbonatite dike and its comparison with the ore-bearing dolomite marble within the deposit, Le Bas et al. (1992) proposed that the marble was originally a carbonatitic tuff that then underwent dolomitization. However, new evidence of intrusive contacts and fenitization found in the footwall and hanging wall of the dolomite marble convinced Le Bas et al. (1996, 1997) that it was originally a polyphase carbonatite intrusion, which was emplaced into the Bayan Obo Group as sills (Zhou et al., 1980; Meng, 1981; Liu, 1986) during the Middle Proterozoic (Yuan et al., 1992) and fenitized wall rocks next to the contacts to develop potassic feldspar and phlogopite rocks (the protolith was shale) in the hanging wall and metasomatic rocks containing sodic amphibole and albite (the protolith was sandstone) in the footwall (Drew et al., 1990; Yuan et al., 1992; Yang et al., 1994, 2000b). These packages were deformed, sheared and folded owing to Caledonian subduction of the Mongolian

oceanic plate, 100 km from north, beneath the North China (Chao et al., 1993, 1997), and then intruded by a Permian granite complex (Drew et al., 1990; Le Bas et al., 1997).

All these above viewpoints were based on geological, mineralogical and geochemical evidence acquired from various investigations, reflecting genetic complexity of the dolomite marbles. However, few data were available in previous studies about the chemistry of carbonate minerals from different geological occurrences at Bayan Obo area. The chemical features of carbonates can provide important information about their origin (Hogarth et al., 1985; Barker and Nixon, 1989; Le Bas and Srivastava, 1989; Dickson, 1990; Clarke et al., 1994; Dawson et al., 1996; Hornig-Kjarsgaard, 1998; Zaitsev and Chakhmouradian, 2002). This contribution employs an electron probe microanalyzer (EPMA) systematically to determine the chemical composition of carbonate minerals from the ore-bearing dolomite marbles, sedimentary limestone and dolostone (H8) of the Proterozoic Bayan Obo Group, and carbonatite dikes in the vicinity of the Bayan Obo ore deposit. These data are used to evaluate the genesis of the ore-bearing dolomite marbles. This paper proposes that SrO and MnO contents of carbonate minerals can be used as important parameters to distinguish sedimentary carbonates and carbonatites.

2. Petrological characteristics of carbonate rocks

Since the Bayan Obo REE–Nb–Fe ore deposit is principally hosted in dolomite marbles, this study focuses on those units. The mineralized dolomite marbles are distributed along east–west direction about 16 km long, 1–2 km wide, together with the quartzite, quartz sandstone and slate of the Middle Proterozoic Bayan Obo Group. The dolomite marbles occur within a series of thrust slices dipping to north, and the whole area is intruded and underlain by a Permian granite, which crops out mainly to the south of the ore deposit (Drew et al., 1990; Le Bas et al., 1997, see Fig. 1). More than 30 carbonatite dikes occur within 3.5 km northeast of the East ore body of the deposit. Most of the dikes are 1 to 2 m wide, some thinner, striking NNE–SSW to NNW–SSE, which can be traced on surface or in exploration trenches.

The longest one was exposed up to 60 m along its strike (Yang et al., 2000a, 2003). These dikes cross cut a thrust inlier of the Wutai Group granitic gneiss and Archaean migmatite in the Kuangou anticline and its southern limb (units H1 to H9 of the Proterozoic Bayan Obo Group) (Le Bas et al., 1992, 1997; Yang et al., 1994, 2000a,b, 2003). New U–Pb zircon ages (1.9–2.0 Ga) for the Wutai Group gneiss reveal a Paleoproterozoic (Wang et al., 2002) rather than Archaean age as previously considered (Zhou et al., 1980; Drew et al., 1990; Zhang et al., 1995; Chao et al., 1997). Since the gneiss is tonalitic to syenitic in composition, Wang et al. (2002) suggest that it is more appropriate to term it a basement complex.

These dikes fenitized their country rocks on both sides of the contact zones, producing fenites characterized by alkali amphibole, albite and phlogopite. Most of the carbonatite dikes are calcic in chemical composition, and three are rich in REE. Only three dikes belong to the group of carbonatites termed magnesio-carbonatite by Woolley and Kempe (1989) and Le Maitre et al. (2002). As previously, most of the dikes are deformed, sheared and locally recrystallized, although their geochemical signatures are preserved (Le Bas et al., 1992; Yang et al., 1994, 2000a, 2003). In addition, there are also several carbonatite dikes recently discovered in the southern limb of the Bayan Obo syncline. Bai and Yuan (1985) noticed that the whole-rock geochemistry of the ore-bearing dolomite marble was obviously different from the sedimentary carbonate rocks (H8) of the Proterozoic Bayan Obo Group north of the Kuangou fault zone, but similar to those of carbonatite dikes (Le Bas et al., 1992, 1996, 1997; Yuan et al., 1992; Bai et al., 1996; Yang et al., 2000a, 2003). Based on geological occurrence and distribution, rock fabrics and mineral paragenesis, the carbonate rocks at Bayan Obo have been divided into four major types in this study in order to compare the chemical attributes of their major rock-forming minerals and petrogenetic significance.

2.1. Sedimentary limestone and dolostone (H8_s)

These rocks are distributed north of the Kuangou fault zone and have medium to thick bedding and massive structure, medium-to-coarse-grained, but occasionally fine-to-medium-grained texture. Calcite and dolomite contain minor opaques (hematite?) and

clays. Curved or sutured carbonate grain boundaries are evident. The limestone and dolostone typically contain fine-grained detrital quartz. There are some late-stage quartz veins and veinlets cutting the rocks. Deformation and recrystallization are relatively weak in these sedimentary carbonate rocks together with quartz sandstone, sandstone and shale (Middle Proterozoic Bayan Obo Group).

2.2. Coarse-grained dolomite marble (H8_c)

This unit hosts REE ore bodies (Fig. 1), and is intermittently distributed along the lower part of the northern limb of the Bayan Obo secondary syncline south of the Kuangou fault zone, and is strongly deformed and sheared. The H8_c dolomite marble has a coarse-grained (grain-size >1 mm) banded and massive structure with a euhedral to subhedral texture. It mainly consists of dolomite, apatite, magnetite, with minor pyrochlore and sodic amphibole (Fig. 2a). Magnetite and apatite have been drawn out into granular schlieren and lineation. This lineation results from shearing, stretching and deformation interpreted to be related to the regional thrusting and folding. The dolomite marble contains minor acicular arfvedsonite and magnesio-riebeckite; the sodic amphiboles in the marble increase towards the footwall contact. Dolomite has euhedral to subhedral shape and the boundaries between grains are normally planar, typically constituting triple junctions similar to those in carbonatites elsewhere (Le Bas and Srivastava, 1989) or recrystallized marbles. The apatite content of the marble is variable, and most crystals are subprismatic to elongate, and have an orientation paralleling the contact zone between the marble and the H4 arkosic quartzite that is a unit of the Proterozoic Bayan Obo Group. In addition, a few phlogopite veins and veinlets cut the marble. The coarse-grained dolomite marble 1.25 km to the northwest of the Main ore body (Fig. 1) demonstrates an intrusive contact to the arkosic quartzite (H4 unit), resulting in fenitization of the quartzite characterized by magnesio-riebeckite and albite (Fig. 2b).

2.3. Fine-grained dolomite marble (H8_f)

This unit is continuously distributed along the upper part of the northern limb of the Bayan Obo

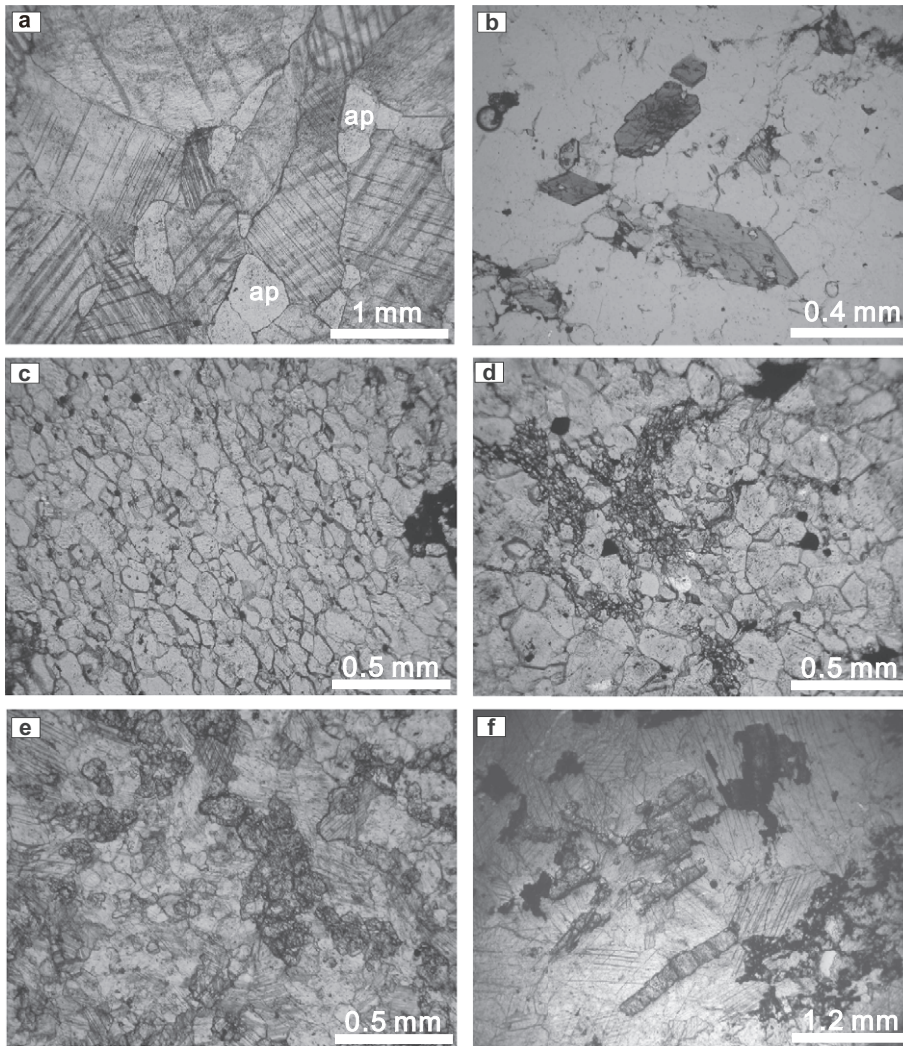


Fig. 2. Photomicrographs of carbonate rocks from the Bayan Obo area: (a) coarse-grained dolomite marble, containing rounded, elongate and irregular apatite (A, low relief, pale), sample B241, plane polarized light (PPL); (b) fenitized quartzite (unit H4 of the Bayan Obo Group), euhedral blue magnesio-riebeckite replaces quartz both along fractures and boundaries between grains, sample B242, PPL; (c) fine-grained dolomite marble, mosaic texture with triple junctions, deformed and sheared, sample B256, PPL; (d) fine-grained dolomite marble, fine monazite clusters (high relief) are distributed along fractures or between dolomite grains, sample B253, PPL; (e) fine-grained carbonatite from a carbonatite dike, fine-grained bastnaesite $[(\text{Ce},\text{La})(\text{CO}_3)\text{F}]$ and parisite $[(\text{Ce},\text{La})_2\text{Ca}(\text{CO}_3)_3\text{F}_2]$ (darker brown and high relief) occur in aggregates that are interlocked with calcite, sample 93/149, PPL; (f) coarse-grained carbonatite consists mainly of calcite, containing large euhedral to subhedral bastnaesite grains, sample CD11(1), PPL.

secondary syncline south to the Kuangou fault zone, about 16 km long from east to west and also distributed along the southern limb of the fold, constituting the major ore-hosted rock (Fig. 1). This REE ore has an average RE_2O_3 content of 1.5 wt.% (Institute of Geochemistry, Chinese Academy of

Sciences, 1988). The dolomite marble (grain-size < 1 mm) has discontinuous lenses, massive to banded structure, with euhedral to subhedral mosaic texture (approximate 120° triple junctions) typical of carbonatites and recrystallized marbles elsewhere; it is composed mainly of dolomite and ankerite, and also

contains variable magnetite, monazite, bastnaesite, barite and fluorite (Fig. 2c). Dolomite is usually euhedral with a mosaic of triple junctions, and untwinned. Very fine monazite, bastnaesite and fluorite bands occur in the dolomite marble, together with magnetite bands, demonstrating so-called banded structure and schlieren structure, which show strong lineation interpreted to be associated with regional thrusting and folding (Chao et al., 1993; Le Bas et al., 1997). Later clusters of fine-grained monazite fill microfractures in dolomite (Fig. 2d), indicating that this monazite may have crystallized from hydrothermal fluids. The fine-grained dolomite marble in the southern limb of the Bayan Obo syncline (Fig. 1) also contains banded magnetite, monazite, bastnaesite and fluorite aggregates, suggesting that the fold was formed after the major REE mineralization.

Field relations and thin section investigations indicate that the fine-grained dolomitic marble (H8_f) is later than the coarse-grained dolomitic marble (H8_c) based on intrusive relationships (Zhou et al., 1980; Meng, 1981; Liu, 1986) that were confirmed by the authors (Yang et al., 1994; Le Bas et al., 1996, 1997). The coarse-grained variety was locally recrystallized to form the fine-grained equivalent, e.g. a large dolomite grain surrounded and/or replaced by numerous smaller dolomite grains. Chao et al. (1993) emphasized that the fine-grained dolomite marble formed by deformation and recrystallization of the coarse-grained dolomite marble, and both of them underwent more than two regional metamorphic episodes (Chao et al., 1993, 1997). The whole-rock geochemistry reveals that the fine-grained variety is more evolved than the coarse-grained marble (Le Bas et al., 1997; Yang unpublished data), compared to C1 and C2 carbonatites (Le Bas, 1999). For example, the H8_f contains higher REE, Mn, Ba, F, lower Sr, and heavier $\delta^{18}\text{O}$ and $\delta^{13}\text{C}$ ratios, compared to the H8_c; both of them show relatively low $^{87}\text{Sr}/^{86}\text{Sr}$ ratios (<0.704) (cf. Le Bas, 1981, 1989, 1999, for discussion on differentiation of carbonatites).

2.4. Calcite carbonatite and dolomite carbonatite (D)

There are more than 30 carbonatite dikes occurring within 3.5 km northeast of the East ore body of the deposit, cross-cutting a thrust inlier of the Paleopro-

terozoic granitic gneiss complex (Wang et al., 2002) and the Wutai Group migmatite (Archaean) in the Kuangou anticline and its southern limb consisting of sedimentary rocks (units H1 to H9) of the Proterozoic Bayan Obo Group. Several carbonatite dikes have recently been discovered in the southern limb of the Bayan Obo syncline. It is significant that these dikes have metasomatized the country rocks on their contact zones, producing fenites characterized by the presence of sodic amphiboles, albite, and phlogopite (Le Bas et al., 1992; Yang et al., 1994, 2000b). Whole-rock XRF analyses and EPMA data show that most of the carbonatite dikes are calcic in chemical composition. Three are also rich in REE; only three analyses indicate magnesiocarbonatites according to the nomenclature by Woolley and Kempe (1989) and Le Maitre et al. (2002).

Carbonatite dikes can also be grouped into coarse-grained and fine-grained based on grain size, which are similar in trace element and REE geochemistry to the ore-bearing dolomite marbles (Le Bas et al., 1992, 1996, 1997; Yang et al., 2003). The fine-grained carbonatite dikes (D_f) are locally porphyritic. Since the dikes were affected by later tectonism, they were strongly deformed and sheared, which is demonstrated by the alignment of rock-forming minerals, the curved cleavage and undulatory extinction of large calcite phenocrysts. A typical fine-grained carbonatite dike was previously described (Yang et al., 2000a), which contains extremely high REE (average REE concentration of 8 wt.% for six samples, see Yang et al., 2003). The carbonatite dike is composed mainly of calcite as euhedral to subhedral grains 0.2–0.4 mm across, but a few calcite phenocrysts are up to 5–7 mm in diameter. Calcite forms interlocking grains producing a mosaic texture with triple junctions as is typical of fine-grained carbonatite (Fig. 2e). This carbonatite could be termed alvikite based on the carbonatite classification of the IUGS Sub-commission (Le Maitre et al., 2002, p. 10). The alvikite contains minor minerals including euhedral to subhedral bastnaesite and parisite, 0.01–0.07 mm across, with some large grains up to 0.3–0.5 mm in diameter. These minerals form fine-grained aggregates that are interlocked with calcite, suggesting that they are primary minerals directly crystallizing from carbonatite magma (Fig. 2e, see Yang et al., 1998). This texture is similar to that

observed in the Mountain Pass carbonatite that was also interpreted as a magmatic crystallization texture (see Mariano, 1989; Wyllie et al., 1996). Optically, bastnaesite and parisite are light brown to light yellow, weakly pleochroic, high relief and high-grade interference color. Cebaite, huanghoite and cordylite are relatively rare in the alvikite as accessory minerals, and occur randomly as very fine aggregates (Yang et al., 2000a). Their single crystals are subhedral–anhedral, on the rims of which were often dissolved by late-stage solution, leaving irregular shapes. They are usually colorless or light brown to light yellow, weakly pleochroic, high relief and high-grade interference color. In addition, the dike contains riebeckite, magnesio-riebeckite, apatite, hematite, magnetite, monazite, allanite, dolomite, quartz, fluorite, and barite (Yang et al., 2000a, 2003).

The coarse-grained carbonatite dikes (D_c) are usually equigranular with euhedral to subhedral and often twinned calcite, 1–3 mm across, up to 5 mm in diameter (Fig. 2f). The calcite forms interlocking large plates producing a mosaic texture with triple junctions as is typical of sövitic carbonatites (Le Bas et al., 1992). Minor minerals are magnetite, apatite, pyrochlore, bastnaesite, monazite and barite. Field relations show that the coarse-grained carbonatite dike (D_c) formed earlier than the fine-grained carbonatite dikes (D_f). Le Bas et al. (1992) presented detailed descriptions of a coarse-grain calcite carbonatite dike (i.e. 1988 dike) cutting the Archaean Wu Tai Group migmatite. They show that this dike is similar in trace element (including REE) distribution patterns to those of the coarse-grained dolomite marble that host ores in the deposit, suggesting possible petrogenetic relations between them.

3. Analytical procedures

All EPMA analyses were performed at the Geology Department of Leicester University, UK, with a JEOL JXA-8600 “superprobe”, using wavelength dispersive spectrometers under experimental conditions of 15 kV and 3 nA, and electron beam size of 5 μm . Count times of 20 s were used for Ca, Mg, Fe, Mn, Sr, Ba, and 50 s for La and Ce. Standard samples used in analyses included natural minerals, synthetic glass and metal. Analytical precision for the elements

of interest is better than 1.0% relative. Detection limits for La_2O_3 and Ce_2O_3 are better than 0.10 wt.% (3σ), and for the other oxides better than 0.05 wt.% (3σ). The raw data were corrected with a routine ZAF program. Several grains of each sample were analyzed, and each of the grains was analyzed from rim to core (>3 spots). The average of all these analytical data represents the mineral chemistry of a sample. Some samples were re-analyzed at the Mineralogy Department of the Natural History Museum, London, with a Cameca SX50 electron microprobe analyzer, which, within analytical errors, produced the same results as those by the EPMA at Leicester University. Formula was calculated based on two CO_3^{2-} groups. Table 1 presents the summary of the analytical results. Detailed data are available upon request from the authors.

4. Results

Dolomite in the dolostone ($H8_s$) has higher CaO and MgO contents, and lower FeO, MnO and SrO contents than those in both of the coarse-grained ($H8_c$) and fine-grained dolomite marbles ($H8_f$) which host the ore bodies (Fig. 3, Table 1). The minor elements (i.e. FeO, MnO, SrO) in dolomite from the dolostone ($H8_s$) are significantly lower than 0.1 wt.%, in fact mostly cannot be detected by EPMA owing to very low concentration. They are similar to those in normal sedimentary dolostone (Table 1) from Lachaoshan, 50 km south of the Bayan Obo deposit, which is distal and not affected by any mineralization. The dolostone at Lachaoshan area belongs to the Bayan Obo Group in terms of stratigraphic correlation (Institute of Geochemistry, Chinese Academy of Sciences, 1988). Although BaO content is also low in carbonates from the sedimentary unit, there is no obvious difference compared to the mineralized dolomitic units ($H8_c$ and $H8_f$). All carbonate minerals from the samples analyzed in this study have very low La_2O_3 and Ce_2O_3 contents, and mostly below the EPMA detection limits (Table 1). Hence, La_2O_3 and Ce_2O_3 concentration data of the carbonates may have important implications for petrogenesis as will be discussed in Section 5.2. This information may provide indirect evidence that REE were formed as discrete phases in the ore deposit, such as bastnaesite

Table 1
EPMA results of representative carbonate minerals from Bayan Obo

Samples	90/65		LCS-05		88/150		90/62		B239		B239		B240	
Rock type	H8 _s		H8 _s		H8 _c		H8 _c		H8 _c		H8 _c		H8 _c	
Mineral	dolomite		dolomite		dolomite		dolomite		calcite		dolomite		dolomite	
Number of grains	7		2		5		8		3		2		2	
N	21		5		19		29		11		9		15	
	Average	STDEV	Average	STDEV	Average	STDEV	Average	STDEV	Average	STDEV	Average	STDEV	Average	STDEV
CaO	30.12	0.71	30.10	0.17	28.68	0.31	28.50	0.15	51.98	0.94	29.04	0.38	28.30	0.16
MgO	20.77	0.47	20.27	0.10	17.66	0.48	18.16	0.30	0.17	0.55	18.27	0.19	17.72	0.29
FeO	0.08	0.04	0.07	0.01	3.59	0.35	4.53	0.20	0.06	0.15	2.32	0.33	2.48	0.34
MnO	0.02	0.01	0.04	0.02	0.68	0.13	0.71	0.14	0.03	0.02	0.56	0.06	0.48	0.07
SrO	0.03	0.02	0.05	0.04	0.29	0.12	0.68	0.09	1.09	0.09	0.43	0.10	0.49	0.08
BaO	0.10	0.08	0.10	0.08	0.13	0.08	0.15	0.10	0.13	0.09	0.10	0.07	0.10	0.08
La ₂ O ₃	0.10	0.06	0.09	0.06	0.07	0.07	0.09	0.06	0.16	0.08	0.07	0.05	0.08	0.07
Ce ₂ O ₃	0.09	0.06	0.08	0.04	0.09	0.06	0.07	0.05	0.28	0.22	0.08	0.06	0.09	0.05
Total	51.31		50.79		51.19		52.88		53.91		50.88		49.74	
<i>Formulae based on two CO₃²⁻ groups</i>														
Ca ²⁺	1.017	0.007	1.029	0.005	1.009	0.011	0.977	0.006	1.956	0.031	1.018	0.012	1.02	0.00
Mg ²⁺	0.975	0.007	0.964	0.005	0.864	0.018	0.866	0.008	0.009	0.028	0.891	0.007	0.886	0.012
Fe ²⁺	0.002	0.001	0.002	0.000	0.099	0.010	0.121	0.005	0.002	0.004	0.063	0.009	0.070	0.010
Mn ²⁺	0.001	0.000	0.001	0.001	0.019	0.004	0.019	0.004	0.001	0.001	0.016	0.002	0.014	0.002
Sr ²⁺	0.000	0.000	0.001	0.001	0.006	0.002	0.013	0.002	0.022	0.002	0.008	0.002	0.010	0.001
Ba ²⁺	0.001	0.001	0.001	0.001	0.002	0.001	0.002	0.001	0.002	0.001	0.001	0.001	0.001	0.001
La ³⁺	0.001	0.001	0.001	0.001	0.001	0.001	0.001	0.001	0.002	0.001	0.001	0.001	0.001	0.001
Ce ³⁺	0.001	0.001	0.001	0.000	0.001	0.001	0.001	0.001	0.004	0.003	0.001	0.001	0.001	0.001
Mg/Fe	436.495	6.015	532.846	15.737	8.751	1.806	7.151	1.589	4.951	6.672	14.041	0.785	12.742	1.199
Ca/(Ca+Mg+Fe)	0.510	0.483	0.516	0.491	0.512	0.292	0.497	0.314	0.995	0.492	0.516	0.431	0.516	0.172
Mg/(Ca+Mg+Fe)	0.489	0.443	0.483	0.478	0.438	0.456	0.441	0.421	0.004	0.442	0.452	0.250	0.449	0.451
Fe/(Ca+Mg+Fe)	0.001	0.074	0.001	0.030	0.050	0.252	0.062	0.265	0.001	0.066	0.032	0.319	0.035	0.376

Samples	B241		88/147		88/147		B222		B222		B253		B256	
Rock type	H8 _c		H8 _f		H8 _f		H8 _f		H8 _f		H8 _f		H8 _f	
Mineral	dolomite		ankerite		dolomite		dolomite		calcite		dolomite		dolomite	
Number of grains	6		7		7		5		2		7		7	
N	26		32		29		22		8		36		24	
	Average	STDEV	Average	STDEV	Average	STDEV	Average	STDEV	Average	STDEV	Average	STDEV	Average	STDEV
CaO	28.86	0.51	27.95	0.49	26.14	0.78	29.99	0.79	50.76	1.76	29.22	0.36	28.53	0.35
MgO	18.34	0.30	14.60	0.55	15.41	0.91	15.81	0.41	2.73	0.54	18.16	0.56	17.86	0.57
FeO	3.28	0.59	7.23	0.61	5.41	1.99	4.00	0.56	1.23	0.18	2.78	1.12	3.83	1.05
MnO	0.67	0.04	1.45	0.20	1.70	0.32	1.45	0.43	1.10	0.09	0.73	0.05	2.02	0.55
SrO	0.49	0.15	0.14	0.04	0.35	0.08	0.22	0.04	0.50	0.08	0.53	0.06	0.33	0.14
BaO	0.10	0.07	0.14	0.09	0.07	0.07	0.09	0.07	0.32	0.19	0.11	0.07	0.11	0.08
La ₂ O ₃	0.10	0.07	0.08	0.06	0.08	0.06	0.09	0.06	0.20	0.11	0.10	0.06	0.10	0.06
Ce ₂ O ₃	0.06	0.05	0.06	0.05	0.06	0.04	0.14	0.08	0.62	0.28	0.08	0.05	0.15	0.09
Total	51.91		51.65		49.21		51.79		57.46		51.71		51.93	
<i>Formulae based on two CO₃²⁻ groups</i>														
Ca ²⁺	0.998	0.008	1.012	0.011	0.979	0.036	1.061	0.014	1.775	0.038	1.014	0.008	0.979	0.012
Mg ²⁺	0.882	0.013	0.735	0.023	0.802	0.042	0.778	0.010	0.133	0.028	0.876	0.026	0.852	0.026
Fe ²⁺	0.089	0.016	0.204	0.017	0.158	0.057	0.111	0.018	0.034	0.005	0.075	0.030	0.103	0.028
Mn ²⁺	0.018	0.001	0.041	0.006	0.050	0.009	0.041	0.012	0.030	0.002	0.020	0.001	0.055	0.015
Sr ²⁺	0.009	0.003	0.003	0.001	0.007	0.002	0.004	0.001	0.010	0.001	0.010	0.001	0.006	0.003
Ba ²⁺	0.001	0.001	0.002	0.001	0.001	0.001	0.001	0.001	0.004	0.002	0.001	0.001	0.001	0.001
La ³⁺	0.001	0.001	0.001	0.001	0.001	0.001	0.001	0.001	0.002	0.001	0.001	0.001	0.001	0.001
Ce ³⁺	0.001	0.001	0.001	0.001	0.001	0.001	0.002	0.001	0.007	0.004	0.001	0.001	0.002	0.001
Mg/Fe	9.962	0.837	3.599	1.378	5.076	0.738	7.034	0.592	3.962	5.325	3.763	23.449	8.309	0.910
Ca/(Ca+Mg+Fe)	0.507	0.224	0.519	0.217	0.505	0.268	0.544	0.327	0.914	0.532	0.902	0.454	0.506	0.181
Mg/(Ca+Mg+Fe)	0.448	0.354	0.377	0.454	0.414	0.311	0.399	0.250	0.068	0.394	0.077	0.524	0.441	0.390
Fe/(Ca+Mg+Fe)	0.045	0.422	0.105	0.329	0.082	0.421	0.057	0.423	0.017	0.074	0.021	0.022	0.053	0.429

(continued on next page)

Table 1 (continued)

Samples	90/64		90/58		90/63		CD7(2)		CD9(1)		CD14(1)		CD11(1)	
Rock type	H8 _f		H8 _f		H8 _f		D _c		D _c		D _c		D _c	
Mineral	dolomite		dolomite		dolomite		ankerite		ankerite		dolomite		calcite	
Number of grains	4		9		7		7		3		5		5	
<i>N</i>	13		29		30		41		25		20		27	
	Average	STDEV	Average	STDEV	Average	STDEV	Average	STDEV	Average	STDEV	Average	STDEV	Average	STDEV
CaO	28.17	0.67	28.88	0.28	28.98	0.19	29.08	0.17	29.15	0.19	30.12	0.81	54.66	1.53
MgO	16.83	0.55	16.70	0.67	17.58	0.35	14.06	0.46	13.22	0.29	17.62	0.68	0.65	0.08
FeO	5.43	0.36	5.02	0.76	4.90	0.39	7.27	0.78	8.46	0.55	2.78	1.49	1.46	0.45
MnO	1.01	0.10	1.01	0.07	1.16	0.09	3.13	0.38	3.63	0.20	1.54	0.10	1.45	0.20
SrO	0.17	0.06	0.21	0.05	0.23	0.05	0.42	0.10	0.47	0.13	0.14	0.04	0.62	0.24
BaO	0.15	0.10	0.11	0.10	0.11	0.07	0.11	0.08	0.11	0.08	0.11	0.08	0.11	0.08
La ₂ O ₃	0.07	0.06	0.09	0.07	0.10	0.07	0.10	0.07	0.10	0.07	0.08	0.07	0.09	0.07
Ce ₂ O ₃	0.07	0.05	0.07	0.04	0.08	0.05	0.08	0.05	0.08	0.05	0.07	0.05	0.09	0.05
Total	51.88		52.10		53.14		54.25		55.21		52.45		59.14	
<i>Formulae based on two CO₃²⁻ groups</i>														
Ca ²⁺	0.992	0.007	1.012	0.009	0.992	0.004	1.018	0.009	1.016	0.007	1.035	0.036	1.874	0.019
Mg ²⁺	0.824	0.010	0.814	0.024	0.837	0.012	0.685	0.020	0.641	0.014	0.842	0.032	0.031	0.004
Fe ²⁺	0.149	0.009	0.138	0.022	0.131	0.011	0.199	0.021	0.230	0.015	0.074	0.038	0.039	0.013
Mn ²⁺	0.028	0.003	0.028	0.002	0.032	0.002	0.087	0.010	0.100	0.005	0.042	0.003	0.039	0.005
Sr ²⁺	0.003	0.001	0.004	0.001	0.004	0.001	0.008	0.002	0.009	0.003	0.003	0.001	0.012	0.005
Ba ²⁺	0.002	0.001	0.001	0.001	0.001	0.001	0.001	0.001	0.001	0.001	0.001	0.001	0.001	0.001
La ³⁺	0.001	0.001	0.001	0.001	0.001	0.001	0.001	0.001	0.001	0.001	0.001	0.001	0.001	0.001
Ce ³⁺	0.001	0.001	0.001	0.001	0.001	0.001	0.001	0.001	0.001	0.001	0.001	0.001	0.001	0.001
Mg/Fe	5.527	1.151	5.920	1.111	6.390	1.133	3.448	0.926	2.786	0.909	11.340	0.837	0.790	0.296
Ca/(Ca+Mg+Fe)	0.505	0.259	0.515	0.160	0.506	0.157	0.535	0.185	0.538	0.203	0.530	0.339	0.964	0.539
Mg/(Ca+Mg+Fe)	0.419	0.397	0.415	0.442	0.427	0.448	0.360	0.392	0.340	0.380	0.432	0.301	0.016	0.105
Fe/(Ca+Mg+Fe)	0.076	0.345	0.070	0.398	0.067	0.395	0.104	0.423	0.122	0.418	0.038	0.360	0.020	0.355

Samples	88/141		88/141		88/142		88/142		93/149		90/42		CD15(2)	
Rock type	D _f		D _f		D _f		D _f		D _f		D _f		D _f	
Mineral	calcite		Mg-calcite		calcite		Mg-calcite		calcite		calcite		calcite	
Number of grains	7		1		9		5		5		7		7	
N	28		2		29		8		17		30		22	
	Average	STDEV	Average	STDEV	Average	STDEV	Average	STDEV	Average	STDEV	Average	STDEV	Average	STDEV
CaO	50.37	5.92	39.06	3.86	54.14	2.99	38.26	5.18	54.37	2.48	54.32	1.78	54.39	0.99
MgO	0.78	0.49	6.60	2.52	0.12	0.17	7.51	2.63	0.20	0.01	0.28	0.32	0.39	0.17
FeO	1.10	0.83	2.53	0.25	0.14	0.13	2.01	0.61	0.55	0.07	0.53	0.34	0.60	0.35
MnO	1.24	0.22	0.99	0.11	1.04	0.15	0.94	0.14	1.22	0.23	2.12	0.28	1.31	0.08
SrO	0.44	0.23	0.28	0.01	0.20	0.10	0.16	0.13	1.72	0.66	0.91	0.85	0.84	0.07
BaO	0.16	0.09	0.13	0.01	0.13	0.09	0.15	0.06	0.13	0.10	0.16	0.09	0.16	0.08
La ₂ O ₃	0.70	2.75	0.06	0.01	0.25	0.81	0.10	0.07	0.35	1.06	0.10	0.07	0.08	0.06
Ce ₂ O ₃	1.11	4.47	0.15	0.01	0.31	1.09	0.09	0.11	0.47	1.61	0.09	0.06	0.08	0.05
Total	55.89		49.78		56.33		49.21		59.02		58.52		57.84	
<i>Formulae based on two CO₃²⁻ groups</i>														
Ca ²⁺	1.841	0.165	1.522	0.131	1.943	0.047	1.490	0.165	1.892	0.053	1.891	0.035	1.908	0.013
Mg ²⁺	0.040	0.025	0.359	0.141	0.006	0.009	0.411	0.150	0.010	0.000	0.014	0.016	0.019	0.008
Fe ²⁺	0.031	0.024	0.077	0.007	0.004	0.004	0.061	0.020	0.015	0.002	0.015	0.009	0.016	0.009
Mn ²⁺	0.036	0.006	0.030	0.003	0.029	0.004	0.029	0.004	0.033	0.006	0.058	0.007	0.036	0.002
Sr ²⁺	0.009	0.005	0.006	0.000	0.004	0.002	0.003	0.003	0.032	0.012	0.017	0.016	0.016	0.001
Ba ²⁺	0.002	0.001	0.002	0.000	0.002	0.001	0.002	0.001	0.002	0.001	0.002	0.001	0.002	0.001
La ³⁺	0.011	0.044	0.001	0.000	0.003	0.011	0.001	0.001	0.004	0.014	0.001	0.001	0.001	0.001
Ce ³⁺	0.017	0.071	0.002	0.000	0.004	0.015	0.001	0.001	0.006	0.021	0.001	0.001	0.001	0.001
Mg/Fe	1.261	1.026	4.673	21.488	1.526	2.278	6.676	7.517	0.655	0.235	0.954	1.712	1.173	0.894
Ca/(Ca+Mg+Fe)	0.963	0.772	0.777	0.471	0.995	0.783	0.759	0.492	0.987	0.961	0.985	0.580	0.982	0.430
Mg/(Ca+Mg+Fe)	0.021	0.116	0.183	0.506	0.003	0.151	0.209	0.449	0.005	0.007	0.007	0.265	0.010	0.269
Fe/(Ca+Mg+Fe)	0.016	0.113	0.039	0.024	0.002	0.066	0.031	0.060	0.008	0.032	0.008	0.155	0.008	0.301

N, number of spot analyses; H8_c, coarse-grained dolomite marble; H8_f, fine-grained dolomite marble; H8_s, sedimentary dolostone; D_c, coarse-grained carbonatite; D_f, fine-grained carbonatite. Mg-calcite denotes magnesian calcite. See Fig. 1 for sample locations.

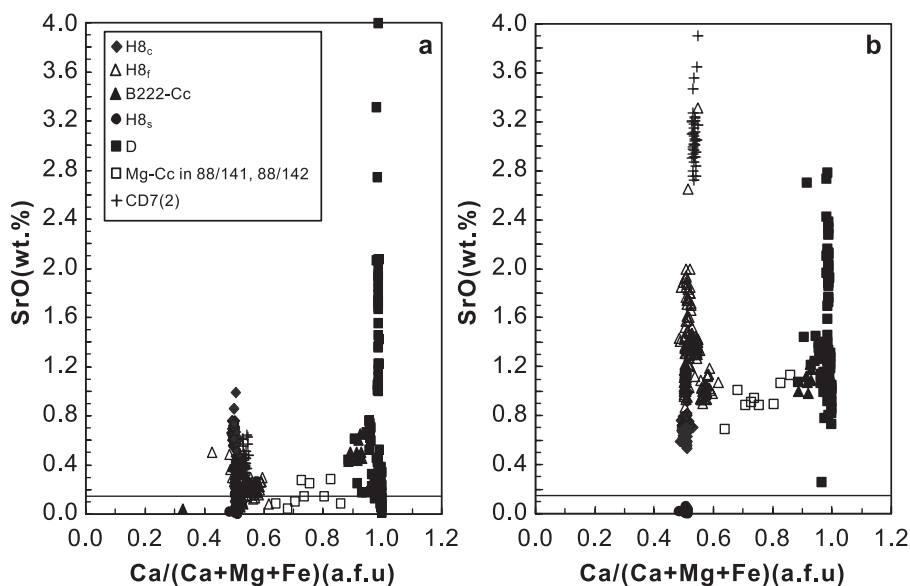


Fig. 3. Compositional variation of carbonate minerals from Bayan Obo: (a) SrO (wt.%) versus $[Ca/(Ca+Mg+Fe)]$ (a.f.u.), (b) MnO (wt.%) versus $[Ca/(Ca+Mg+Fe)]$ (a.f.u.). Mineral species are clearly defined by the ratio of $[Ca/(Ca+Mg+Fe)]$ (a.f.u.): calcites cluster around just below 1; dolomite and ankerite around 0.5, with the ratio of Mg/Fe < 4.0 for ankerite; $0.9 >$ magnesian calcite > 0.6 . The major differences for the carbonate minerals from various occurrences are their minor elements; see detailed discussion in the text and Fig. 4.

and monazite (Zhang et al., 1995; Yang et al., 1998, 2000a).

Comparison of dolomite in the ore-bearing coarse-grained dolomite marble (H8_c) and the fine-grained dolomite marble (H8_f) makes it clear that dolomite from the former contains higher SrO, and relatively lower MnO and FeO (Table 1). The SrO and MnO content in dolomite from the coarse-grained dolomitic marble (H8_c) are higher than 0.50 and 0.69 wt.%, respectively, while SrO and MnO content in dolomite from the fine-grained dolomitic marble (H8_f) are higher than 0.22 and 1.30 wt.%. All these dolomites may be termed Sr–Mn-bearing ferroan dolomite, which contains minor components (e.g. SrO and MnO) much higher than those in dolomite from the sedimentary unit (H8_s). The major components CaO and MgO in dolomite have a minimal range indicative of very small variation in $Ca/(Ca+Mg+Fe)$ ratios (atoms per formula unit, a.f.u.), whereas the minor elements such as SrO and MnO have a wide variation (Fig. 3). Therefore, these components in carbonate minerals can be used as discriminant parameters to determine the origin of the carbonate rocks. More interestingly, from coarse-grained marble (H8_c) to

fine-grained marble (H8_f), the SrO contents in dolomite always decreases, whereas MnO and FeO increase. These variations are discussed further in Section 5.2.

5. Discussion

5.1. Partitioning of SR and MN in carbonates

Partitioning of Sr and Mn between calcite and dolomite in an equilibrium system is controlled by crystal chemistry and physio-chemical conditions, where Sr prefers to enter calcite to replace Ca, whereas Mn and Fe tend to substitute Mg in dolomite (Kretz, 1982; Dickson, 1990). Using a partition coefficient K_D defined by Kretz (1982) can better describe these relations, which can also be used as criterion for recognizing equilibrium mineral assemblages (Table 2) (Kretz, 1982; Lentz, 1994).

In a typical sample (sample B222) of fine-grained dolomitic marble (H8_f), a few calcite grains were found to coexist with dolomite. The calcite contains higher SrO, but lower MnO and FeO than in the

Table 2
Summary of chemical characteristics of the carbonate minerals for different origins at Bayan Obo

	Primitive carbonatite ^a	Evolved carbonatite ^b	Hydrothermal carbonatite ^c	Sediments
Calcite	MnO < SrO	MnO > SrO	MnO > SrO	MnO < SrO
Dolomite	MnO < SrO	MnO > SrO	MnO > SrO	MnO > SrO
Ankerite	MnO < SrO	MnO > SrO	MnO > SrO	MnO > SrO
Calcite, dolomite, ankerite	MnO < 0.4 wt.% SrO > 0.4 wt.%	MnO > 0.4 wt.% SrO < 0.4 wt.%	MnO > 0.4 wt.% SrO < 0.4 wt.%	MnO < 0.15 wt.% SrO < 0.15 wt.%
Calcite–dolomite (equilibration)	$K_D(\text{MnO}) < 1$ $K_D(\text{SrO}) > 1$	$K_D(\text{MnO}) < 1$ $K_D(\text{SrO}) > 1$	$K_D(\text{MnO}) < 1$ $K_D(\text{SrO}) > 1$	$K_D(\text{MnO}) < 1$ $K_D(\text{SrO}) > 1$
Calcite–dolomite (disequilibrium)	$K_D(\text{MnO}) > 1$ $K_D(\text{SrO}) < 1$	$K_D(\text{MnO}) > 1$ $K_D(\text{SrO}) < 1$	$K_D(\text{MnO}) > 1$ $K_D(\text{SrO}) < 1$	$K_D(\text{MnO}) > 1$ $K_D(\text{SrO}) < 1$

$K_D(\text{MnO}) = \text{MnO (wt.\%)} / \text{in calcite/MnO (wt.\%)} / \text{in dolomite}$; $K_D(\text{SrO}) = \text{SrO (wt.\%)} / \text{in calcite/SrO (wt.\%)} / \text{in dolomite}$, as defined by Dawson et al. (1996); data referred from Veizer (1983), Hogarth et al. (1985), Le Bas and Srivastava (1989), Le Bas et al. (1992), Barker and Nixon (1989), Clarke et al. (1994), Ting et al. (1994), and Dawson et al. (1996).

^a Primitive carbonatite (C1, see Le Bas, 1977, 1981, 1999): the earliest carbonatite phase emplaced within a alkaline–carbonatite complex, that shows least differentiation and usually is cut by late evolved carbonatite phase such as C2. It contain higher Sr and P, lower REE, Ba, Mn, and relatively narrow $\delta^{18}\text{O}$ (SMOW) and $\delta^{13}\text{C}$ (PDB) isotope ratios similar to those of the Mantle.

^b Evolved carbonatite (C2, see Le Bas, 1977, 1981, 1999): temporally late carbonatite emplaced within a alkaline–carbonatite complex, that shows apparent differentiation cutting C1 and contains higher incompatible elements, e.g. REE, Ba, F, Mn, but lower Sr and P. It is usually associated with intensive REE, B, and F mineralization of economic significance. Its $\delta^{18}\text{O}$ isotope ratios are elevated up to +30‰, although $\delta^{13}\text{C}$ ratios remain a narrow range (0–9‰).

^c Hydrothermal carbonatite: carbonatites associated with alkaline metasomatites (fenites) but without associated silica undersaturated magmatic rocks (e.g. nephelinite, ijonite) occur along deep fault zones in the Precambrian continental crust, being the derivatives of the mantle-derived carbonatite–alkaline fluids (Samoilov, 1991) or devolatilization (Borodin and Kapustin, 1968; Hogarth et al., 1985).

associated dolomite (Table 1). A few calcite crystals occurring within a predominantly dolomite sample (B239) from coarse-grained dolomite marble (H8_c) have similar minor element partition relations (Table 1). These chemical characteristics are common for coexisting calcite and dolomite in carbonatites, where Sr partitions preferentially into calcite relative to coexisting dolomite, while Fe and Mn favor dolomite (Dawson et al., 1996). The element partition patterns between dolomite and calcite suggest chemical equilibrium may be achieved (Kretz, 1982) or preserved for these particular specimens.

According to the dolomite–calcite geothermometer of Anovitz and Essene (1987), equilibration temperature is estimated to be 530 °C (± 50) and 600 °C (± 50) for samples B239 and B222, respectively. These temperatures are similar to the temperatures of evolved carbonatite magmas (Wyllie et al., 1996), indicating that the igneous signatures of carbonate minerals may be preserved during the regional metamorphisms in the Bayan Obo area. The metamorphic grade of the Bayan Obo Group strata is most probably the lower part of greenschist facies in terms of mineral paragenesis (Chao et al.,

1992, 1997). Based on the studies of Chao et al. (1992, 1997), the Bayan Obo Group includes the dolomite marble unit (H8) although they did not divide the unit into the coarse-grained dolomitic marble (H8_c) and fine-grained dolomitic marble (H8_f) as is shown in this paper (Fig. 1). The temperatures estimated from the geothermometer (Anovitz and Essene, 1987), therefore, rule out the possibility of sedimentary origin for the dolomitic marble units. We interpret the dolomitic marble units (H8_c and H8_f) to be of carbonatitic origin, which is consistent with the conclusion of Le Bas et al. (1997) based on C, O and Sr isotope study.

It is necessary to notice that the dolomite–calcite geothermometer could be affected by the FeO content in calcite, which is not considered in the geothermometer of Anovitz and Essene (1987), and the effect of Mg diffusion (Gittins, 1979; Essene, 1983). Significant FeO content in calcite usually raises the estimated temperatures, but the effect of Mg diffusion results in low temperature estimation (Gittins, 1979). Both effects may cause the estimated temperatures for samples B239 and B222 to be lower than the “real” values.

5.2. Role of crystal fractionation in carbonatites on Sr, Mn and REE

Petrological observation and experimental studies show that carbonate minerals that precipitate from a carbonatite magma vary from calcite through dolomite to ankerite (Le Bas, 1989; Wyllie, 1989), resulting in Sr decrease, but such elements as Mn, REE, U, Th, F, and Ba increase in the residual melt (Le Bas, 1977, 1981, 1989; Wyllie et al., 1996). Hence, the late-stage evolved carbonatites are associated with REE (e.g. bastnaesite, monazite), fluorite and barite mineralization, when these minerals are saturated and crystallize. Similarly, the fine-grained dolomite marble (H8_f) may originally be a product of late-stage carbonatite prior to metamorphism, based on its geochemical similarity to the carbonatite dikes (Le Bas et al., 1996, 1997; Yang et al., 2003).

The SrO and MnO variations in carbonate minerals from the H8_c and H8_f (Table 1, Fig. 4a and b) are consistent with those from coarse-grained sövite to fine-grained alvikite controlled by crystal fractionation of carbonatite magmas (Le Bas, 1981; 1989; Clarke et al., 1994). Whole-rock XRF analyses indicate that bulk REE concentrations in the coarse-grained marble (H8_c) are much lower than those in the fine-grained marble (H8_f) (Le Bas et al., 1996, 1997; Bai et al., 1996; Yang unpublished data). This may reflect fractional crystallization of carbonate minerals resulting in REE enrichment in the residual carbonatitic magma. This consequence can be ascribed to the very low REE concentrations in these carbonates, which is consistent with REE partition coefficients between carbonates and carbonatitic melts that are much less than 1. Dawson et al. (1996) used a synchrotron XRF probe to analyze carbonates, e.g. calcite and dolomite, from seven African carbonatites, illustrating that the carbonates always have very low REE content (Ce ≤ 250 ppm). With the help of ICP-AES, Hornig-Kjarsgaard (1998) analyzed calcite and dolomite separates (24 samples) from eight typical carbonatite occurrences worldwide, and achieved a similar conclusion. These data together with our data presented here suggest that REE partition into the residual carbonatitic magmas as carbonates undergoes fractional crystallization.

5.3. Role of hydrothermal fluids

Hydrothermal fluids interacting with carbonates must change their compositions. Magmatic calcite in carbonatites is typically rich in Sr (Le Bas, 1989), and secondary or late-stage calcite invariably shows some depletion in Sr that is in some cases accompanied by enrichment in Mn (Clarke et al., 1994; Zaitsev and Chakhmouradian, 2002) depending on the nature of fluids involved. If calcite is in equilibrium with meteoric, or sea water, then its Sr and Mn contents will be reduced to the 0.1- to 1000-ppm level (Dickson, 1990). These values are much lower than those from carbonatites (Dawson et al., 1996). Le Bas et al. (1997) considered the possibility that the dolomite marbles (particularly H8_f) were originally sedimentary and later pervaded by carbonatitic fluids prior to the mineralization. However, the O, C, and Sr isotopic evidence together with fenitization in the hanging wall and foot wall of the marbles at Bayan Obo rule out the formation of carbonatite from a carbonate sediment in this manner (Le Bas et al., 1996, 1997).

The role of hydrothermal fluids reacted with carbonates has been noticed in the present study. For example, a fine-grained calcite carbonatite dike cuts the dark slate (H9) that forms the roof of the main mineralized (H8_c and H8_f) marbles, from which two samples (88/141 and 88/142) were collected (Fig. 1; Table 1). Calcite from these two samples contains average SrO of 0.20–0.43 wt.% and MnO of 1.05–1.28 wt.% (Table 1), similar to Sr–Mn-bearing calcite from common carbonatites elsewhere. It is interesting that several magnesian calcite grains (average mole Mg/Ca ratio of 0.35) were found from sample 88/142 (taken from margin of the dike); their SrO and MnO contents are obviously lower than those in the other calcites in the same sample, ruling out equilibration between the mineral pairs. These data are interpreted to indicate that the carbonatite dike may be affected by late Mg-bearing fluids that reacted with the calcite. As a result, Ca in the calcite would be substituted by Mg from the fluids, which resulted in formation of the magnesian calcite and partial removal of Sr and Mn. $\delta^{18}\text{O}(\text{SMOW})$ ratio of this sample is unusually low (2.6‰) although the $\delta^{13}\text{C}(\text{PDB})$ ratio retains a typical carbonatite value (–4.33‰), suggesting the convection, along the contact, of meteoric water in the adjacent slate driven by heat supplied from the dike

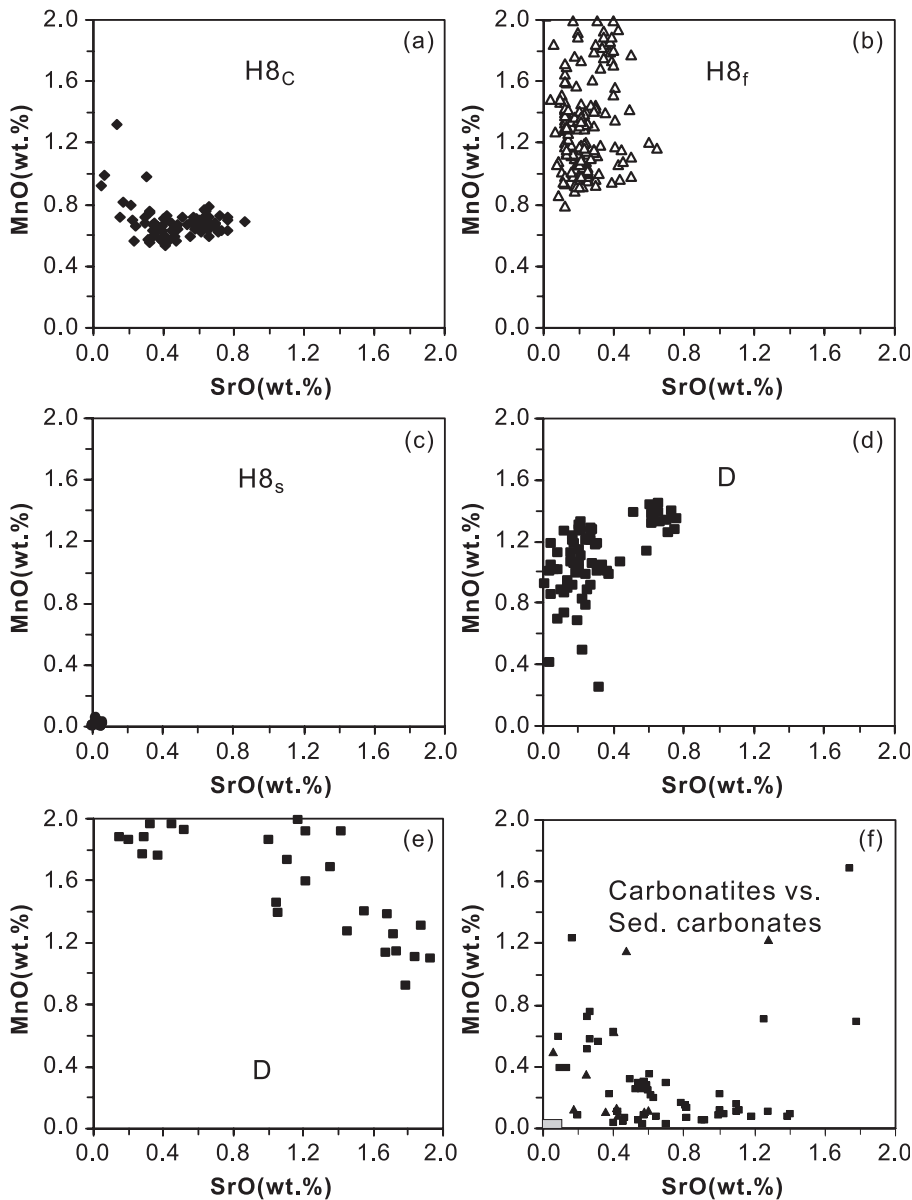


Fig. 4. Plot of SrO (wt.%) versus MnO (wt.%) in carbonate minerals from Bayan Obo and comparison with other occurrences elsewhere: (a) solid diamond—coarse-grained dolomite marble ($H8_c$); (b) open triangle—fine-grained dolomite marble ($H8_f$); (c) solid circle is sedimentary dolostone ($H8_s$); (d) and (e) solid square is carbonatite dikes (D); (f) a compilation of carbonate data for typical sedimentary carbonate rocks and carbonatites elsewhere in the world: sedimentary dolomite and calcite located in a small box close to the origin (the highest SrO in dolomite and calcite are 0.07 and 0.11 wt.%, respectively; and maximum MnO in dolomite and calcite are 0.05 wt.%; data taken from Veizer et al., 1978; Scoffin, 1987; Veizer, 1983; Dickson, 1990; Tucker and Wright, 1990); carbonatites: dolomite (solid triangle) and calcite (solid square). Carbonatite data sources: Gatineau, Quebec (Hogarth et al., 1985); Sukulu, Uganda (Ting et al., 1994; Dawson et al., 1996); Phalaborwa and Goudini, South Africa; Tundulu Hill, Malawi; Panda Hill and Kerimasi, Tanzania; Tororo, Uganda (Dawson et al., 1996); Oka, Quebec; Kaiserstuhl, Germany; Fen, Norway; Alnö, Sweden; Siilinjärvi and Sokli, Finland; Jacupiranga, Brazil; and Loolekop, Phalaborwa, South Africa (Hornig-Kjarsgaard, 1998); Kola Peninsula, Russia (Zaitsev and Chakhmouradian, 2002), showing the distinction between sedimentary carbonate rocks and carbonatites.

or from the Permian granite not far below it (Le Bas et al., 1997). Additionally, slightly higher $^{87}\text{Sr}/^{86}\text{Sr}$ ratio of 0.70686 for this sample (Le Bas et al., 1997) supports that it interacted with fluids carrying the signature of the H9 slate.

5.4. Comparison with other carbonatites and sedimentary carbonate rocks

The plot of detailed analytical data of the carbonate minerals on SrO versus MnO diagrams (Fig. 4) demonstrates their distribution fields, which discriminate different occurrences of the carbonate rocks at Bayan Obo: the dolomites from the sedimentary dolostone (H8_s) north of the Kuangou fault zone fall into the field S near the origin of the graph (Fig. 4c), which completely differ from those from the ore-bearing dolomite marbles (H8_c and H8_f) and carbonatite dikes (D) (Fig. 4a, b, d and e). However, dolomites from the coarse-grained dolomite marble (H8_c) and the fine-grained dolomite marble (H8_f) are similar to those from the carbonatite dikes, which are located in the fields of carbonatites produced by fractional crystallization of carbonatitic magmas (Fig. 5) except for samples from a fine-grained extremely rich REE

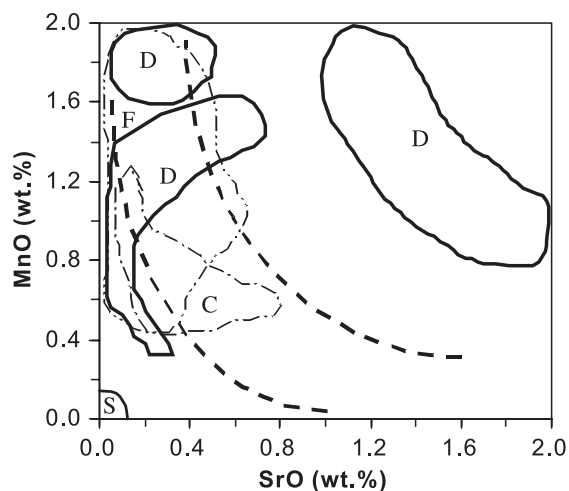


Fig. 5. SrO versus MnO discrimination diagram for carbonatites and sedimentary carbonate rocks based on the combination of Fig. 4a to e: Field S represents the distribution field for H8_s, C for H8_c, F for H8_f, and D for carbonatite dikes. The dashed lines show the trend of fractional crystallization of carbonatite magma, based on the study of Clarke et al. (1994).

carbonatite dike (Yang et al., 2000a, 2003). Calcites from this carbonatite dike (samples 93/149 and 90/42) are very rich in SrO (0.91–1.72 wt.%) and MnO (1.23–2.12 wt.%) (Table 1), and fall into the upper part on the SrO versus MnO variation diagram (Fig. 4e), but their variation trend is still parallel to the fractional crystallization trend (Fig. 5) of carbonatites defined by Clarke et al. (1994). This relation suggests that the evolution of the carbonatite may be controlled by crystal fractionation, resulting in further REE enrichment in the residual carbonatite magmas as described by Wyllie et al. (1996). It is not clear why the calcites from this dike contain significantly higher SrO and MnO than those from the other dikes, even though the dike may be affected by late-stage hydrothermal fluids (Yang et al., 2000a). The Sr–Mn-bearing calcite is typical of carbonatites elsewhere (Le Bas et al., 1992). Calcite from carbonatite lava at Fort Portal, Uganda, is recorded to contain very high SrO (1.36–2.71 wt.%), but relatively low MnO (0.03–0.04 wt.%) (see Barker and Nixon, 1989). These authors interpreted these low MnO contents in carbonatites as a result of substantial exchange with meteoric water at low temperature during post-emplacement alteration, based on oxygen and carbon isotope data. Experimental study shows that Mn in calcite is readily removed by low temperature fluids if the solution is depleted in Mn (Reeder et al., 1990). According to the mole percentage of MgCO₃ dissolved in calcite equilibrated with dolomite (Anovitz and Essene, 1987), samples 93/149 and 90/42 yield temperatures of 550° (± 50) and 620° (± 50) °C, respectively. These temperatures are within the range of evolved carbonatite melts (Wyllie et al., 1996), but much higher than those of carbonatites (100°–300 °C) of hydrothermal origin derived from the mantle devolatilization (Borodin and Kapustin, 1968; Hogarth et al., 1985).

Stable isotope data (Liu, 1986; Le Bas et al., 1997) show that the sedimentary dolostone (H8_s) has values of $\delta^{18}\text{O}$ (SMOW) ca. +20‰ and $\delta^{13}\text{C}$ (PDB) ca. 0‰. In contrast, the coarse-grained dolomite marble (H8_c) has $\delta^{18}\text{O}$ ratios between +8‰ and +12‰, and $\delta^{13}\text{C}$ ratios between –5‰ and –3‰, whereas the fine-grained dolomite marble (H8_f) has $\delta^{18}\text{O}$ ratios between +12‰ and +16‰, and $\delta^{13}\text{C}$ ratios between –4‰ and 0‰. These variations in O and C isotopes of the H8_c and H8_f are similar to those of carbonatite

fractionation from sövite to alvikite elsewhere (Le Bas, 1981; Clarke et al., 1994), where they are interpreted as the carbonatite fractionation sequence at mole ratios $\text{H}_2\text{O}/\text{CO}_2 < 1$ (Knudsen and Buchardt, 1991). Clearly, this distribution is the opposite of the direction of change of $\delta^{18}\text{O}$ and $\delta^{13}\text{C}$ during post-depositional alteration for dolomite in limestones (Hall and Veizer, 1996). $^{87}\text{Sr}/^{86}\text{Sr}$ data confirm this distinction: >0.710 for the $\text{H}8_s$ and <0.704 for the $\text{H}8_c$ and $\text{H}8_f$ (Le Bas et al., 1996, 1997).

A compilation of carbonate data for typical sedimentary carbonate rocks and carbonatites elsewhere in the world clearly indicates that sedimentary dolomite and calcite are located in a small box close to the origin (Fig. 4f) (the highest SrO in dolomite and calcite are 0.07 and 0.11 wt.%, respectively; maximum MnO in dolomite and calcite are 0.05 wt.%; data taken from Veizer et al., 1978; Scoffin, 1987; Veizer, 1983; Dickson, 1990; Tucker and Wright, 1990), whereas dolomite and calcite from carbonatites show typical high SrO and MnO (Fig. 4f). The carbonatite data cited here include Gatineau, Quebec (Hogarth et al., 1985), Sukulu, Uganda (Ting et al., 1994; Dawson et al., 1996), Phalaborwa and Goudini in South Africa, Tundulu Hill in Malawi, Panda Hill and Kerimasi in Tanzania, and Tororo in Uganda (Dawson et al., 1996), Oka in Quebec, Kaiserstuhl in Germany, Fen in Norway, Alnö in Sweden, Siilinjärvi and Sokli in Finland, Jacupiranga in Brazil, and Loolekop, Phalaborwa in South Africa (Hornig-Kjarsgaard, 1998), and Kola Peninsula in Russia (Zaitsev and Chakhmouradian, 2002). The distinction between sedimentary carbonate rocks and carbonatites is evident, and is comparable to those carbonate rocks from Bayan Obo presented in this study.

Calcite, ankerite and dolomite in carbonatites of hydrothermal origin from the mantle devolatilization usually contain higher MnO than SrO, indicating that Mn activity is relatively higher in the fluids (Hogarth et al., 1985). Evolved carbonatites appear to have the same features. However, carbonates in more primitive carbonatites seem to have higher SrO than MnO (Clarke et al., 1994; Ting et al., 1994; Dawson et al., 1996). The distinction between hydrothermal and evolved carbonatites may not be possible, but both of them are obviously different from primitive carbonatites, which can be determined through study of Sr and

Mn partitioning between coexisting calcite and dolomite (and/or ankerite) as well as their relative concentrations in the minerals (Table 2). An interesting study of spheroidal calcite inclusions in apatite shows that the early magmatic calcite contains higher SrO (0.54 ± 0.08 wt.%, $n=5$) than MnO (0.25 ± 0.12 wt.%) (Ting et al., 1994), suggesting that calcite crystallizing from more primitive carbonatite may have higher SrO relative to MnO. These values do not change significantly when the calcite has interacted with late-stage fluids through microfractures as is shown in pitted calcite inclusions (SrO = 0.46 ± 0.05 wt.%, MnO = 0.25 ± 0.02 wt.%, $n=5$) associated with microfractures traversing the apatite (Ting et al., 1994).

Sedimentary calcite and dolomite commonly contain very low SrO (<0.01 wt.%) and MnO (<0.01 wt.%) contents (Veizer et al., 1978; Veizer, 1983; Dickson, 1990; Tucker and Wright, 1990), which are normally not detected by EPMA. They apparently differ from those in carbonatites (Figs. 4e and 5). Metamorphic marbles of sedimentary origin usually preserve their trace element geochemistry (Hall and Veizer, 1996). When the marbles of sedimentary origin are interbedded with slate, quartzite, schist or paragneiss, there are no fenites developed at the borders between the carbonate units and the others (Yang et al., 2000b). However, carbonatite dikes and intrusions always have fenite halos that are usually well preserved even in relatively high-grade metamorphism (Barker, 1989; Yang et al., 2000b). More recently, Boundy et al. (2002) recognized that calcite in eclogite facies marbles from the Caledonian Orogen, western Norway contains variable SrO (0–1.21 wt.%) and MnO (0–0.51 wt.%). Their studies suggest that the marbles themselves are a product of metasomatic reactions (i.e. breakdown of scapolite in granulite into carbonates and sulfates) and precipitated from a fluid along shear zones during eclogite facies metamorphism. Although the calcite from these eclogite facies marbles is indistinguishable in chemical composition (e.g. SrO and MnO) with that from carbonatites, there are no fenites occurring in the country rocks immediately around those marbles. Furthermore, interaction of sedimentary carbonates with various fluids could not raise their Sr and Mn contents (Bouzenoune and Lécalle, 1997), although they could partially be

removed. These empirical investigations provide substantial support that the ore-bearing dolomitic marbles at Bayan Obo could not be significantly modified during deformation and low-grade metamorphism (lower greenschist facies). Therefore, detailed EPMA analyses of carbonate minerals of different occurrences from the Bayan Obo area support a carbonatitic origin for the ore-bearing dolomite marbles.

6. Conclusions

In conclusion, SrO and MnO contents in carbonate minerals can be employed as important parameters to distinguish sedimentary carbonate rocks from carbonatites as shown at Bayan Obo. The main rock-forming minerals, i.e. dolomite or ankerite in ore-bearing dolomite marbles of the Bayan Obo giant REE–Nb–Fe ore deposit are similar to those from carbonatite dikes near the deposit, but different from those in the normal sedimentary dolostone, implying that these mineralized dolomite marbles may be carbonatitic in origin. Variation of MnO and SrO content in dolomite from the ore-bearing dolomite marbles ($H8_c$ versus $H8_f$) may be interpreted by fractional crystallization of a carbonatitic magma, which results in REE enrichment in the evolved residual magma. Coexisting calcite and dolomite from the dolomite marbles yield temperatures of 530 to 600 °C, similar to the temperatures (550 to 620 °C) for carbonatite dikes. These temperatures, thought to be underestimated, are within the ranges of temperatures of evolved carbonatites. Although the sedimentary carbonates from different geological ages and sedimentary environments may contain variable SrO and MnO contents, there are no reports that their SrO and MnO concentrations are higher than 0.15 wt.%, but rather are normally less than 0.01 wt.%. However, carbonates from carbonatites typically contain both SrO and MnO higher than 0.15 wt.%. Therefore, this paper assigns the value of MnO and SrO contents in carbonate minerals higher than 0.15 wt.% as an indicator of carbonatites. These criteria together with fenitization may be used as key signatures to distinguish magmatic carbonatites from sedimentary carbonate rocks, even when both were metamorphosed to marbles.

Acknowledgements

We are grateful to Mr. Rob Wilson giving valuable help and advice on EPMA analyses at the Geology Department of Leicester University. Drs. T.C. Williams, L.S. Campbell and P. Henderson at the Natural History Museum, London kindly provided help on CAMECA SX50 probe analyses. Prof. Daniel S. Barker (the University of Texas at Austin), Prof. Eric J. Essene (the University of Michigan), and Prof. David R. Lentz (the University of New Brunswick) are greatly appreciated for their critical reviews of an earlier version of this manuscript. Constructive discussions with Profs. David R. Lentz, Bruce E. Broster, Ke-Jie Tao and Pei-Shan Zhang are very helpful to us for understanding the chemical distinctions between carbonatites and sedimentary carbonate rocks. We are indebted to the journal reviewers Prof. K. Bell and Dr. Martin Smith for their comments and helpful suggestions that improve the manuscript. The revised manuscript is benefited by useful suggestions and editorial handling from Prof. S. Foley. The Royal Society, London (UK) and the British Council are acknowledged for providing fellowships and funding to support us while working at Leicester. The project is also supported by Natural Science Foundation of China, through a grant No. 49872032 to XMY.

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