

## Geochemistry of carbonatites in Maoniuping REE deposit, Sichuan Province, China

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**Abstract** Carbonatites in the Maoniuping REE deposit, Sichuan Province, which are spatially and temporally associated with rare earth mineralization, were emplaced at the time of Himalayan. The rocks are carbonatite-syenite complexes, with the mineral assemblages of calcite-aegirine-acmite-arfvedsonite-mica-orthoclase. The rocks are characterized by the enrichment in incompatible elements, such as Sr, Ba and REE, with C and O isotopic compositions of the "primary igneous carbonatites", relatively high initial  $^{87}\text{Sr}/^{86}\text{Sr}$  ratios and low  $\epsilon_{\text{Nd}}$  values. All of these suggest that the rocks were derived from the metasomatic enriched mantle. It is demonstrated by geological and geochemical evidence that the mixing of the Himalayan subducting crustal materials with mantle source EM1 is probably the main factor responsible for the formation of carbonatites. The carbonatite-syenite complexes were generated from liquid immiscibility of  $\text{CO}_2$ -rich alkalic silicate magma, which was derived from partial melting of the metasomatic mantle.

**Keywords:** carbonatite, geochemistry, EM1-EM2, Maoniuping REE deposit.

Carbonatites are rarely igneous rocks distributed on the earth. The rocks usually form ring complexes with alkalic rocks, occurring in the environments of continental rift, collisional orogenic zone and oceanic island<sup>[1, 2]</sup>. Numerous facts and experiments have revealed<sup>[3-5]</sup> that the parental magma of carbonatites originated from the metasomatic enriched mantle; compared with silicate melts, carbonatite melts are characterized by the relative enrichment in volatile components, low solidus and liquidus temperatures, viscosities, densities, and high mobility of carbonatite magma relatively little contaminated by crustal materials during magma emplacement or eruption. The rocks are closely associated with REE, Nb, P, Fe and Th. Thus, comprehensive investigations of carbonatites are very important for a better understanding of mantle metasomatism, the origin and evolution of magma and related mineralization.

Although carbonatites have been found in many parts of China, such as Miaoya of Hubei Province, Zijinshan of Shanxi Province, Dashigou of Shaanxi Province, Fanshan of Hebei Province, central Shandong Province, Bayan Obo of Inner Mongolia and Maoniuping of Sichuan Province<sup>[6]</sup>, relatively little work has been published on the geochemistry and petrogenesis of the rocks. Carbonatites are closely associated with rare earth mineralization in Maoniuping, Sichuan

Province. Some petrological and geochemical features of the rocks were reported by Pu<sup>[7]</sup> and Wang et al<sup>[8]</sup>. In this paper, we systematically analyze the compositions of major elements, trace elements and isotopes, and discuss the characteristics of carbonatite source and genetic relationship between carbonatites and syenites.

## 1 Geological settings and fundamental features of carbonatites

The Maoniuping REE deposit is located in the northern Panxi rift. The area is divided into the east rift zone, the middle transitional zone and the west geosyncline fold zone by the NE-trending Nanhe fault and the Jingpingshan fault, and the Maoniuping REE deposit is located in the middle transitional zone. In the area are primarily distributed the Middle Devonian silt-clasolite, carbonate and the Quaternary proluvium and talus. The main structure is the NE-trending fault. Magmatites are distributed extensively in this area, including Yanshanian granites (Mianxi granites), Himalayan syenites and minor basalts and rhyolites of unknown ages<sup>[9]</sup>.

Vein-like carbonatites intruded into the center of syenites, with the attitude generally in consistence with the NE-trending fault. Veinlets on both sides of the main vein also intruded into syenites (fig. 1). The carbonatite veins (90—200 m wide) extend 400 m without pinching. Carbonatites are mostly composed of calcite (>90%), with minor amounts of biotite, aegirine, acmite, arfvedsonite, orthoclase, etc. (<10%), and the accessory minerals such as apatite, arpidelite etc. (<1%). On the margins of carbonatite veins the fenitization and arfvedsonitization are developed. According to Pu<sup>[10]</sup>, the K-Ar age of carbonatites is about (31.7±0.7) Ma, which is generally equivalent to that of syenites ((27.8±0.5)—(40.3±0.7) Ma).

## 2 Samples and analytical methods

Fresh calcites were selected from carbonatites and analyzed for major elements, trace elements and isotopes. The major elements of syenites and calcites were determined by using the wet-chemical method at the Institute of Geochemistry, the Chinese Academy of Sciences. The trace elements were analyzed by ICP-MS with the precision being better than 10%, also at the Institute of Geochemistry, the Chinese Academy of Sciences. The published paper<sup>[11]</sup> can be referred to for the details of the analytical procedure. In the processes of analysis the international standard AMH-1 (andesite) was used as the standard specimen for quality control. The C and O isotopic compositions were analyzed at the Institute of Geology, the Chinese Academy of Geological Sciences (CAGS) by the 100% phosphorate method on the MAT 251 EM mass spectrometer, with the analytical precision being ±0.2‰. The Sr and Nd isotopic compositions were analyzed at the Institute of Geology and Geophysics, the Chinese Academy of Sciences. The analytical procedures for Sr and Nd isotopes were similar to each other. Samples were first dissolved in HF+HClO<sub>4</sub>, then Rb, Sr and REE were separated on the AGW50×12 (100—200 meshes) cation exchange columns and were dried, respectively. The REE were further dissolved in 200 μL of 0.1 mol/L HCl solution, then Sm and Nd were separated on the levextrel resin ion exchange columns.

The blanks were  $Rb=50 \times 10^{-11}$  g/g,  $Sr=50 \times 10^{-11}$  g/g,  $Sm=7 \times 10^{-11}$  g/g and  $Nd=8 \times 10^{-11}$  g/g. NBS987 and LA standards analyzed during this work gave  $^{87}Sr/^{86}Sr=0.710234 \pm 7$  and  $^{143}Nd/^{144}Nd=0.511838 \pm 8$ .

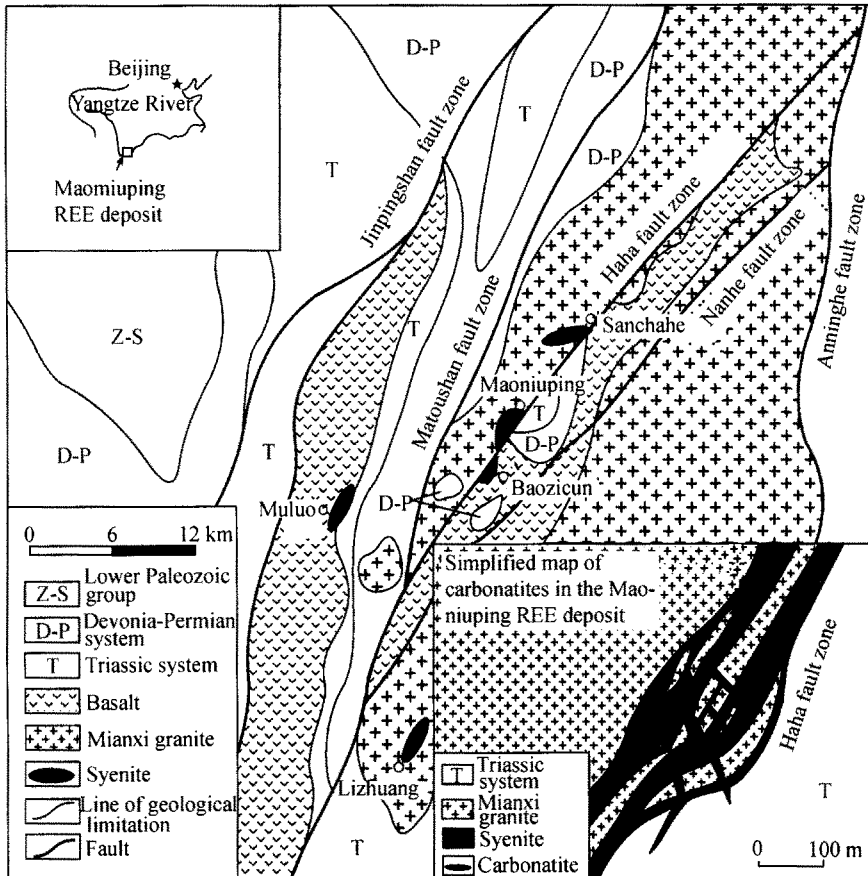


Fig. 1. Simplified geological map of the Maoniuping REE deposit. The map is revised according to data from No. 109 Geological Party.

### 3 Geochemistry

#### 3.1 Major elements

Listed in table 1 are the major element contents of carbonatites in the Maoniuping mineral district. The concentrations of  $SiO_2$  range from 1.67% (weight percent) to 3.65% (weight percent, the same hereafter), and the contents of  $CaO$ ,  $MgO$  and  $Fe_2O_3+FeO$  range from 54.20% to 55.50%, 0.01% to 0.20%, and 0.20% to 0.36%, respectively. The contents of  $Na_2O$  and  $K_2O$  are extremely low (0.05%—0.08%, 0.001%—0.12%, respectively). The resulting data are fallen into the range of the major element contents of carbonatites reported by Samoiliv et al.<sup>[12]</sup>. The  $CaO/(CaO + MgO + FeO + Fe_2O_3 + MnO)$  ratios of 95.8%—98.5% are consistent with those of the calico-carbonatites.

### 3.2 Trace elements

In comparison with the average values of the world calcio-carbonatites<sup>[13]</sup>, those of the rocks in the Maoniuping area are characterized by relatively low Nb and Ta contents and relatively high Sr/Sm, Zr/Hf, Sm/Hf, Nb/Ta and La/Nb ratios (128—183, 1.04—2.44, 703—1401, 1.20—18.4 and 7526—28380, respectively). It is shown in fig. 2 that carbonatites in the Maoniuping area relatively enrich Ba, Sr and LREE, and deplete Nb, Ta, Zr, Hf and Ti.

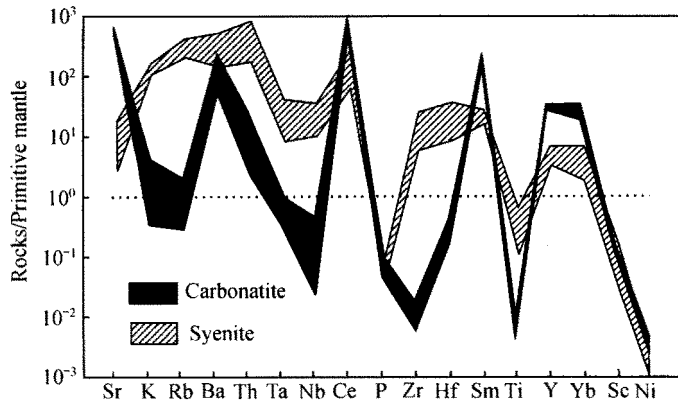


Fig. 2. Primitive mantle-normalized trace elements of carbonatites and syenites in the Maoniuping orefield. The values of primitive mantle are cited from ref. [14].

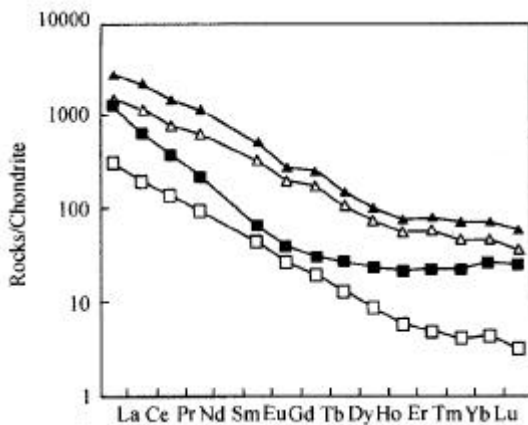


Fig. 3. Chondrite-normalized REE distribution patterns of carbonatites and syenites in the Maoniuping orefield. The data of chondrites are cited from ref. [15]. The solid area stands for the highest contents of REE, and the hollow area indicates the lowest contents of REE.  $\square$ , calcites;  $\triangle$ , syenites.

The  $\Sigma$ REE contents of carbonatites range from  $2051 \times 10^{-6}$  to  $3652 \times 10^{-6}$ , with LREE and HREE concentrations ranging from  $1922 \times 10^{-6}$  to  $3508 \times 10^{-6}$  and  $106 \times 10^{-6}$  to  $144 \times 10^{-6}$ , respectively. The LREE/HREE ratios range from 14 to 25. The chondrite-normalized REE distribution patterns are of the LREE enrichment type (fig. 3), with  $(\text{La}/\text{Yb})_n = 23.1\text{—}44.4$ ,  $(\text{La}/\text{Nd})_n = 2.09\text{—}2.68$ ,  $(\text{Gd}/\text{Yb})_n = 2.53\text{—}4.18$ , and show weak negative Eu anomalies ( $d\text{Eu} = 0.79\text{—}0.82$ ), and unremarkable Ce anomalies ( $d\text{Ce} = 0.88\text{—}1.06$ ). It is also shown in fig. 3 that the chondrite-normalized REE distribution patterns of carbonatites are similar to those of syenites with  $(\text{La}/\text{Yb})_n = 72\text{—}213$ ,  $(\text{La}/\text{Nd})_n =$

$3.80\text{—}4.80$ ,  $(\text{Gd}/\text{Yb})_n = 3.45\text{—}7.93$ ,  $d\text{Eu} = 0.86\text{—}1.01$ , and  $d\text{Ce} = 0.77\text{—}1.02$ , except that the former rocks enrich HREE, with low  $(\text{Gd}/\text{Yb})_n$  ratios.

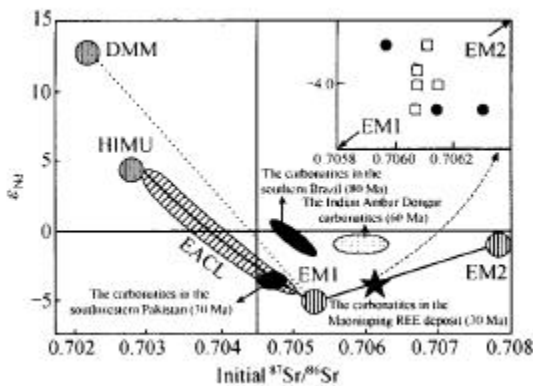


Fig. 4.  $\epsilon_{\text{Nd}}$  vs. initial  $^{87}\text{Sr}/^{86}\text{Sr}$  for carbonatites in the orefield. To limit the potential distortions caused by epsilon values, the comparison of Sr and Nd isotope is restricted to the foreign carbonatites younger than 100 Ma. The Brazilian data are cited from ref. [20], the Pakistani data from ref. [1], and the Indian data from ref. [21]. Squares stand for carbonatites, circles for syenites in the Maoniuping REE deposit. The range of EACL are cited from ref. [22]. DMM, HIMU, EM1, EM2 are the mantle end-member components from ref. [23],  $(^{87}\text{Sr}/^{86}\text{Sr})_{\text{UR}} = 0.7045$  [18].

from the Maoniuping area are fallen into the area between EM1 and EM2 in fig. 4, deviating from the EACL. They are characterized by high initial  $^{87}\text{Sr}/^{86}\text{Sr}$  ratios, and low  $\epsilon_{\text{Nd}}$  ratio values.

## 4 Discussion

### 4.1 Genesis of carbonatites and relationship between carbonatites and syenites

In the Maoniuping REE deposit, carbonatites are composed of calcite-aegirine-acmite-arfvedsonite-mica-orthoclase. The rocks are characterized by enrichment in incompatible elements, such as Sr, Ba, REE, with C and O isotopic compositions of the “primary igneous carbonatites”, Sr and Nd isotopic compositions fall into the area between EM1 and EM2. All of these suggest that carbonatites are probably the products of mantle carbonate magma. Three genetic models for carbonatites are described as follows: ( ) carbonatites are derived from low-degree partial melting (<1%) of  $\text{CO}_2$ -rich asthenosphere or lithosphere mantle [22]; ( ) the rocks are the products of fractional crystallization of  $\text{CO}_2$ -rich alkali silicate melt [24]; ( ) they are the products of liquid immiscibility of  $\text{CO}_2$ -rich alkalic silicate melt [25, 26]. The evidence obtained from the present study supports the third viewpoint. Further discussions are given below:

(1) Consistent with the occurrences of other carbonatite-syenite complexes both at home and abroad [27], carbonatites in the Maoniuping area are closely associated in space with syenites, forming ring complexes with carbonatites at the center (fig. 1). Both rocks were emplaced approximately at the same time ((31.7±0.7) Ma for carbonatites, (27.8±0.5) to (40.3±0.7) Ma for syenites) [10]. Besides, both rocks have similar chondrite-normalized REE distribution patterns (fig.

Viewing from the statistics data on the initial  $^{87}\text{Sr}/^{86}\text{Sr}$  of carbonatites of different ages from the Six Continents, we found that initial Sr isotopic values tend to increase from the older carbonatites (2600 Ma) to younger ones (30 Ma), and their initial Sr isotopic compositions also tend to increase from 0.7015 to 0.7050. In contrast, the initial  $^{87}\text{Sr}/^{86}\text{Sr}$  values of carbonatite from the Maoniuping REE deposit are higher, which apparently deviate from the above trend of variation. It is shown in fig. 4 that the young (<100 Ma) carbonatites in the world are mainly fallen into the area between HIMU and EM1 mantle end-member, consistent with the Sr and Nd isotopic compositions of the East African Carbonatites Line (EACL). The Sr and Nd isotopic compositions of carbonatites

carbonatites in the Indian Ambar Dangar and southern Brazil deviate from the range of EACL (fig. 4). Toyoda et al.<sup>[20]</sup> and Simonetti et al.<sup>[21]</sup> proposed that negative  $\epsilon_{\text{Nd}}$  values could be explained by the contamination of the mantle source (DMM or HIMU) with lower crustal compositions. Carbonatites in the Maoniuping area ((31.7±0.7) Ma) are obviously different from the above-mentioned carbonatites with ages less than 100 Ma, fallen into the area between EM1 and EM2, relatively close to the EM1, deviating from the EACL range in  $\epsilon_{\text{Nd}}$  vs. initial  $^{87}\text{Sr}/^{86}\text{Sr}$  diagram (fig. 4).

Because the origin of EM2 compositions is related with subducting and recycling continental materials<sup>[31]</sup>, there are two explanations for Sr and Nd isotopic compositions of carbonatites in the studied area: First, the primitive magma derived from EM1 is contaminated by crustal materials during its ascending. Second, the EM1 mantle source is mixed with the subducting crustal materials. The major elements and REE contents of carbonatites in the Maoniuping area are relatively constant. Initial  $^{87}\text{Sr}/^{86}\text{Sr}$ ,  $^{143}\text{Nd}/^{144}\text{Nd}$ ,  $d^{13}\text{C}_{\text{PDB}}$  and  $d^{18}\text{O}_{\text{SMOW}}$  values of the rocks show little variation. These features suggest that the primitive magma is little contaminated by crustal materials in the process of ascending. For this reason, the first explanation can be ruled out. That is to say, the mantle source of the Maoniuping carbonatites was mixed with subducting crustal materials. So carbonatites were formed in a subduction environment instead of a rift environment. Although the deduction requires more geological and geochemical investigations, the following facts support the conclusion.

(1) Although the Maoniuping carbonatites occur in the Panxi rift, they were emplaced during the Himalayan period when the Panxi rift closed<sup>[32]</sup>. Affected by the collision between the Indian plate and the Yangtze plate, a strongly orogenic movement is expected in this area during the Himalayan period, and inevitably the intracontinental subduction would take place, as supported by the evidence provided by Wang et al.<sup>[8]</sup>.

(2) It has been documented<sup>[33–36]</sup> that the depth of intracontinental subduction could exceed the Moho, even reach the mantle, thereby inducing partial melting of the mantle materials. The studied area is located in the transitional zone between the Panxi rift and the Longmenshan-Jinpingshan orogenic zone, where the Yangtze plate penetrated as a wedge into the crust at Longmenshan, resulting in the Moho dislocation. The subduction event took place during the Himalayan period<sup>[37, 38]</sup>. Using the “upper thrusting and lower wedging” model, Luo<sup>[38]</sup> explained successfully the evolution of tectonics-magma-ore-deposition system in the Himalayan Longmenshan-Jinpingshan orogenic zone. According to this model, the intracontinental subduction event should have taken place in the Maoniuping area during the Himalayan period.

(3) The “TNT”-shaped anomalies in the incompatible elements distribution patterns of carbonatites and syenites in the Maoniuping REE deposit (fig. 2) also support the Himalayan subduction event<sup>[39]</sup>.

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