Significance of Sm-Nd isotope systematics in crustal genesis: A case study of Archaean metabasalts of the eastern Dharwar Craton

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Determination of the age of rocks by whole rock Sm-Nd isochron method has several limitations imposed by petrogenetic processes. If the age of the rocks can be determined by other independent methods, the Sm-Nd system provides a wealth of information to understand crustal genesis. Sm-Nd isotopic studies of metabasaltic rocks of the Archaean Kolar and Ramagiri Schist belts in the eastern Dharwar Craton indicate that the system was disturbed by postmagmatic fluid alteration processes associated with terrane accretion.

1. Introduction

In studies of crustal genesis, the time of formation of rocks, nature of their sources, time of deformation and metamorphism of rocks and the nature of associated fluids, are the essential information required. Several radiogenic parent-daughter isotope systems have been used to estimate timing of the formation of rocks (and to some extent metamorphism and deformation) and to understand the nature of their sources. Among these, the Sm-Nd system is being widely used as it is considered to have the following advantages over others: (1) it is relatively less mobile (2) it is relatively more difficult to rehomogenize the Sm-Nd system in minerals and (3) ^{147}Sm has a longer half life than ^{87}Rb, making the Sm-Nd system more appropriate to study older rocks.

However, the Sm-Nd system has its own disadvantages too. To obtain a precise age estimate using the isochron method of dating, the spread in the Sm/Nd ratio of the analyzed rock samples should be large. For a suite of petrogenetically related rocks, the range in Sm/Nd ratio would be too small to get the precise isochron age. This is because, both Sm and Nd are rare earth elements (atomic numbers 62 and 60, respectively) having similar ionic radii and charge, and they are similar in their geochemical behaviour. Hence, it is difficult to fractionate them and cause variation in the Sm/Nd ratio of rocks or magmas by various melting or fractionation processes.

The extent to which the Sm/Nd and Nd isotope ratios in rocks can be disturbed is not well understood. Fluids are active in the mantle source regions before and during partial melting. Fluids also play a vital role during metamorphism and deformation of the continental crust. Several studies have reported that the Rb-Sr system can be reset or disturbed by the interaction of crustal fluids during metamorphism and deformation. If independent age information is available on the Archaean rocks, then it is possible to test the effect of fluid interaction on the Sm-Nd systematics. In this paper the problems associated with the whole-rock Sm-Nd isochron method of dating are discussed and its usefulness in understanding the nature of sources, nature of the fluids and the crustal evolution are emphasized in the context of the eastern Dharwar Craton.

Keywords. Sm-Nd isotope; Archaean metabasalt; eastern Dharwar Craton.

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2. Sm-Nd systematics

^{147}Sm decays to ^{143}Nd by emitting an alpha particle. The number of atoms of ^{143}Nd in any rock or mineral is the sum of atoms ^{143}Nd initially present at the time of its formation ($^{143}\mathrm{Nd}_i$) and ^{143}Nd atoms formed by the decay of ^{147}Sm ($^{143}\mathrm{Nd}^*$).

$$^{143}\mathrm{Nd} = {}^{143}\mathrm{Nd}_i + {}^{143}\mathrm{Nd}^*, \qquad (1)$$

$$^{143}\mathrm{Nd}^* = {}^{147}\mathrm{Sm}(e^{\lambda t} - 1). \qquad (2)$$

Substituting this in equation (1), we have

$$^{143}\mathrm{Nd} = {}^{143}\mathrm{Nd}_i + {}^{147}\mathrm{Sm}(e^{\lambda t} - 1). \qquad (3)$$

Dividing the above equation by ^{144}Nd, stable and non-radiogenic isotope of Nd, we have

$$(^{143}\mathrm{Nd}/^{144}\mathrm{Nd}) = [^{143}\mathrm{Nd}/^{144}\mathrm{Nd}]_i + (^{147}\mathrm{Sm}/^{144}\mathrm{Nd})(e^{\lambda t} - 1), \qquad (4)$$

where $\lambda = 6.54 \times 10^{-12} y^{-1}$.

The above equation is commonly used for whole rock isochron dating of rocks. In the whole-rock Sm-Nd isochron method of age determination the daughter isotope ratios (^{143}Nd/^{144}Nd) and the abundances of parent and daughter elements (Sm and Nd) are measured on several samples. Invariably in all rocks, at the time of their formation, a certain amount of isotopes of daughter element is also incorporated. Therefore, the term initial ratio ($[^{143}\mathrm{Nd}/^{144}\mathrm{Nd}]_i$ in equation 4) cannot be equal to zero and has some positive value. In a cogenetic suite of rocks, at the time of their formation ($t = 0$), all the samples should have had the same isotopic ratio of the daughter element ($[^{143}\mathrm{Nd}/^{144}\mathrm{Nd}]_i$), although they may have had different Sm and Nd abundances and Sm/Nd ratios.

Therefore, in equation (4), both time (t) and initial ratio $^{143}\mathrm{Nd}/^{144}\mathrm{Nd}_i$ are unknowns for older rocks. By solving the simultaneous equations, both t and *initial ratio* for a cogenetic suite of rocks can be calculated. A graphical solution to the simultaneous equations is given in figure 1. If 1, 2, 3 and 4 represent cogenetic suite of rocks and satisfy other conditions of isochron outlined below the slope of the line equals to the term $(e^{\lambda t} - 1)$ and the y-axis intercept corresponds to the initial ratio.

^{143}Nd/^{144}Nd ratios and ^{147}Sm/^{144}Nd ratios of samples and errors associated with their determinations are plotted in isotope evolution diagram, such as, figure 1. If the scatter of points away from the best-fit line is over and above the assigned errors, then either the assigned errors are too low or there are some other factors responsible for the scatter. These factors may include:

- mobility of parent- and/or daughter-elements and/or daughter-isotopes,
- inhomogeneity in initial ratios and
- variations in age or initial ratios of the samples plotted.

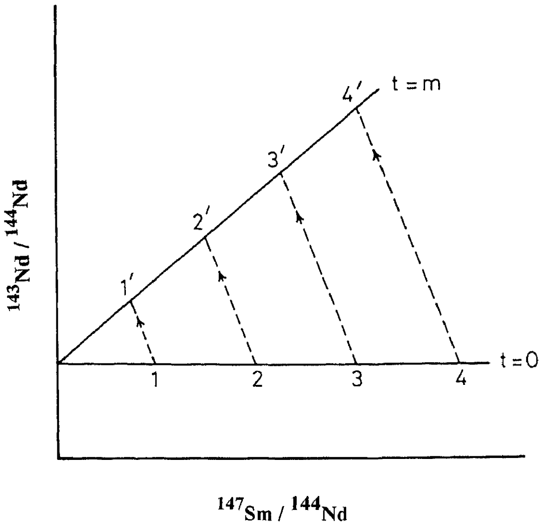

Figure 1. Sm-Nd isochron diagram. Numbers 1, 2 etc. represent rock samples which have variable Sm/Nd ratios but with one value of initial Nd isotopic ratio at time 't' before present. Numbers 1′, 2′ etc. represent the present positions of the same rock samples, as measured today.

A perfect linear fit simply means that the points plotted have satisfied statistical conditions for graphical solution of a set of simultaneous equations in the form $y = mx + c$. But it does not specify the equation. If the best fit line is interpreted as an *isochron*, then it becomes a graphical solution to the decay equation (4). If it is interpreted as a *mixing line* then it becomes a solution to a mixing equation.

Hence, there is a need to study geological and geochemical background of whole-rock samples involved, to constrain possible processes that might have caused variation in Sm/Nd ratios in a suite of samples. This is necessary for finding out the significance of the regression line. Processes that are considered to cause variation in Sm/Nd ratio of a suite of samples could be 'closed' or 'open' and are evaluated below.

3. Closed system igneous processes (first order processes)

Igneous rocks form from crystallization of melts generated by melting of deep crustal and mantle sources. Melting is always partial and crystallization could be equilibrium or fractional. These first order processes will not change the isotopic ratio of the daughter element (^{143}Nd/^{144}Nd) in a melt relative to the source. This is because isotopic fractionation is thought to be negligible at high temperatures, particularly for heavier elements.

As for the parent-daughter ratio, variation is possible by these first order processes only when the parent and daughter elements are geochemically dissimilar. For example, if plagioclase is involved during partial melting or fractional crystallization then fractionation of Sr from Rb is possible because plagioclase has a $K_d > 1$ for Sr and a $K_d < 1$ for Rb ($K_d =$ mineral-melt equilibrium distribution coefficient). Biotite can also fractionate Sr from Rb but in a sense opposite to that of plagioclase. However, there are very few common rock forming minerals that can fractionate Nd from Sm. Therefore, whereas variation in Rb/Sr ratios is theoretically possible, first order petrogenetic processes cause very little variation of Sm/Nd ratios.

Before interpreting an isochron, it is essential to know if (a) the variation in parent-daughter ratios was brought about by the first order petrogenetic processes, or (b) the variation in daughter isotopic ratios is brought about by radioactive decay only, in the samples used. There are several chemical elements whose abundance in minerals and melts are systematically changed due to the first order igneous processes. Using the isotopic composition of Nd and the elemental abundance of Sm and Nd, it may be possible to verify whether a suite of rock samples was cogenetic and had behaved as a closed system (see below).

4. Open system processes (second order processes)

There are several second order processes that can cause variation in parent-daughter ratios and in the isotopic composition of the daughter element as well. These include: (a) magma mixing, (b) crustal assimilation, (c) zone refining and (d) fluid-rock interaction. Most of the magmas that made it to the outer most layer of the earth were likely to have been affected by one or more of the above processes to varying degrees. Thus it is necessary to test whether a suite of samples are cogenetic and have formed and evolved as a closed system.

5. Evaluation of the dominant process responsible for Sm/Nd fractionation

One of the ways to do this is by studying the relationship between $f_{\mathrm{Sm/Nd}}$ and $\varepsilon_{\mathrm{Nd}}$ values in the suite of samples. Epsilon Nd ($\varepsilon_{\mathrm{Nd}}$) is the fractional difference between the $^{143}\mathrm{Nd}/^{144}\mathrm{Nd}$ ratio in the sample and the chondrite uniform reservoir (CHUR) at time t, multiplied by 10,000.

$$\varepsilon_{\mathrm{Nd}} = [(^{143}\mathrm{Nd}/^{144}\mathrm{Nd}_{\mathrm{sample}}/^{143}\mathrm{Nd}/^{144}\mathrm{Nd}_{\mathrm{CHUR}}) - 1] \times 10^4.$$

The $f_{\mathrm{Sm/Nd}}$ is the fractional difference in the Sm/Nd ratio between the sample and CHUR

$$f_{\mathrm{Sm/Nd}} = [(^{147}\mathrm{Sm}/^{144}\mathrm{Nd}_{\mathrm{sample}}/^{147}\mathrm{Sm}/^{144}\mathrm{Nd}_{\mathrm{CHUR}}) - 1].$$

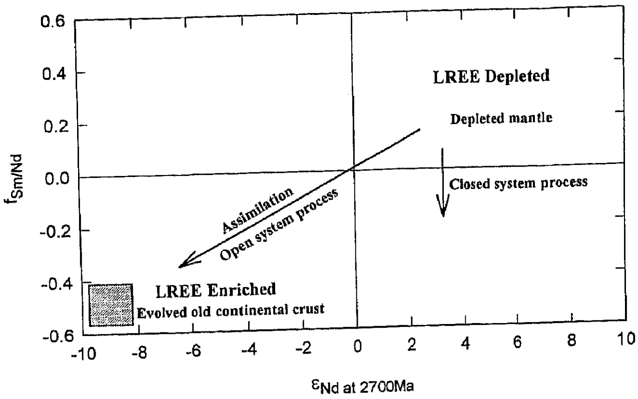

Figure 2. $f_{\mathrm{Sm/Nd}}$ vs $\varepsilon_{\mathrm{Nd}}$ schematic diagram. For explanation see the text.

$\varepsilon_{\mathrm{Nd}}$ values are calculated for the time of their formation (t) and plotted against the $f_{\mathrm{Sm/Nd}}$ as suggested by Shirey and Hanson (1986) to understand petrogenetic processes (figure 2). The light REE depleted magmas derived from long term light REE depleted sources plot in the top-right quadrant. While those derived from long term light REE enriched sources would plot in the bottom-left quadrant. If light REE enriched magmas were derived from long term depleted sources which were enriched in light REE shortly before melting by mantle-derived components, then they would plot in the bottom right quadrant. Light REE enriched magmas representing very low extents of melting of depleted mantle would also plot in the bottom right quadrant (see figure 2).

If the magmas derived from long term light REE depleted sources had undergone variable extents of contamination or assimilation by old, light REE enriched continental crustal rocks, then they would form a linear 'mixing' array as indicated in figure 2. Similar mixing trends could be formed by a suite of samples that represent magmas derived from variably contaminated long term light REE depleted sources by old enriched rocks. On the contrary a cogenetic suite of rocks that represent magmas derived either by different degrees of partial melting of a homogeneous source or by fractional crystallization of a parent magma would define a vertical spread. Such a set of rocks will have $\varepsilon_{\mathrm{Nd}}$ values indistinguishable from each other within the experimental uncertainty and may show variation in $f_{\mathrm{Sm/Nd}}$ values to the extent allowed by the closed system petrogenetic process.

Thus by using $\varepsilon_{\mathrm{Nd}}$ vs. $f_{\mathrm{Sm/Nd}}$ plot it is possible to find out the petrogenetic relationship amongst a suite of samples and then decide whether they can give any reliable age information. Even if they cannot be used for dating, they give valuable information regarding processes that might have affected the magmas or their sources. Here, we take two examples from the eastern Dharwar Craton and use them to illustrate these points.

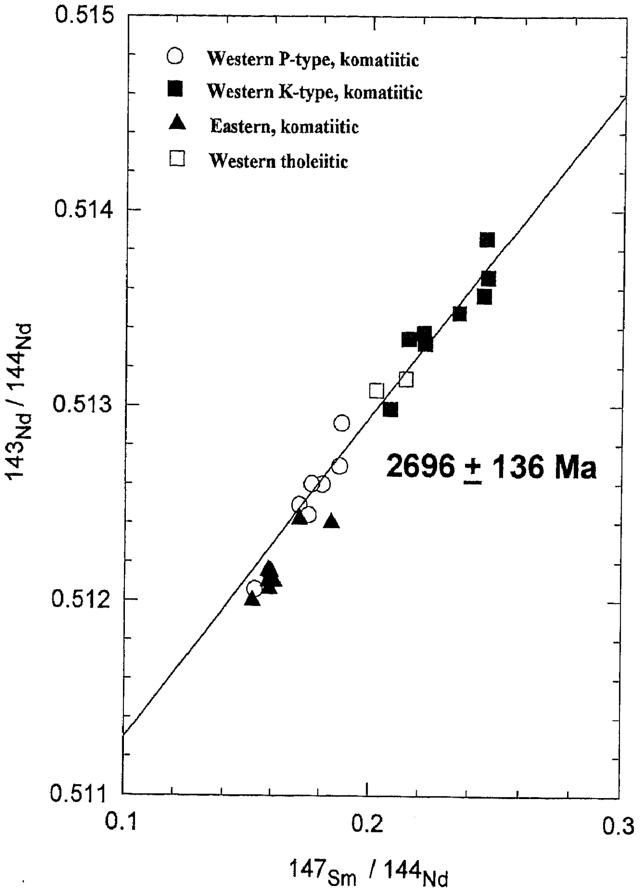

Figure 3. Sm-Nd isochron diagram of Kolar amphibolites. Komatiites from the east side of the belt are shown to have had mantle sources different from those of the west side.

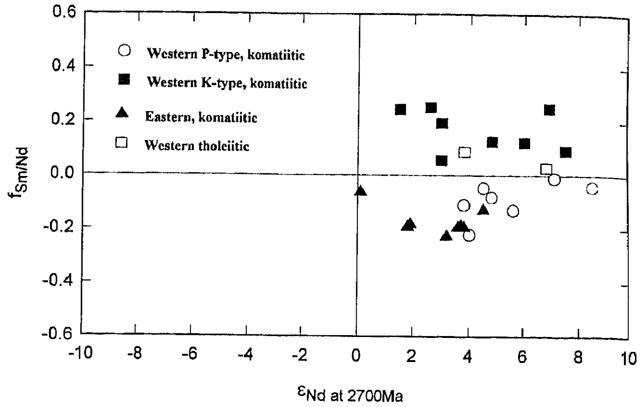

Figure 4. $f_{\mathrm{Sm/Nd}}$ vs $\varepsilon_{\mathrm{Nd}}$ diagram for Kolar amphibolites. Western komatiitic had sources that were more depleted and heterogeneous in LREE than those of the eastern komatiitic samples.

6. A summary of geochemistry of Kolar and Ramagiri schist belts

6.1 *Kolar Schist Belt*

The Kolar Schist Belt is a 4 × 80 km, N-S trending belt made up of tholeiitic to komatiitic rocks surrounded by late Archaean granitoid rocks (Rajamani *et al* 1985, 1989; Balakrishnan and Rajamani 1987; Krogstad *et al* 1989). The schist belt rocks have undergone middle to upper amphibolite grade of metamorphism. The mafic and ultramafic rocks from the eastern part of the belt have, by and large, light REE enriched REE patterns, whereas, the komatiitic rocks from the western part of the belt have Ce only depleted to light REE depleted REE patterns. The western and central tholeiitic amphibolites have nearly flat REE patterns.

Balakrishnan *et al* (1990) obtained a Pb-Pb age of 2732 ± 155 Ma on samples from an outcrop of massive metabasalts from the central part of the belt. Sm-Nd systematics on the amphibolites of the Kolar Schist Belt have been studied by Balakrishnan *et al* (1990) and only relevant aspects are discussed here. About 25 samples of light REE enriched and light REE depleted komatiitic amphibolites have been analysed. The eastern komatiitic amphibolites do not show enough spread in Sm/Nd ratio to give any geologically meaningful age (figure 3). The eastern and western komatiitic samples, together have enough spread in Sm/Nd ratio, but, as they are considered to have been derived from different mantle sources, they cannot be considered as a cogenetic suite. Hence it is not justified to fit an isochron through them.

The western komatiitic amphibolites, which include Ce only depleted (P-type) and light REE depleted varieties (K-type), together show spread in the Sm/Nd ratio and the 2696 ± 136 Ma isochron date obtained on these samples was considered to represent the time of enrichment of long term light REE depleted sources for the P-type komatiitic amphibolites, if at all it had any age significance (Balakrishnan *et al* 1990).

All the samples of the Kolar Schist Belt have positive $\varepsilon_{\mathrm{Nd}}$ values ranging from +1.5 to +8. When plotted against $f_{\mathrm{Sm/Nd}}$ they do not show any trend indicating mixing or assimilation with the older evolved continental crust (figure 4). Hence it was suggested that the spread in the $\varepsilon_{\mathrm{Nd}}$ could have been a result of mantle heterogeneity (Balakrishnan *et al* 1990).

6.2 *Ramagiri Schist Belt*

The Ramagiri Schist Belt is a 80 km long, N-S trending, trident shaped belt occurring 200 km NNW of the Kolar Schist Belt. The main central arm of the belt is made up of eastern, central and western blocks. The eastern block has amphibolite facies meta-tholeiites with LREE depleted REE patterns. Greenschist facies mafic to felsic rocks having LREE enriched REE patterns occur in the central block. Amphibolite facies meta-tholeiites with flat to slightly depleted

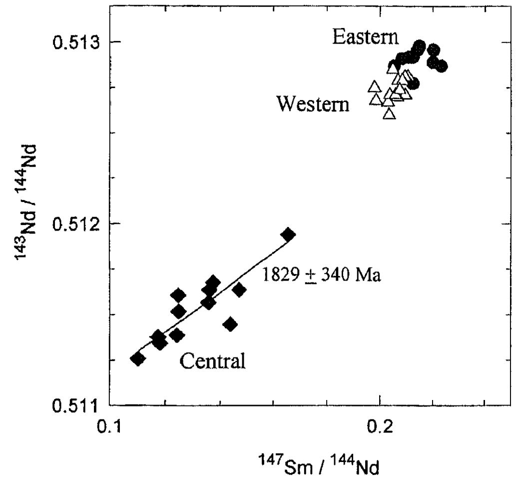

Figure 5. Sm-Nd isochron diagram of the tholeiitic amphibolites of the Ramagiri belt. Pb isotope data on the same samples from the central block tholetiic rocks (2746 ± 64 Ma) and the U-Pb Zircon age of the associated acid volcanics (2710 Ma) indicate that the Sm-Nd age of 1829 Ma has no geological significance.

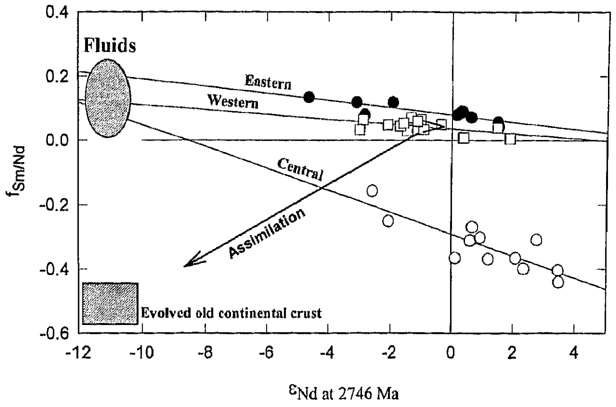

Figure 6. $f_{Sm/Nd}$ vs ε_{Nd} diagram for Ramagiri tholeiitic amphibolites. For mantle derived basaltic rocks, negative ε_{Nd} values are anomalous. This and the trends shown by the three suites of rocks indicate that their Nd isotopic systematics were disturbed by interaction with crustally derived fluids.

REE patterns occur in the western block. The analysed mafic rocks of the central block are related to each other and to a common source with limited variation in their REE patterns. The meta-tholeiitic rocks of the eastern and western blocks are related to each other but not to the central block (Zachariah 1992). The source of the central block rocks are considered to have been developed in response to mantle metasomatism by components derived from a subducting oceanic crust (Zachariah *et al* 1997). Zachariah *et al* (1995) obtained a Pb-Pb age of 2746 ± 64 Ma on a set of closely spaced meta-tholeiite samples from one outcrop in the central block, which was interpreted as the time of igneous crystallization of the rocks. Zircons separated from the acid volcanics of the central block gave a precise U-Pb age of 2710 Ma (Balakrishnan *et al* in press).

The Sm-Nd systematics of about 45 samples of the Ramagiri Schist Belt were studied (complete data set is available in Zachariah *et al* 1995). Those of the eastern and western blocks have little spread to give any meaningful age information, while those of the central block gave an Sm-Nd whole rock isochron age of 1829 ± 340 Ma (figure 5). However, as shown below, this Sm-Nd age has no geological significance as it is very different from the Pb-Pb age of 2746 ± 64 Ma and from a more precise U-Pb age of 2710 Ma on zircons.

Each of the three blocks of Ramagiri Schist Belt samples show a large range, of up to 8 units, in ε_{Nd} values (at 2746 Ma ago). The three suites of metabasalts contain some samples which have negative ε_{Nd} values. They show two distinct trends when plotted in ε_{Nd} versus $f_{Sm/Nd}$ diagram (figure 6). Interestingly, these trends when extrapolated intersect in the quadrant with positive $f_{Sm/Nd}$ and negative ε_{Nd} values. Because, the observed correlations do not point towards the field with negative ε_{Nd} and $f_{Sm/Nd}$ values representing old continental crust, they cannot be explained by either mixing of older continental crust in the sources or its assimilation by magmas represented by the Ramagiri samples. Therefore, Zachariah *et al* (1995) considered that the isotope composition and ε_{Nd} values of Ramagiri metabasalts were disturbed by interaction of fluids. As the intersection of trends defined by the two suites of samples lie between −10 and −12 ε_{Nd} values, it was suggested that the fluids were derived from older evolved continental crust.

Fluids derived from continental crust are generally thought to have light REE enriched and fractionated REE patterns, similar to the rocks that predominate the continental crust. However, the REE characteristics of the fluids constantly evolve, because, (a) they interact with compositionally different rock types, (b) undergo chemical precipitation and (c) exchange with lowering pressure and temperature and changing chemical activities of various species.

If the light REE enriched fluids interact with a large volume of light REE depleted tholeiitic rocks at lower amphibolite facies conditions, the light REE could be preferentially adsorbed as they have larger ionic radii than the heavy REE. This will result in the fluids that have passed through such rocks having light REE depleted pattern. The hydrothermal fluids associated with gold ores commonly precipitate minerals like quartz, calcite, epidote, allanite, scheelite, pyrite and other sulfides (Narayanaswamy *et al* 1960; Sivasiddaiah and Rajamani 1989). Amongst these minerals, precipitation of quartz and sulfides may not have any effect on the REE pattern of the remaining fluid, whereas precipitation of light REE bearing or enriched minerals such as allanite, calcite and epidote would result in the depletion of the light REE in the remaining fluids.

These processes may leave the Nd isotope composition of the fluids unchanged if a significant amount of Nd was not introduced into it even though the fluid has evolved from light REE enriched to depleted type. Therefore, one may expect fluids having a wide range of Sm/Nd ratios, covering from enriched to depleted types and consequently a range of $f_{\mathrm{Sm/Nd}}$ values. However these fluids may have only a small range in $\varepsilon_{\mathrm{Nd}}$ values if their Nd isotope composition had remained unchanged.

Thus the field of intersection of the trends defined by the Ramagiri samples in figure 6 defines the composition of fluids that had interacted with them. $\varepsilon_{\mathrm{Nd}}$ values for the fluids (-8 to -12) therefore warrant their derivation from older continental crustal rocks. There have been several direct and indirect evidences for the presence of old granitoid rocks in the Ramagiri and adjoining Kolar areas. These include evidences for inherited zircons in Banded Gneiss, Dod Gneiss and in the Champion Gneiss granitoid conglomerate and variably high Sr and low Nd ratios in the Dod and Dosa granodioritic rocks in the Kolar area (Krogstad *et al* 1989, 1991 and 1995) and inherited zircons in Chenna Gneiss in the Ramagiri area (Balakrishnan *et al* in press). Highly radiogenic Os isotope ratios reported from samples of ores and host rocks in the Kolar Schist Belt (Walker *et al* 1989) also points to the presence of old continental crustal rocks in this area. Interestingly, the Ramagiri belt itself includes thin marble units which are about 3098 ± 100 Ma and with an $\varepsilon_{\mathrm{Nd}}$ -1.7 (Zachariah *et al* 1996).

The fluid related alteration of Ramagiri rocks appears to have occurred at lower temperatures. The preservation of distinct Nd isotope composition in closely spaced samples collected 50 cm from each other and the alteration mineral assemblage all indicate a low temperature fluid interaction (Zachariah *et al* 1995).

Although the 2750 Ma old meta-tholeiitic rocks from all the three blocks of the Ramagiri belt are disturbed by the extraneous fluids, a 2450 ± 110 Ma old E-W trending diabase dike that cuts across the belt is not disturbed by the fluids (Zachariah *et al* 1995). This suggests that the fluid interaction is most likely to have occurred between 2750 and 2450 Ma ago. The accretion of various blocks of the Ramagiri area, the shearing of rocks and, possibly, passage of crustally derived fluids through them must have occurred within the above time interval. Such crustal processes are known to generate fluids and facilitate large scale movement of fluids through the earth's crust resulting in vein type gold mineralization (e.g., Kerrich and Wyman 1990).

Based on the continuing work on the Ramagiri Schist Belt, and with the recognition of the role of fluids in changing Nd isotope composition of sheared and altered mafic rocks, we can reevaluate the Sm-Nd systematics of the Kolar amphibolites. We wonder if the spread in the $\varepsilon_{\mathrm{Nd}}$ values of the three suites of komatiitic amphibolites of the Kolar Schist Belt could be at least partly explained by alteration by fluids derived from an old evolved continental crust. Re-Os and O isotope data on the komatiitic and tholeiitic amphibolites and gold-calcite-quartz veins indicate that the amphibolites had undergone at least two episodes of post crystallization alterations by crustally derived fluids enriched in radiogenic Os (Walker *et al* 1989). Considering that crustal Nd could bring down the $\varepsilon_{\mathrm{Nd}}$ to negative values the sources for the P- and K- type high Mg amphibolites must have had a $\varepsilon_{\mathrm{Nd}}$ value of +8 at 2700 Ma (see figure 4). If so, the mantle sources for the high Mg komatiitic magmas in the Kolar belt must have been ultradepleted for a considerable length of time 2700 Ma ago.

The $\varepsilon_{\mathrm{Nd}}$ values for the mantle sources of Ramagiri tholeiitic rocks and Kolar komatiitic rocks are distinct. They must have been derived from diverse mantle sources under different tectonic conditions. However, the rocks of the Kolar and Ramagiri schist belts were formed and accreted at similar time and affected by crustally derived fluids. The accretion of rocks in these belts is considered to have occurred ca. 2500 Ma ago.

7. Conclusion

Thus the Sm-Nd systematics is very useful in the study of Archaean rocks and in understanding the crustal processes. The system can be disturbed by hydrothermal fluid processes at least in ancient metabasaltic rocks. Even if age information cannot be obtained from an Sm-Nd isotope study of a suite of rocks the following valuable information on the petrogenetic and fluid alteration processes can be extracted:

- The nature and evolutionary history of the sources of igneous rocks.
- The extent of any open system magmatic processes like assimilation.
- The nature and source of fluids and their evolutionary history.
- The geological processes involved in the formation of the continental crust through the information obtained from the three aspects listed above.

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