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4. Petrogenetic Implications of Isotopic System

Isotope ratios in a magma are characteristic of the source region from where the magma was extracted and the ratios remain unchanged during subsequent fractionation event. This is because the mass difference between any pair of radiogenic isotopes used in geochemistry is so small that the isotopic pair cannot be fractionated by the process controlled by crystal-liquid equilibria. This simple observation has led to two important developments in isotope geochemistry, having significant petrogenetic implications. *Firstly*, distinct source region can be recognized with their own unique isotopic character and *secondly*, mixing can be recognized between isotopically distinct sources.

The different elements used in radiogenic isotope studies in geology vary greatly in their physical and chemical properties so much so that different isotope systems vary in their sensitivity to particular petrological processes. This variability is primarily due to the fact that the parent and the daughter elements behave in different ways under certain circumstances so that the two become fractionated. The extent to which an element is fractionated into the crust relative to the depleted mantle is indicated by the order of incompatibility for the elements of interest, which is Rb>Th> U >Pb> (Nd, Hf) > (Sr, Sm, Lu).

4.1 Rb-Sr System

It is evident from the order of incompatibility that Rb will be most concentrated in the crust relative to depleted mantle whereas Sr, Sm and Lu will be least concentrated. This property can be used to develop a simplified model for the long term Sr isotopic evolution of the upper mantle and the crust. Based on the extrapolation back in time from the present chondritic meteorite values, the $^{87}Sr/^{86}Sr$ ratio of the mantle about 4.6 Ga before present was about 0.699. Since then, the $^{87}Sr/^{86}Sr$ of the upper mantle has slowly increased following the decay equation represented by the growth curve.

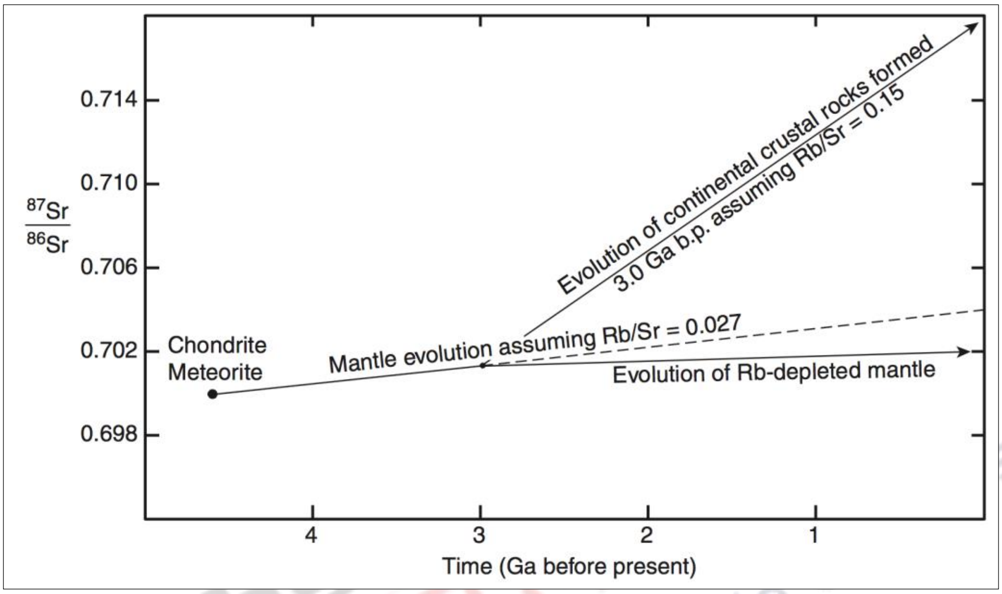

Figure 5: Estimated Rb and Sr isotopic evolution of Earth's upper mantle and continental crust, assuming a large-scale melting event producing granitic-type continental rocks at 3.0 Ga. Adopted from Winter (2011).

If at any time the mantle should be partially melted, and the melts so derived become part of the continental crust, Rb will be preferentially incorporated into the crust. Considering a hypothetical massive melting event at 3.0 Ga (Fig. 5), the subsequent mantle growth curve will have a lower slope as Rb has been selectively removed by the melts, so that less ^{87}Sr is subsequently generated. The crustal growth curve, on the other hand, has a dramatically greater slope because Rb will be greatly concentrated from a large mantle reservoir into a smaller volume of sialic crust (Fig. 5). The crustal rock evolution curve is based on an assumed initial Rb/Sr ratio of 0.15. The exact value of the $^{87}Sr/^{86}Sr$ will naturally depend on the initial Rb/Sr ratio (related to the degree of partial melting) and the age of the rock. Moreover, within the continental crust Rb and Sr are further separated by remelting, metamorphism and sedimentation as Sr is partitioned into and retained by plagioclase, whereas Rb is preferentially partitioned into the melt or fluid phase. Therefore, the Rb-Sr system stands out to be an excellent petrogenetic indicator for crustal and mantle processes.

4.2 Sm-Nd System

Analogous to the Sr isotopic evolution over time, a simplified model for the evolution of Nd isotope can also be developed. CHUR (Chondrite Uniform Reservoir) is an estimate of the average chondritic composition by DePaolo and Wasserburg (1976). From the chondrite model, the CHUR bulk Earth line shows the $^{143}Nd/^{144}Nd$ evolution of the mantle if it were a closed system (Fig. 6). Because ^{144}Nd is not a radiogenic product, it is constant over time. As ^{147}Sm breaks down to ^{143}Nd, the $^{143}Nd/^{144}Nd$ ratio gradually increases, just as $^{87}Sr/^{86}Sr$ did in figure 5.

Similar to the Rb-Sr system, if at any time the mantle is partially melted (postulated here to be 3.0 Ga), the parent and the daughter elements will be fractionated (Fig. 6). However, the Sm-Nd systematics will behave in an opposite manner to the Rb-Sr systematics. Nd being more incompatible than Sm, partial melting of the mantle removes more Nd than Sm from the mantle. This depletes the mantle in the *daughter* isotope, whereas in the Rb-Sr system, the *parent* is depleted. As a result of the daughter depletion in partially melted areas of the mantle, the higher Sm/Nd ratio will, over time, generate more radiogenic ^{143}Nd from ^{147}Sm relative to the original ratio of $^{143}Nd/^{144}Nd$. On the other hand, the partial melt derived from the mantle, mostly concentrated in the crust, will reflect a lower Sm/Nd ratio. The low Sm/Nd results in the generation of smaller quantities of ^{143}Nd and will follow the melt trend in figure 6. Rocks derived from any source will naturally have the same $^{143}Nd/^{144}Nd$ as that of the source at the time of partial melting. Therefore, the initial isotopic ratio $(^{143}Nd/^{144}Nd)_0$, which can be easily obtained from the isochron method, will clearly reflect the source of the rock, either mantle or crust, and also indicate any mixing between the two.

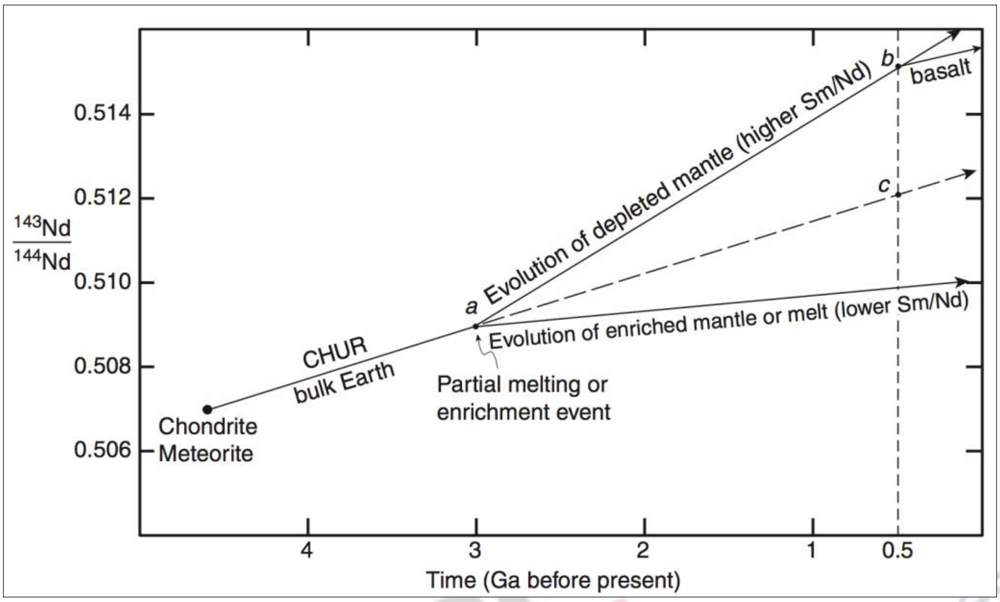

Figure 6: Estimated Sm and Nd isotopic evolution of Earth's upper mantle and continental crust, assuming a large-scale melting event producing granitic-type continental rocks at 3.0 Ga. Adopted from Winter (2011).

4.3 Degree of Nd and Sr Enrichment (ε_{Nd} and ε_{Sr})

As the differences in Nd isotope ratios are small, DePaolo and Wasserburg (1976) introduced the term ε (epsilon) to express the degree of Nd enrichment. ε is defined as:

$$\varepsilon_{Nd} = \left[\frac{(^{143}Nd/^{144}Nd)_0}{(^{143}Nd/^{144}Nd)_t^{CHUR}} - 1\right] * 10^4 \tag{14}$$

where:

$(^{143}Nd/^{144}Nd)_t^{CHUR}$ = $^{143}Nd/^{144}Nd$ ratio of CHUR at time (*t*) of formation of the rock.

A positive ε_{Nd} value is depleted (higher ^{143}Nd), and a negative ε_{Nd} is enriched (lower ^{143}Nd), both with respect to the CHUR standard (corrected for time along the CHUR bulk Earth line in figure 6). Thus a positive ε_{Nd} for a rock implies that it was derived from a depleted mantle source, and a negative εNd indicates that the rock was derived from either an enriched mantle or a crustal source enriched over time.

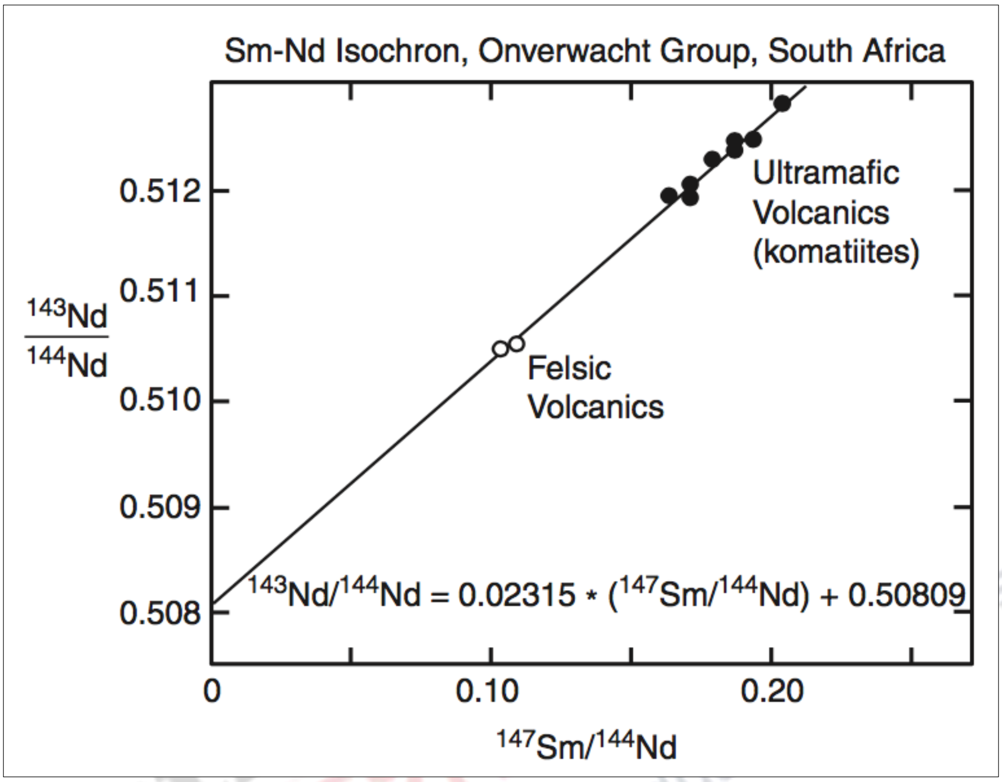

Figure 7: Sm-Nd isochron for whole-rock analysis of ultramafic and felsic volcanics from the Archean Onverwacht Group of South Africa. Age calculated as 3.540±30 Ma. Adopted from Winter (2011)

As a quantitative example, consider the 3.54 Ga volcanics of the Onverwacht Group in Figure 7. $(^{143}Nd/^{144}Nd)_0$is the intercept of the regression line, = 0.5080. $(^{143}Nd/^{144}Nd)_{CHUR}$ at 3.54 Ga can be calculated from a form of Equation (8),

$$(^{143}Nd/^{144}Nd)_{today}^{CHUR} = (^{143}Nd/^{144}Nd)_t^{CHUR} + (^{147}Sm/^{144}Nd)_{today}^{CHUR}(e^{\lambda t} - 1)$$

by substituting $t = 3.54*10^9$, yielding 0.508031. Substituting this value into Equation (14) yields $\varepsilon_{Nd} = [(0.50809/0.508031) - 1]*10{,}000 = 1.16$, suggesting a slightly depleted mantle source.

A similar calculation can be used to derive ε_{Sr} in the Rb-Sr system, where:

$$\varepsilon_{Sr} = \left[\frac{\left(\frac{^{87}Sr}{^{86}Sr}\right)_0}{\left(\frac{^{87}Sr}{^{86}Sr}\right)_t^{CHUR}} - 1 \right] * 10^4 \qquad (15)$$

A positive ε_{Sr} value is enriched (higher ^{87}Sr), and a negative ε_{Sr} is depleted (lower ^{87}Sr), both with respect to the CHUR standard. Thus a negative ε_{Sr} for a rock implies that it was derived from a depleted mantle source, and a positive ε_{Sr} indicates that the rock was derived from either an enriched mantle or a crustal source enriched over time.

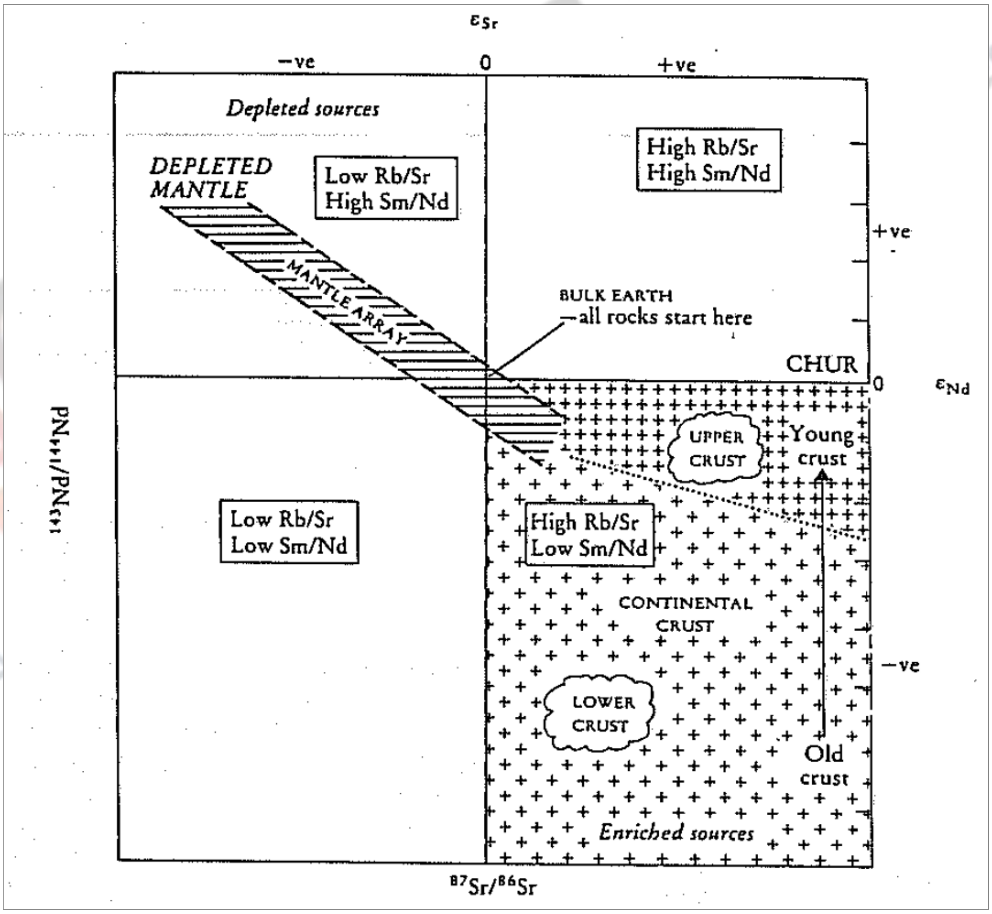

Figure 8: A ^{143}Nd/^{144}Nd vs. ^{87}Sr/^{86}Sr (ε_{Nd} vs. ε_{Sr}) isotope correlation diagram showing the relative positions of depleted and enriched mantle sources. Most non-enriched mantle reservoirs plot in the upper left ‘depleted’ quadrant whereas most crustal rocks plot in the lower right ‘depleted’ quadrant. Adopted from Rollinson (1993).

A $^{143}Nd/^{144}Nd$ vs. $^{87}Sr/^{86}Sr$ (ε_{Nd} vs. ε_{Sr}) isotope correlation diagram can be drawn (Fig. 8) to show the relative positions of depleted and enriched mantle sources. Most depleted mantle reservoirs plot on the upper left 'depleted' quadrant whereas most crustal rocks plot in the lower right 'enriched' quadrant. Upper and lower crust tend to plot in different positions in the crustal quadrants (DePaolo and Wasserburg, 1979). Thus the combination of the Rb-Sr and Sm-Nd isotopic systems is a wonderful petrogenetic indicator and easily reflects the source of the rocks.

5. Radiometric Dating of Whole rocks and Single Minerals

The numeric value of *t* can be conveniently calculated from the concept of decay of a radioactive parent to stable daughter and using the geochronological equations developed for each parent-daughter pair discussed earlier in the chapter. However, it has been commonly noticed that detailed geochronological study of slowly cooled rocks tend to yield discordant results when several different methods of age determination involving different isotopic systems are used. Therefore, it is very important to look closely at different isotopic systems in an attempt to understand what the different forms of geochronological data mean and what is that we are actually measuring when we claim to have calculated the geological age of a rock. In this respect, two important areas to understand are the concepts of *blocking temperature* and *geological ages*.

5.1 Blocking Temperature

The temperature at which a system is closed to any isotopic exchange with the surroundings (behaves as a closed system) is known as the blocking temperature. At this temperature the isotopic clock is switched on. Different minerals have different blocking temperatures for various isotopic systems. Therefore, we always get the age of a mineral at which it has just crossed the blocking temperature. In case of a mineral crystallizing from a magma, the blocking temperature might be same or different from the temperature of crystallization. This depends on the mineral being used for dating and its blocking temperature with respect to a particular isotopic system. Figure 9 illustrates the relationship between age and blocking temperatures of several minerals with respect to various isotopic systems.

5.2 Concept of Geological Age

(a) Crystallization age: It is the age when a mineral in a rock crystallized from magma. In case of an igneous rock, the crystallization age of a mineral records the magmatic age of the rock. In case of a metamorphic rock, the crystallization age denotes the age at which the metamorphic mineral formed.

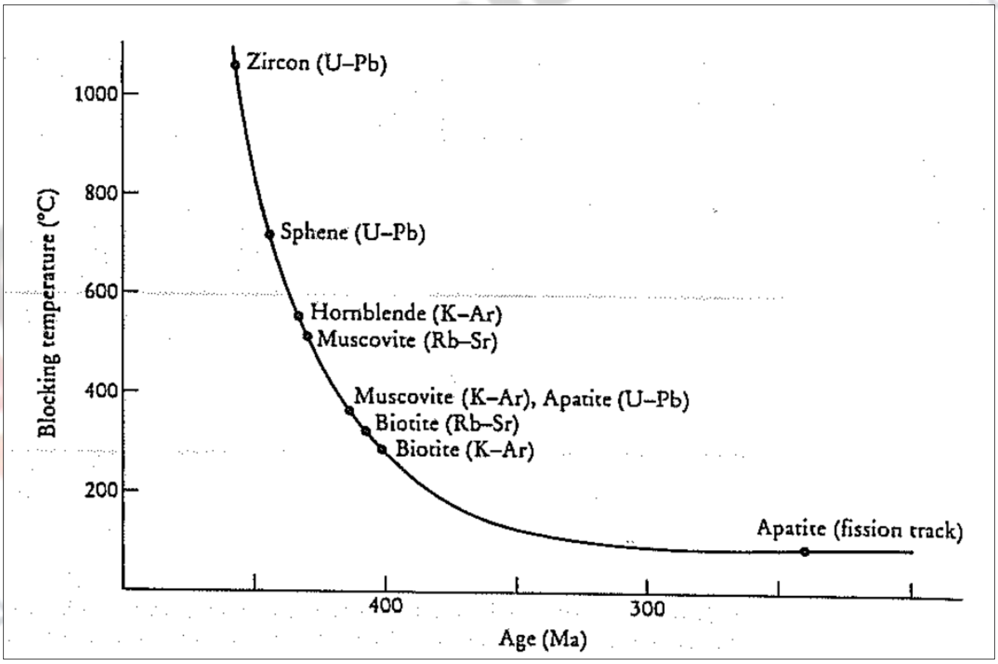

Figure 9: A plot of mineral age vs. blocking temperature for the Glen Dessary syenite from Scotland. The mineral ages define a cooling curve for the pluton. The mineral on which each age determination was made and the method used are indicated. Adopted from Rollinson (1993).

(b) Cooling age: In an igneous rock, it is the time after solidification of the melt at which the mineral passes through the blocking temperature. In a metamorphic rock the term ‘cooling age is’ conveniently used to describe the time, after the main peak of metamorphism, at which a mineral which has

crystallized at the metamorphic peak passes through its blocking temperature.

(c) Metamorphic age: It is the age at which peak metamorphism occurred. To find the age of peak metamorphism, the mineral which has the blocking temperature nearest to the peak temperature conditions need to be used.

(d) Crust formation age: This is the time of formation of a new segment of continental crust by fractionation of material from the mantle. Whether or not it is possible to determine directly the crust formation age will depend upon the geological history of the segment of the crust.

6. Whole rock Ages

As the name suggests, the whole-rock ages are obtained from the bulk composition of the rock and not from the composition of single minerals. The whole rock is normally powdered and a small amount of that powder is used for analysis. Whole-rock systems are commonly used to determine the age of crystallization of an igneous rock or the age of the metamorphic peak during metamorphism. Sometimes it is possible to determine the age of crust formation as well. The Rb-Sr and Sm-Nd systems are most commonly used for determining whole-rock ages.

6.1 The Rb-Sr System

The rubidium-strontium isotopic system is one of the most widely used isotopic whole-rock methods, for the most crustal rocks containing sufficient Rb and Sr (between 10 and 1000 ppm) as the chemical separation of elements and mass spectrometry relatively straightforward. It is a versatile method and can be applied to a range of whole-rock compositions. Equation (6),

$$^{87}\mathrm{Sr}/^{86}\mathrm{Sr} = (^{87}\mathrm{Sr}\ /^{86}\mathrm{Sr})_0 + {}^{87}\mathrm{Rb}/^{86}\mathrm{Sr}\ (e^{\lambda t} - 1)$$

can be used as the geochronological equation. ^{87}Sr/^{86}Sr and ^{87}Rb/^{86}Sr ratios can be directly measured from the sample using a mass spectrometer. The initial (^{87}Sr /^{86}Sr)$_0$ ratio in the rock during the time of its formation can be obtained using the isochron method from different co-genetic samples having different initial Rb/Sr ratio. λ being a constant, t can be easily calculated from the above equation.

Disadvantages: The results of Rb-Sr geochronology are not always easy to interpret as both Rb and Sr are relatively mobile elements, so that the isotopic system may readily be disturbed either by influx of fluids or by a later thermal event. Therefore, the Rb-Sr isochrons are less useful in constraining crust forming ages. Nevertheless, if an Rb-Sr isochron is obtained it can usually be attributed to a definite event such as the age of metamorphism or alteration, or the age of diagenesis in sedimentary rocks, even if the primary age of the rock cannot be determined.

6.2 The Sm-Nd System

The elements Sm and Nd, being more compatible than Rb and Sr, are much less mobile and may be used to 'see through' younger events in rocks whose Rb-Sr isotopic chemistry may have been disturbed during younger events. Thus the Sm-Nd technique seems to be a more efficient whole-rock method from which crust formation ages may be determined. Equation (8),

$$^{143}Nd/^{144}Nd = (^{143}Nd/^{144}Nd)_0 + {}^{147}Sm/^{144}Nd\,(e^{\lambda t} - 1)$$

can be used as the geochronological equation. Similar to that of the Rb-Sr system, $^{143}Nd/^{144}Nd$ and $^{147}Sm/^{144}Nd$ ratios can be directly measured from the sample using a mass spectrometer. The initial $(^{143}Nd/^{144}Nd)_0$ ratio in the rock during the time of its formation can be obtained using the isochron method from different co-genetic samples having different initial Sm/Nd ratios.

Disadvantages: The primary limitation of the method is the long half-life ($T_{1/2} = 1.06 * 10^{11}$yr) for the decay of $^{147}Sm \rightarrow {}^{143}Nd$, meaning that it is only efficiently applicable to very old rocks. Another limitation is the relatively small variation in Sm/Nd ratio in most cogenetic rock suites. This problem can be overcome by combining a wide range of lithologies on the same isochron in order to obtain a spread of Sm/Nd ratios. However, this might result in plotting of samples with different sources (having different initial $^{143}Nd/^{144}Nd$ ratios) and different evolutionary histories being plotted on the same isochron, giving rise to some meaningless results.

A combination of both the Rb-Sr and Sm-Nd whole-rock geochronology on suitable rocks might provide a better understanding of the evolutionary history of the rocks. The Sm-Nd system may be used to determine the crust formation ages while the Rb-Sr system may indicate the age of metamorphism, fluid influx and alteration etc.

7. Dating of Single Minerals

Apart from whole rocks, the constituent minerals of the rocks can also be dated separately using a favorable isotopic system. The mineral to be dated can be physically separated from the rest before analysis or they can be dated by various in-situ techniques. Four popular isotopic systems commonly used for mineral dating have been discussed below.

7.1 Rb-Sr Mineral Ages

Igneous rocks of granitic composition and metamorphic rocks of pelitic composition commonly have minerals like muscovite and biotite which are rich in Rb and therefore frequently dated by the Rb-Sr method. Ages are calculated using a two point isochron in which a mineral with low concentration of Rb, such as plagioclase feldspar, is used along with muscovite or biotite to get a control on the initial $^{87}Sr/^{86}Sr$ ratio. The present $^{87}Sr/^{86}Sr$ and $^{87}Rb/^{86}Sr$ ratios are measured by a mass spectrometer and substituted in Equation (6) to obtain the value of *t*. The geological interpretation of the obtained mineral age depends primarily on whether the rock is igneous or metamorphic. When mineral dates obtained from one rock specimen or from a suite of cogenetic igneous rocks are in agreement, they are said to be ‘concordant’. Unfortunately, ‘discordance’ of mineral dates is more common than ‘concordance’. The reason is that the constituent minerals in the rock may easily gain or lose Rb and/or Sr during a later event, which is the major disadvantage of the method. In such cases, the obtained mineral ages must be interpreted with caution

7.2 K-Ar Mineral Ages

The K-Ar system is most commonly applied in K-rich minerals like biotite, muscovite, hornblende and alkali feldspar. This is the most straightforward method of mineral dating as the minerals do not contain any Ar at the time of its formation, meaning the initial $^{40}Ar/^{38}Ar$ is zero. The Ar that is measured during dating is entirely from the radioactive decay of ^{40}K. So the ages can be obtained from single samples by measuring $^{40}Ar/^{38}Ar$ and $^{40}K/^{38}Ar$ directly from the sample and substituting the values in Equation (7).

The major *disadvantage* of this method is the problem of argon loss. Ar being a noble gas is easily lost in the atmosphere, either partially or completely, even when the minerals are subjected to slight disturbances. So the ages obtained are sometimes very difficult to interpret. The other problem is that the blocking temperature of Ar diffusion is rather low for most minerals. For example, the blocking temperature vary from ~300-450ºC for biotite, 350±50ºC for muscovite and 530±30ºC for hornblende. Therefore, even if there is no Ar-loss, the age determined is likely to be lower than the true age unless the rock cooled very rapidly.

7.3 Sm-Nd Mineral Ages

The Sm-Nd system can be efficiently applied to garnet bearing rocks as garnets have high Sm/Nd ratios, which allows the slope of a mineral isochron to be accurately determined, generating precise mineral isochron and the initial $^{143}Nd/^{144}Nd$ ratio. Similar to the Rb-Sr mineral ages, the Sm-Nd garnet ages are determined on two point isochrons between garnet and one or more of the minerals with low Sm/Nd ratios like plagioclase, clinopyroxene, orthopyroxene and hornblende. The $^{143}Nd/^{144}Nd$ and $^{147}Sm/^{144}Nd$ are measured from the sample and substituted in Equation (8) to obtain a value of *t*. The only *disadvantage* of this method is that it is mostly suitable for older rocks having garnet in the mineral assemblage.

7.4 U-Pb Mineral Ages

The U-Pb dating method has been extensively and most effectively applied to minerals like zircon ($ZrSiO_4$) and monazite [$(Ce,La)PO_4$]. But the method is also applicable to minerals like sphene ($CaTiSiO_5$), baddeleyite (ZrO_2), perovskite ($CaTiO_3$), etc. As discussed in the previous sections, the age of a sample containing uranium can be calculated using Equations (9) and (10) and all concordant points should lie on the *concordia* curve in the $^{207}Pb^*/^{235}U$ vs. $^{206}Pb^*/^{238}U$ plot. If the system experiences loss of Pb, the $^{207}Pb^*/^{235}U$ and $^{206}Pb^*/^{238}U$ ratios in a population of minerals tend to define a linear array, called discordia, with an upper and lower intercept on the concordia, which may have specific geological significance. This is an additional advantage of this method that discordant ages can also provide additional information about the age and evolutionary history of the rock.

Zircon has long been recognized as a robust U-Pb geochronometer. It has a very high closing temperature (>1000ºC) and prevents loss of Pb by diffusion after crystallization. Another important aspect of zircon is that in ideal situation it does not incorporate any Pb in its structure during crystallization, hence the entire Pb is radiogenic. Zircon is also mechanically and chemically durable and the isotopic clock is not easily reset in response to weak tectonothermal events. Microanalysis and high-precision mass spectrometry combined with cathodoluminescence (CL) imaging has revolutionized zircon geochronology, allowing us to obtain ages from texturally complex and zoned zircon crystals that record different times of their growth.

Magmatic zircon grains in felsic igneous rocks (Fig. 10a) can be used to date the time of crystallization of the magma. Granitic rocks which are melts from older crust may contain inherited zircon, overgrown by new magmatically crystallized zircon. The date from the inherited part gives the age of the crustal source while the date from the overgrowth may give the age of new zircon crystallization. Metamorphosed igneous rocks may preserve magmatic zircon cores overgrown by metamorphic zircon rims (Fig. 10b). Metamorphosed pelitic rocks may contain zircon which formed during metamorphism (Fig. 10c). Dates from different zones may constrain the age of crystallization of the igneous protolith and the age of metamorphism thereafter. Detrital zircon grains in sediments may date the provenance of the sediments, and in metasedimentary rocks, detrital zircon grains with metamorphic overgrowths may date the provenance of the sedimentary protolith as well as the age of the metamorphic event. Therefore, U-Pb dating of zircon in a variety of different rocks may be used to constrain ages of a wide range of geological events.

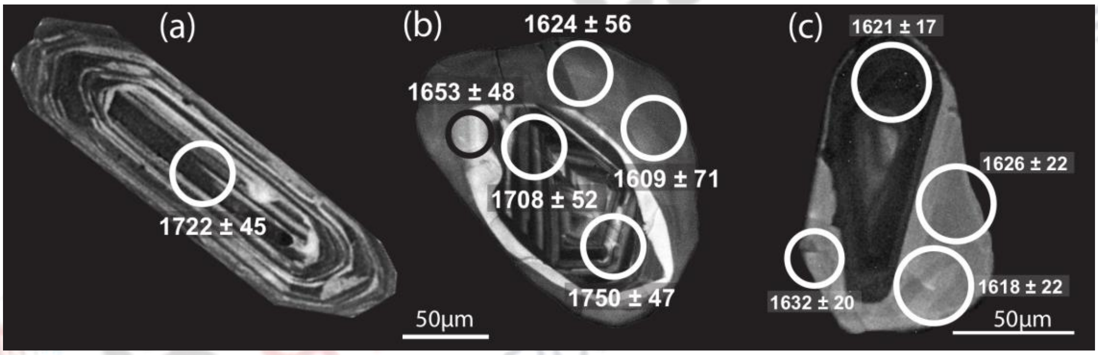

Figure 10: (a) Magmatic zircon grain, (b) magmatic zircon with metamorphic rim, and (c) metamorphic zircon grain. Image taken from Sarkar et al. (2014; 2015).

Monazite has also been widely used for U-Pb mineral dating as it also has very high closing temperatures (>1000ºC), does not incorporate Pb in its structure during crystallization and Pb diffusion is negligible even at high temperatures. Texturally controlled in-situ monazite dating has been proved to be a very effective method, especially in polymetamorphosed terranes. In-situ techniques with high spatial resolution allow selecting grains in defined textures of the rock. Consequently, these textures can be correlated with a specific stage of the tectonometamorphic evolution, thereby enabling us to resolve complex evolutionary history of rocks.

However, monazite is not as durable as zircon and the isotopic clock can be reset in response to weaker tectonothermal events, especially in the presence of fluids, whereas zircon may remain unaffected. Therefore, the combination of zircon and monazite dating of the same samples is ideal to look through the timings of events in multiply deformed and metamorphosed high-grade terranes.

Frequently Asked Questions-

Q1. Derive the basic equation required for radiometric dating from the fundamental concept of radioactivity that $-dN/dt \propto N$?

Q2. If a mantle source rock is partially melted, how will $^{87}Sr/^{86}Sr$ differ between the partial melt and the refractory solid residue? Will crystal fractionation affect the ratio? Explain

Q3. $^{147}Sm \rightarrow {}^{143}Nd$ behaves similarly to the Rb-Sr system. Both Sm and Nd are rare earth elements, and Sm has a larger atomic number than Nd. Which is preferentially fractionated into partial melts, and is the effect as great as for Rb-Sr? How does this affect the $^{143}Nd/^{144}Nd$ ratio in the crust and mantle? Explain

Q4. The following isotopic data for a whole-rock (WR) sample and for plagioclase (Plg), pyroxene (Px), and ilmenite (Ilm) mineral separates in an Apollo 12 lunar basalt (sample 12014)

Mineral	Rb (ppm)	Sr (ppm)	$^{87}Rb/^{86}Sr$	$^{87}Sr/^{86}Sr$
WR	0.926	90.4	0.0296	0.70096
Plg	0.599	323	0.00537	0.69989
Px	0.386	22.7	0.0492	0.70200
Ilm	3.76	96.5	0.1127	0.70490

What is the age of this rock?

Hint: The age and the initial Sr isotopic ratio, $(^{87}Sr/^{86}Sr)_0$, can be determined by plotting these data on an isochron diagram.

Q5. Explain what is meant by the degree of enrichment of Nd and Sr*(εNd and εSr)?* How is it helpful in recognizing the source and evolution of rocks?

Multiple Choice Questions-

1. Which is the most suitable mineral to determine the age of crystallization of a granitic rock?

a) Biotite
b) Hornblende
c) Zircon
d) Garnet

Ans: c

2. ε_{Nd} and ε_{Sr} are positive in

a) Crust and mantle respectively
b) Mantle and crust respectively
c) In the crust
d) In the mantle

Ans: b

3. If we start with 100gm ^{238}U, what would be the amount of ^{238}U and ^{206}Pb after 4 half-lives?

a) 25 and 75 grams
b) 12.5 and 87.5 grams
c) 6.25 and 93.75 grams
d) 3.12 and 96.88 grams

Ans: c

4. Which isotopic system is the most suitable for the study of crustal rocks?

a) Sm-Nd
b) Rb-Sr
c) U-Pb
d) K-Ar

Ans: b

5. Which among the following minerals is commonly dated by K-Ar isotopic system?

a) Garnet
b) Zircon
c) Monazite
d) Muscovite

Ans: d

Suggested Readings:

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