

# Calculation of Rare Earth Element Patterns in Magmatic Fluids: Evidence for Origin of the Lake George Sb-Au-W-Mo Ore Deposit, New Brunswick, Canada

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**Abstract:** This contribution illustrates the procedures of calculating rare earth element (REE) patterns of magmatic fluids in equilibrium with granitic melts. These patterns can then be compared to those of altered and (or) mineralized rocks in order to investigate the source of the ore-fluids. This technique is applied to the Lake George Sb-Au-W-Mo mineral deposit, New Brunswick, Canada. The results suggest that ore-fluids are dominated by magmatic fluids, consistent with the origin of intrusion-related mineralization systems. The magmatic hydrothermal fluids emanating from progressively cooling, voluminous and volatile-saturated magma at depth may have reacted with earlier magmatic sulphide minerals formed by liquation in the Lake George granodiorite stock, scavenged ore components (e.g., Au, S) in the sulphides, which would have enhanced ore materials in the ore-fluids and precipitated ore minerals in hydrofractures and pre-existing faults that crosscut graphitic country rocks (e.g., greywacke) within the metamorphic halos and resulted in intrusion-related gold-antimony mineralization associated with an earlier W-Mo-(Au) mineralization.

**Keywords:** Rare earth element (REE) patterns, partition coefficients, granitoid, intrusion-related gold system, Lake George.

## INTRODUCTION

For simplicity, granitoid is used as a general term for granitic melt crystallized during cooling. During crystallization, a magmatic volatile (or vapour) phase (MVP) is lost to surrounding country rocks. Up to 5 wt% water dissolved in the magma may have been lost during the solidification process [1]. This magmatic fluid may contain a significant amount of various metals; if precipitated in a suitable geological site(s), a mineral deposit may form. Experimental and empirical investigations of element partitioning between the MVP and granitic melt have made it possible to simulate the composition of MVP [2-4]. These data can be used to compare with observed geochemical data of ore deposits and associated altered rocks, which may provide a key clue as to the contribution of ore-forming materials originating from the magmas or intrusions.

This paper presents the procedures of calculating REE patterns of MVPs on the basis of published experimental partition coefficient data. As a case study, this technique is applied to tracing the REE patterns of magmatic fluids associated with the Lake George granodiorite, which is then compared with altered quartz-feldspar porphyry that host mineralization. Detailed calculation procedures together with a spreadsheet are provided, so that similar computation may be performed for other deposits.

## PARAMETER SELECTION

The context of element partition coefficient between MVP and silicate melt used in this study is the same as that suggested by [5, 6]. REE partition coefficient ( $k_p^{REE}$ ) between MVP and silicate melt is defined as: ppm REE concentration in vapour phase divided by ppm REE concentration in silicate melt. Similarly, chlorine partition coefficient ( $k_p^{Cl}$ ) is defined as: ppm chlorine content in vapour phase divided by ppm chlorine content in silicate melt. It is noted that partition coefficient is dimensionless, and is controlled by the pressure, temperature, and composition of magmatic-hydrothermal systems in question [1-9]. In addition, the value of  $k_p^{Eu}$  may be a function of oxygen fugacity of the system, because Eu has two valences, i.e.,  $Eu^{3+}$ ,  $Eu^{2+}$ , depending  $f(O_2)$  [5].

REE concentration of melt is represented by the data of fresh bulk-rock analysis; REE data of altered rocks are considered to represent the result of the fresh rock reacting with or superimposed by MVPs. If this assumption is true, the signature of MVPs may be identified.

## CALCULATION PROCEDURES

A spreadsheet program is compiled to carry out the calculation, which is available upon a request of the author. Detailed procedures of calculation are described as follows step by step, and an example of the calculation is presented in Table 1.

- Calculate the chlorine molarity ( $m_{Cl}^m$ ) of granite melt that is based on chlorine content of granite or based on the methods by [4], although it is difficult to acquire chlorine concentration in granite melt [10].

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**Table 1. Calculation of REE Concentrations in Magmatic Volatile Phases**

REE Content (ppm) in Lake George Granodiorite (Data from [17])		
	A	B
La	31	31
Ce	51	51
Nd	13.7	13.7
Sm	4.2	4.2
Eu	1.1	1.1
Gd		
Tb	0.8	0.8
Yb	2.2	2.2
Lu	0.3	0.3
Cl	1000	
15 wt% NaCl in primary fluid inclusion in apatite (data from [13])		
<i>Calculation of REE partition coefficient</i>		
		<i>Equation</i>
m(m-Cl)	0.0282	1
m(v-Cl)	<b>1.23</b>	<b>2.56</b>
K_Cl (2kb)	43.5	43.5
		Kilinc and Burnham (1972)
K_La	0.0637	0.6762
K_Ce	0.0795	0.6710
K_Nd	0.0555	0.6500
K_Sm	0.0480	0.5344
K_Eu	0.0933	0.2554
K_Gd	0.0559	0.4688
K_Tb	0.0384	0.4092
K_Ho	0.0327	0.3179
K_Yb	0.0320	0.2167
K_Lu	0.0219	0.1795
<i>Calculation of REE concentrations (ppm) in MVPs</i>		
La_v	1.97	20.89
Ce_v	4.07	34.36
Nd_v	0.76	8.90
Sm_v	0.20	2.24
Eu_v	0.10	0.28
Gd_v	0.20	2.19
		(estimated from Sm and Tb)
Tb_v	0.03	0.33
Yb_v	0.07	0.48
Lu_v	0.01	0.06
<i>Condrite-normalized (Boynston 1984) REE in MVPs</i>		
La_v	6.35	67.40
Ce_v	5.04	42.52
Nd_v	1.27	14.84
Sm_v	1.03	11.51
Eu_v	1.37	3.75
Gd_v	0.78	8.44
Tb_v	0.65	6.91
Yb_v	0.34	2.28
Lu_v	0.23	1.85

$$m_{Cl}^m = \frac{(\text{chlorine content} \div 35.5) \times [1000 \div (100 - \text{chlorine content})]}{1} \quad (1)$$

where chlorine content (in whole-rock sample) unit is wt%.

Using this formula (eq. 1), chlorine molarity of the sample (granite) is calculated. This chlorine molarity is assumed to represent a minimum value of the corresponding granitic melt from which the granite solidified, because much chlorine may have been lost [1, 8] due to high chlorine partition coefficient between magmatic volatile phase and silicate melt at magmatic conditions [5, 10-12].

Using the methods of Piccoli and Candela [4], a more accurate chlorine molarity of granite melt may be achieved. Their methods require chemical composition data of apatite, or bulk composition of apatite associated with the main intrusion. Here, for simplicity estimated chlorine molarity of granite melt is computed only based on the chlorine content of the studied samples by using eq. 1, which would provide a minimum value for chlorine molarity of granite melt in question.

- Calculate the chlorine molarity ( $m_{Cl}^v$ ) of aqueous vapour phase in equilibrium with the granite melt at magmatic P-T conditions in terms of chlorine partition coefficient ( $k_p^{Cl}$ ) [5, 7]. The calculation can be made by using the following equation if,  $m_{Cl}^m$  on the basis of Step 1 is given above (see column B in Table 1).

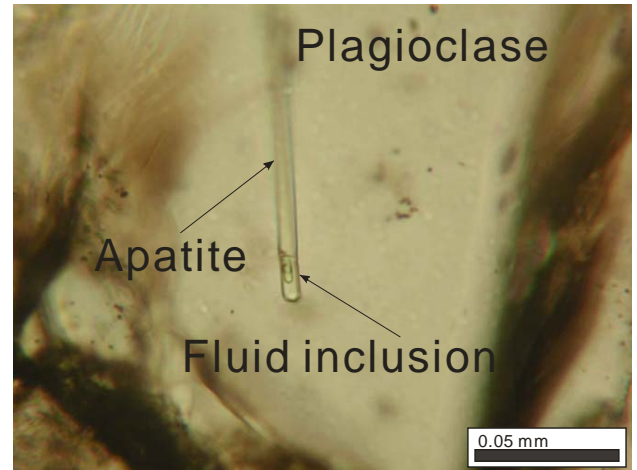
$$m_{Cl}^v = k_p^{Cl} \times m_{Cl}^m \quad (2)$$

Kilinc and Burnham [5] reported chlorine partition coefficients between chloride-bearing fluids and granitic melts (composition ranging from granite to granodiorite and trondjemite) under pressure of 2 to 8 kb and near 700 and 750°C isotherms. Their data show that the values of  $k_p^{Cl}$  are strongly pressure dependent, increasing from 43.5 at 2 kb to 83.3 at 6 kb, and then decreasing sharply to 13.0 at 8 kb. A similar result was reported by [7] for the pressure range of 2 to 4 kb; Webster *et al.* [7] noticed that the fluorine-free system exhibits higher values of  $k_p^{Cl}$ .

These results suggest that chlorine partitions strongly in favour of aqueous fluids under magmatic conditions, although some pressure effects were also observed. However, for haplogranite melts containing high fluorine (> 7 wt%) and low chlorine (0.12 wt%) at 2 kb and 1000°C, chlorine partitions in favour of the granite melt [7]. So far, there has not been a report that a natural granitic melt (magma) to contain such anomalously high fluorine as shown in that experiment [7].

Also, using the salinity data of fluid-inclusions to estimate the molarity of chlorine may be a quick solution for estimation of  $m_{Cl}^v$ , if the fluid-inclusions

trapped in minerals represents the primary magmatic aqueous phase in equilibrium with the granite magma. The salinity of primary fluid inclusions trapped in apatite enclosed in a plagioclase crystal (Fig. 1) from a granodiorite sample is a good example of the type of fluid inclusions suitably recommended to use to estimate chlorine molarity of magmatic fluid [13], as shown in column B of Table 1.



**Fig. (1).** Primary fluid inclusion trapped in an acicular apatite crystal enclosed in plagioclase grain from the Lake George granodiorite. This inclusion is comprised of liquid and gas phases, and has salinity of 15.0% NaCl<sub>equiv</sub>. Data from [13].

Webster and Holloway [7] showed that fluorine partitions strongly into a granitic melt relative to an aqueous fluid phase, and  $k_p^F$  increases with increasing fluorine in the melt with < 7 wt% fluorine. If a topaz rhyolite melt contains ≥ 7 wt% fluorine at 2 kb and 800°C, fluorine would partition in favour of the aqueous fluids.

- Calculate REE partition coefficient ( $k_p^{REE}$ ) between aqueous fluid phase and granite melt, according to the experimental results by [6, 9]; the values of  $k_p^{REE}$  are proportional to the cubic power of the chlorine molarity of the aqueous fluid, except for europium that is to the fifth power. The mathematic relationship of REE partition coefficients and the chlorine molarity of aqueous fluids can be obtained, based on a least square regression analysis of the experimental data by [6, 9] as follows.

The REE partition coefficients ( $k_p^{REE}$ ) data used for the least square regression analysis were acquired under the conditions of 4.0 kb, 800°C, and QFM (quartz-fayalite-magnetite) buffer by [6], and the data were obtained at 2.0 kb and 800°C in the experiments in [9]. Although the effect of temperature on the partition coefficients is not well defined in these experiments, the pressure effect is remarkable, showing that the partition coefficients of the trivalent REE increase with decreasing pressure. Therefore, the volatile phase exsolved from granite melts at

$$k_p^{La} = 0.0408(m_{Cl}^m)^3 - 0.0116 \quad (R^2 = 0.95, n = 13) \quad (3)$$

$$k_p^{Ce} = 0.0394(m_{Cl}^m)^3 + 0.0068 \quad (R^2 = 0.94, n = 18) \quad (4)$$

$$k_p^{Nd} = 0.0396(m_{Cl}^m)^3 - 0.0176 \quad (R^2 = 0.94, n = 13) \quad (5)$$

$$k_p^{Sm} = 0.0324(m_{Cl}^m)^3 - 0.0118 \quad (R^2 = 0.92, n = 13) \quad (6)$$

$$k_p^{Eu} = 0.0015(m_{Cl}^m)^5 + 0.0891 \quad (R^2 = 0.95, n = 18) \quad (7)$$

$$k_p^{Gd} = 0.0275(m_{Cl}^m)^3 + 0.0052 \quad (R^2 = 0.92, n = 20) \quad (8)$$

$$k_p^{Tb} = 0.0247(m_{Cl}^m)^3 - 0.0072 \quad (R^2 = 0.92, n = 13) \quad (9)$$

$$k_p^{Ho} = 0.0190(m_{Cl}^m)^3 - 0.0024 \quad (R^2 = 0.92, n = 13) \quad (10)$$

$$k_p^{Yb} = 0.02275(m_{Cl}^m)^3 + 0.0116 \quad (R^2 = 0.92, n = 18) \quad (11)$$

$$k_p^{Lu} = 0.0105(m_{Cl}^m)^3 + 0.0025 \quad (R^2 = 0.90, n = 13) \quad (12)$$

pressures below 4 kb may contain higher REE concentration than those calculated using the eqs. 3 to 12. The limitation of this type of modelling should be noted. Clearly, more experiments are required to cover a range of pressures at least from lower to upper crust.

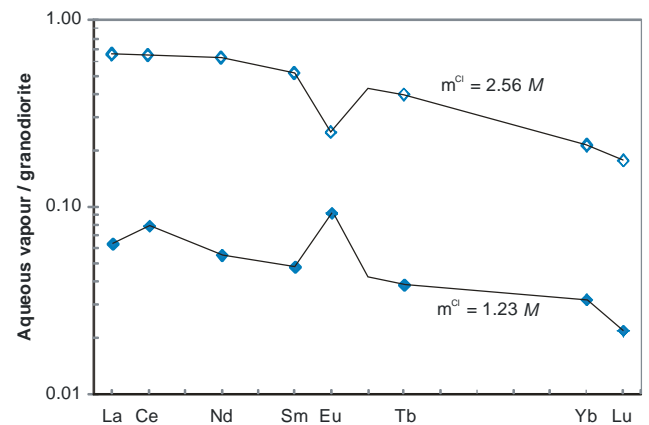
- Calculate the REE concentration of MVP on the basis of the  $k_p^{REE}$  values calculated described above. For example, Ce content in the MVP equals to  $k_p^{Ce} \times Ce$  content in the melt, given that Ce content in the sample (granite sample) equals to Ce content in the granite melt (Table 1).
- Plot the REE data of the MVPs in equilibrium with the granite melt as a commonly-used REE pattern; here, using REE values in the fresh granite as the normalization values rather than chondrite, in order to illustrate REE fractionation between the aqueous vapour phase and granite melt (Fig. 2).
- Compare the REE patterns of mineralized or altered rocks host to gold mineralization to those of the calculated aqueous vapour phase to trace the source of ore fluids (Fig. 3).

## APPLICATION

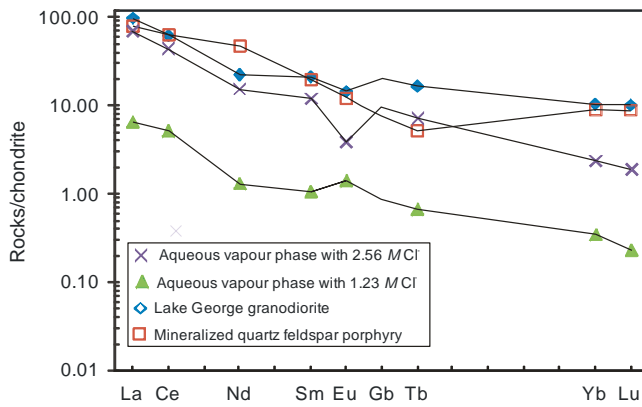
To illustrate the utility and applicability of the procedure as described above, it is applied to the Lake George Sb-Au-W-Mo mineral deposit, New Brunswick (Canada), which was the largest antimony producer in North America until mid-1990s. This deposit is temporally and spatially associated with the Early Devonian Lake George granodiorite stock [14-20]. The occurrence of Au-bearing quartz-carbonate veinlets and stockworks associated with W-Mo mineralization within the granodiorite stock, quartz-

feldspar dyke, and the proximal metamorphic aureole strongly suggests that associated Au mineralization is related to the hydrothermal system associated with this hypabyssal intrusion [15-17, 19], analogous to intrusion-related gold systems [13, 20-24].

Table 1 presents the procedures for calculating REE concentrations in magmatic volatile phases described above using the Lake George granodiorite as an example. Table 2 lists the REE concentrations in the Lake George granodiorite [17], mineralized quartz-feldspar porphyry [19], and magmatic volatile phases at chlorine molarity of  $m_{Cl}^v = 1.23$  and  $2.56 M$  in MVPs calculated in terms of the procedures described above, based on the granodiorite chlorite content and the salinity of primary fluid inclusions trapped in apatite enclosed in a plagioclase crystal fluid [13], respectively.



**Fig. (2).** Granodiorite-normalized REE patterns of magmatic fluids that have chlorine molarity of  $m_{Cl}^v = 1.23 M$  and  $2.56 M$  (see text for detailed calculation). The normalization values in Table 1.



**Fig. (3).** Chondrite-normalized REE patterns of the Lake George granodiorite (data from [17]) and mineralized quartz-feldspar porphyry (Data from [19]), comparing with the REE patterns of MVPs. The chondrite values from Boynton [28].

**Table 2. REE Concentrations (ppm)**

	1	2	3	4
La	31	25	1.97	20.89
Ce	51	52	4.07	34.36
Nd	13.7	29	0.76	8.90
Sm	4.2	3.9	0.20	2.24
Eu	1.1	0.9	0.10	0.28
Tb	0.8	0.25	0.03	0.33
Yb	2.2	1.9	0.07	0.48
Lu	0.3	0.29	0.01	0.06

\*Notes:

- 1 - The Lake George granodiorite (data from [17]).
- 2 - Mineralized quartz-feldspar porphyry (data from [19]).
- 3 - Magmatic vapour phase (mv-Cl = 1.23 M) in equilibrium with granite melt.
- 4 - Magmatic vapour phase (mv-Cl = 2.56 M) in equilibrium with granite melt.

Fig. (2) shows the calculated REE patterns in MVPs at  $m_{Cl}^v = 1.23$  and  $2.56 M$ , normalized by the Lake George granodiorite. With increasing value of  $m_{Cl}^v$ , REE concentrations in the respective MVPs increase significantly (Table 1). At  $m_{Cl}^v = 1.23 M$ , the REE pattern of MVP displays a positive Eu anomaly, whereas at  $m_{Cl}^v = 2.56 M$  the pattern show a pronounced negative anomaly.

When a comparison with the Lake George granodiorite and mineralized and altered quartz-feldspar porphyry is made, intriguing information stands out that these MVPs appear to decouple with the granodiorite that is roughly similar to the altered porphyry, except the latter lacks a Eu anomaly (Fig. 3). This may result from interaction of MPVs with the quartz-feldspar porphyry, leading to the disappearance of Eu anomaly and in some of the REE portion intersecting with that of the granodiorite. This also suggests that hydrothermal fluid(s) causing alteration as well as mineralization (sericite; see [19]), and gold mineralization are derived from the progressively cooling voluminous magma. This interpretation is supported by the prior studies

of fluid inclusion, mineral chemistry, stable isotopes, and litho geochemistry [13, 20, 22-24].

**DISCUSSION**

Although the experimental data on REE partitioning between MVPs and granite melt published are very limited and incomplete, implications for calculating REE patterns in MVPs used for tracing magmatic fluids are encouraging. This technique is relatively cheap compared to stable isotopes, and can be used as a tool to investigate the metallogeny of mineral deposits associated with granitoid intrusions.

A few problems are noted when using REE patterns of MVPs calculated in terms of partition coefficients to study of fluid source(s) forming a mineral deposit. First, eqs. 3 to 12 may be only applied to the conditions defined in (or similar to) the experiments [6,9], although the pressure effect may be inferred to some extent. Secondly, the experimental values of REE partition coefficient ( $k_p^{REE}$ ) are considered to be controlled only by the chlorine concentrations of aqueous fluids in these experiments. The other chemical components, such as  $CO_3^{2-}$ ,  $HS^-$ , have not been studied on the control of  $k_p^{REE}$  [6, 9]. Thirdly, it is hard to estimate the chlorine molarity ( $m_{Cl}^m$ ) of granite melt as pointed out by [10], as chlorine is readily lost. Using the salinity of primary fluid inclusions trapped in apatite enclosed in plagioclase as shown in the Lake George granodiorite (Fig. 1; [13]) may be a quick solution to this problem (i.e., estimate of chlorine molarity of magmatic fluid; see Step 2 above). Also, the methods proposed by [4] may offer an estimation of chlorine molarity ( $m_{Cl}^m$ ) of granite melt.

Another issue arises in tracking the source(s) of ore materials including metals (e.g., Au, Cu, S) and fluids ( $H_2O$ ). A use of the estimated abundance data of REE in MVPs presented in this contribution means that both REE and  $H_2O$  are derived from the cooling granite magma(s), but does not provide for evidence for the source of Au, Cu, and S. Sulphur isotopes in conjunction with fluid inclusions may better constrain sulphur source in the study of the Lake George deposit [13, 20]. It is known that magmatic pyrrhotite is present in the Lake George granodiorite, suggesting that most of the Au and Cu partitions into early sulphide liquid because their partition coefficients between sulphide and granite melt are very high. In such a case, MVPs must have low contents of Au and Cu, thus the potential of forming a gold deposit via simple melt-volatile partitioning could be low, as those metals were sequestered by earlier sulphide liquation processes. However, it is also well known that the primary magmatic sulphide minerals are not stable when interacting with MPVs releasing from progressively cooling, voluminous and volatile-saturated magma(s)/intrusion at depth. Resorption, oxidation, and destructions of these liquation sulphides could liberate Au, Cu, and S, and enhance the metalliferous endowment of the magmatic fluids [22, 25-27], which may then precipitate ore metals in a suitable setting, such as shear zone, hydrofracture system, to generate an intrusion-related gold deposit; the Lake George deposit is an example of such a process.

In conclusion, the similarities of the mineralized rocks and the calculated magmatic fluids in REE patterns suggest that the ore-forming fluids are derived dominantly from the granodiorite, providing evidence that origin of the Lake George Sb-Au-W-Mo mineral deposit is analogous to an intrusion-related gold system.

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#### CONFLICT OF INTEREST

Declared none.

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