

## **1. Incongruent Continuous Melting Calculations**

We follow the definition of McKenzie (1985) and Williams and Gill (1989), and refer to continuous melting as the integrated processes of melting and removal of magma from a static residue having a critical porosity. Continuous melting in this sense is similar to dynamic melting (commonly applied in the case of adiabatic melting of upwelling mantle), but with the difference that the solid residue in continuous melting is static (i.e., has no upwelling velocity). In the case of stable elements, dynamic melting and continuous melting are mathematically equivalent (Williams and Gill, 1989), and equations describing incongruent dynamic melting of stable elements have been presented previously (Zou and Reid, 2001). In this section we present the derivation of equations describing U-series nuclide behavior during incongruent, continuous melting. We first derive equations describing the behavior of stable elements (section 1.1), which are mathematically identical to those obtained by Zou and Reid (2001) although derived in a somewhat different manner. We then expand these equations to account for the effects of ingrowth and decay during the melting process (section 1.2). In this paper, we apply these equations to modeling of  $^{226}\text{Ra}$ - $^{230}\text{Th}$  and  $^{230}\text{Th}$ - $^{238}\text{U}$  disequilibria during incongruent melting of amphibolite, but the equations are general and can be applied to any parent-daughter pair and to any incongruent melting reaction.

### **1.1 Stable Element Calculation**

During continuous melting, initial melt fractions remain in equilibrium with the solid residue (batch melting) until a critical porosity is reached. Any additional melt produced beyond this critical porosity is instantaneously extracted and pooled elsewhere and the extracted melt fraction is referred to as  $X$ . As the critical mass porosity ( $\Phi$ ) approaches 0, the continuous melting process approaches incongruent fractional melting. We derive in this section equations describing the concentration of stable trace elements in the melt phase as a function of the extracted melt fraction and the critical porosity.

During incongruent melting some solid phases are consumed while others are produced along with the melt. Those which are consumed or produced in different proportions from their initial modal abundance are termed “incongruent” phases, whereas those which contribute to the melting reaction in their initial modal proportions are termed “congruent” phases. Hence the bulk partition coefficient of element  $i$  ( $D_i$ ) becomes a function of the melt fraction. Phase proportions are governed by an incongruent reaction which can be written as:



where  $\alpha_i$  are the incongruently melting phases,  $\theta_i$  are the congruently melting phases and  $\beta_i$  are the incongruently produced phases (Zou and Reid, 2001). For the case of amphibolite dehydration melting, only incongruently consumed and produced phases are present.

A general conservation equation for a stable element  $i$  (neglecting advective and diffusion terms) is given by:

$$(1 - \phi)\rho_s \frac{\partial c_s^i}{\partial t} + \rho_f \phi \frac{\partial c_f^i}{\partial t} = (c_s^i - c_f^i)M, \quad (2)$$

where  $\phi$  is the critical volume porosity,  $\rho_s$  is the solid residue density,  $\rho_f$  is the magma density,  $c_s^i$  is the concentration in the residue,  $c_f^i$  is the concentration in the magma and  $M$  is the melting rate, assumed to be constant. Note that the concentration of stable trace elements depends on the melt fraction but not on the timescale of melting and therefore is independent of the melting rate; we include it here to make the derivation parallel to that of the derivation of equations relevant to radioactive nuclides (section 1.2). We can replace time with extracted melt fraction as a variable using the following relationship between extracted melt fraction  $X$  and time (McKenzie, 1985):

$$X = 1 - \exp\left[\frac{-M_e}{\rho_f\phi + \rho_s(1-\phi)}t\right], \quad (3)$$

where  $M_e$  is the melt extraction rate. As pointed out by Zou (1998) the melt extraction rate is not strictly the melt production rate, but they are related as:

$$\frac{M}{M_e} = 1 - \Phi, \quad (4)$$

where  $\Phi$  is the critical mass porosity rather than the critical volume porosity and is related to the critical volume porosity by:

$$\Phi = \frac{\rho_f\phi}{\rho_f\phi + \rho_s(1-\phi)}. \quad (5)$$

As the stable element concentration is ultimately related to reaction progress rather than strictly to time, it is advantageous to write the differential equation in (2) in terms of extracted melt fraction rather than time. Using (3) we can perform a change of variables in (2), and using (4) and (5) and noting also that  $c_f^i = D^i c_s^i$  we get the following expression:

$$(1-\Phi)(1-X)\frac{\partial D^i c_f^i}{\partial X} + (1-X)\Phi\frac{\partial c_f^i}{\partial X} = (1-\Phi)c_f^i(D^i - 1). \quad (6)$$

During incongruent melting  $D^i$  is not constant and remains inside the differentiation operator. To perform this differentiation we first need to express  $D^i$  as a function of the melt progress through the extracted melt fraction variable. Both Zou (2001) and Hertogen and Gijbels (1975) give essentially the same expression for  $D^i$  in incongruent melting situations based upon mass balance arguments. Using the notation of Zou:

$$D^i = \frac{1}{1-F} [D_0^i - Q_0^i F], \quad (7)$$

where

$$D_0^i = \sum x_0^j K_i^j, \quad (8)$$

and

$$Q_0^i = \frac{K_i^j - \sum t^j K_i^j}{t^i}. \quad (9)$$

$F$  is the total melt fraction (which is related to the extracted melt fraction by

$F = \Phi + (1 - \Phi)X$ ),  $K_i^j$  is the partition coefficient of  $i$  in the  $j$ th phase (here the partition coefficient is assumed constant through the melting range),  $x_0^j$  is the fraction of the  $j$ th phase originally present, and  $t^j$  is the fraction of the  $j$ th solid incongruently produced phase and  $t^i$  is the fraction of melt produced.  $D_0^i$  is the initial bulk partition coefficient and  $Q_0^i$  is a constant related to the rate of change of the partition coefficient as a result of reaction progress as  $t^j$  phases are being produced. Note that (9) would have to be modified if congruently melting phases were also present (Zou and Reid, 2001). Substituting (9) in (7), and using the chain rule gives the simplified expression:

$$\frac{\partial c_f^i}{c_f^i} = \left( \frac{Q_0^i + \Phi(1 - Q_0^i) - 1}{D_0^i + \Phi(1 - Q_0^i) - X[Q_0^i + \Phi(1 - Q_0^i)]} \right) \partial X \quad (10)$$

Integrating and using the fact that  $c_{f0}^i = \frac{c_0^i}{D_0^i + \Phi(1 - Q_0^i)}$  (Zou and Reid, 2001) gives us

the following expression for  $c_{if}$  as a function of critical mass porosity and extracted melt fraction:

$$c_f^i = \frac{c_0^i}{D_0^i + \Phi(1-Q_0^i)} \left[ 1 - \frac{Q_0^i + \Phi(1-Q_0^i)}{D_0^i + \Phi(1-Q_0^i)} X \right]^{\frac{1}{Q_0^i + \Phi(1-Q_0^i)} - 1} \quad (11)$$

To get the concentration of element i in the aggregate melt this expression must be integrated over X and then divided by X to give:

$$\bar{c}_f^i = \frac{c_0^i}{X} \left( 1 - \left[ 1 - \frac{Q_0^i + \Phi(1-Q_0^i)}{D_0^i + \Phi(1-Q_0^i)} X \right]^{\frac{1}{Q_0^i + \Phi(1-Q_0^i)}} \right) \quad (12)$$

This is identical to the expression given in Zou and Reid (2001) eqn. 16 although derived in a somewhat different manner.

## 1.2 Radioactive Nuclides

The same general solution procedure is followed for the U-series nuclides, although in this case there is an added source term in the conservation equation that complicates the calculation. The half-life of <sup>238</sup>U is much longer than the melting timescale considered, therefore it can be treated as a stable element and equations (11) and (12) can be used to determine the concentration. However, for <sup>230</sup>Th and <sup>226</sup>Ra a new conservation equation must be used:

$$(1-\phi)\rho_s \frac{\partial c_s^D}{\partial t} + \phi\rho_f \frac{\partial c_f^D}{\partial t} = (c_s^D - c_f^D)M + \lambda^P [\rho_s c_s^P (1-\phi) + \rho_f c_f^P \phi] - \lambda^D [\rho_s c_s^D (1-\phi) + \rho_f c_f^D \phi] \quad (13)$$

Here the superscripts D and P refer to daughter and parent nuclide, respectively, and  $\lambda$  is the decay constant. The first term on the right-hand side accounts for transfer of daughter atoms from the solid to the liquid through melting (? is that right), the second term

accounts for ingrowth of the daughter due to decay of the parent during the melting process, and the third term accounts for decay of the daughter. Using (3) and changing the independent variable from time to extracted melt fraction gives:

$$(1-\phi)\rho_s \frac{M_e(1-X)}{\rho_f\phi + \rho_s(1-\phi)} \frac{\partial c_s^D}{\partial X} + \phi\rho_f \frac{M_e(1-X)}{\rho_f\phi + \rho_s(1-\phi)} \frac{\partial c_f^D}{\partial X} = (c_s^D - c_f^D)M + \lambda^p [\rho_s c_s^p (1-\phi) + \rho_f c_f^p \phi] - \lambda^D [\rho_s c_s^D (1-\phi) + \rho_f c_f^D \phi] \quad (14)$$

We can express this equation in terms of the concentration of daughter in the magma by

dividing (14) through by M, using  $\frac{M}{M_e} = \frac{\rho_s(1-\phi)}{\rho_s(1-\phi) + \rho_f\phi}$ , defining

$\rho_{avg} = \rho_s(1-\phi) + \rho_f\phi$ , and using the partition coefficient expression

$$(D^D = D_0^D + \Phi(1 - Q_0^D) - X[Q_0^D + \Phi(1 - Q_0^D)]):$$

$$\begin{aligned} & \frac{\partial c_f^D}{\partial X} \left[ \frac{D_0^D + \Phi(1 - Q_0^D) - X[Q_0^D + \Phi(1 - Q_0^D)]}{(1 - \Phi)\rho_{avg}} \right] + \\ & c_f^D \left[ \frac{D_0^D - Q_0^D}{(1 - \Phi)(1 - X)\rho_{avg}} - \frac{D_0^D + \Phi(1 - Q_0^D) - X[Q_0^D + \Phi(1 - Q_0^D)] - (1 - X)}{(1 - \Phi)(1 - X)\rho_{avg}} + \frac{\lambda^D}{M} \left( \frac{D_0^D + \Phi(1 - Q_0^D) - X[Q_0^D + \Phi(1 - Q_0^D)]}{(1 - X)} \right) \right] = \\ & c_f^p \left[ \frac{\lambda^p}{M} \left( \frac{D_0^p + \Phi(1 - Q_0^p) - X[Q_0^p + \Phi(1 - Q_0^p)]}{1 - X} \right) \right] \end{aligned} \quad (15)$$

To simplify the expression, we define  $A = D_0 + \Phi(1 - Q_0)$  and  $B = Q_0 + \Phi(1 - Q_0)$ :

$$\begin{aligned} & \frac{\partial c_f^D}{\partial X} \left[ \frac{A^D - X[B^D]}{(1 - \Phi)\rho_{avg}} \right] + \\ & c_f^D \left[ \frac{D_0^D - Q_0^D}{(1 - \Phi)(1 - X)\rho_{avg}} - \frac{A^D - X[B^D] - (1 - X)}{(1 - \Phi)(1 - X)\rho_{avg}} + \frac{\lambda^D}{M} \left( \frac{A^D - X[B^D]}{(1 - X)} \right) \right] = \\ & c_f^p \left[ \frac{\lambda^p}{M} \left( \frac{A^p - X[B^p]}{1 - X} \right) \right] \end{aligned} \quad (16)$$

This equation cannot be solved by separation of variables but can be written in the form,

$$c_f^{D'} + P c_f^D = L \quad (17)$$

where P and L are constants. The solution is:

$$c_f^D = e^{-I} (T + c_0^D A^D \frac{1}{B^D}), \quad (18)$$

where

$$T = \int L e^I dX, \quad (19)$$

and

$$I = \int P dX \quad (20)$$

P and L can be simplified to:

$$P = \frac{(1 - B^D)}{(A^D - B^D X)} + \frac{\lambda^D (1 - \Phi) \rho_{avg}}{M(1 - X)}, \quad (21)$$

and

$$L = \frac{c_f^p \lambda^p \rho_{avg} (A^p - B^p X)(1 - \Phi)}{M(1 - X)(A^D - B^D X)}. \quad (22)$$

So,

$$e^I = (A^D - B^D X)^{\frac{B^D - 1}{B^D}} (1 - X)^{\frac{-\lambda^D (1 - \Phi) \rho_{avg}}{M}} \quad (23)$$

and,

$$T = \frac{\lambda^p \rho_{avg} (1 - \Phi)}{M} \int c_f^p (A^p - B^p X) (A^D - B^D X)^{\frac{1}{B^D}} (1 - X)^{\frac{-\lambda^D (1 - \Phi) \rho_{avg} - 1}{M}} \partial X . \quad (24)$$

$c_f^p$  is also a function of X. The abundance of  $^{238}\text{U}$  can be computed with (18), but since there is no parent nuclide  $T=0$ , the solution simplifies to that obtained by separation of variables. The abundance of  $^{238}\text{U}$  can then be used to compute the abundance of  $^{230}\text{Th}$  using (18). The integral in T is difficult to solve analytically because in general  $A^D \neq A^p \neq B^D \neq B^p$ , and in this study T was evaluated numerically using the trapezoidal method. With this strongly non-linear function care must be taken that the integration steps are sufficiently small so that errors are small. This requires iterative refinement until activities ratios changed by less than 0.01. . Once  $c_f^{230Th}$  is determined as a function of X we can use it as the parent in (18) to determine  $c_f^{226Ra}$ . Equation (18) gives the concentration of the daughter nuclide in the melt at any point in the melting process (the instantaneous melt), but the concentration in the aggregated extracted melt is more relevant to this work. This is again calculated using:

$$\bar{c}_f^i = \frac{1}{X} \int c_f^i \partial X . \quad (26)$$

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