

SUPPLEMENTARY MATERIALS

Materials and Methods

1. Temperature Decay

We used an exponential time dependent heat decay equation of the form

$$T = T_o \exp(-bt), \quad \text{S1}$$

that represent the cooling of a magma body. Here, T = temperature (K), T_o = initial temperature (K), b = proportionality constant (for rapid cooling $b = 0.0000007 \text{ s}^{-1}$; for slow cooling $b = 0.00000035 \text{ s}^{-1}$; and for very slow cooling $b = 3.4926 \times 10^{-9} \text{ s}^{-1}$) and t = time (s).

2. Diffusion

The $CO_{2,t}$ diffusivity used in this study is expressed in the form of

$$\ln D_{CO_{2,t}} = \left(a_o + \frac{b_o}{T} + c_o \frac{P}{T} \right) + \left(a_1 + \frac{b_1}{T} \right) c_w. \quad \text{S2}$$

Equation (2) is adapted from Zhang et al. (2007) and it correlates the effect of pressure, temperature and H_2O wt% on the $CO_{2,t}$ diffusivity in silicate melt. For the anhydrous silicate melt ($\leq 0.2 \text{ wt\% } H_2O_i$) this equation is modified in the form

$$\ln D_{CO_{2,t}} = \left(-14.69 - \frac{16915}{T} + 0.2056 \frac{P}{T} \right), \quad \text{S3}$$

and for the hydrous silicate melt the equation is

$$\ln D_{CO_{2,t}} = \left(-14.34 - \frac{17360}{T} + 0.6527 \frac{P}{T} \right) + \left(-0.7172 + \frac{1436.8}{T} \right) c_w \quad \text{S4}$$

where, $D_{CO_{2,t}}$ = $CO_{2,t}$ diffusion coefficient (m^2s^{-1}), and P = pressure (MPa).

For quantitatively estimating the diffusive velocity of the $CO_{2,t}$ front the continuity equation (Fick's law) of the form

$$\frac{\partial C}{\partial t} = D \frac{\partial^2 C}{\partial x^2} \quad S5$$

is used. Where, $C = CO_{2,t}$ concentration (moles/L) and x = length (m).

3. Advection

For the metasomatic scenario, the diffusivity coefficient is adapted in the modified form of the Stokes – Einstein equation from Tamimi et al. (1994)

$$D_{CO_{2,t}} = \frac{5.35 \times 10^{-10} T}{\left(\frac{\mu}{10^{-3}} \right)^{1.035}}. \quad S6$$

Where, μ = temperature dependent dynamic viscosity (Pa.s) and is related with the temperature in the form of

$$\mu = A \times T^{\frac{B}{(T-C)}}, \quad S7$$

where, $A = 2.414 \times 10^{-5}$ Pa.s; $B = 247.8$ K and $C = 140$ K.

$D_{CO_{2,t}}$ front migration due to advection – diffusion in the porous media is estimated using the continuity equation in the form of

$$\frac{\partial C}{\partial t} = D\phi \frac{\partial^2 C}{\partial x^2} - v\phi \frac{\partial C}{\partial x}. \quad S8$$

Where, ϕ = porosity and v = flow velocity (ms^{-1}). For the metamorphic scenario the rock porosity (ϕ) and advective velocity (v) is assumed constant throughout the simulation time.

4. Initial and Boundary Conditions

For simultaneously solving the system of equations following initial and boundary conditions were used. For the initial condition we used a constant concentration of $CO_{2,t}$

$$C(x, t = 0), C_{ic}. \quad S9$$

On the other hand the boundary conditions that we used can be expressed as

$$\left. \begin{aligned} C(x=0, t) &\rightarrow C_{bc} \\ C(x=\infty, t) &\rightarrow \frac{\partial C}{\partial x} = 0 \end{aligned} \right\}. \quad \text{S10}$$

The boundary conditions imposed indicate a constant CO₂ concentration at the entry point ($x=0$) and no flux boundary condition at the exit point ($x=\infty$). For the silicate melt we also used flux boundary condition as

$$\left. \begin{aligned} C(x=0, t) &\rightarrow \frac{\partial C}{\partial x} = -\frac{J}{D} \\ C(x=\infty, t) &\rightarrow \frac{\partial C}{\partial x} = 0 \end{aligned} \right\}. \quad \text{S11}$$

Here, at the entry point ($x=0$) diffusive flux and at the exit point ($x=\infty$) no flux boundary condition is imposed.

5. *Parameter Choices*

The initial CO₂ concentration (C_{ic}) is chosen as 10^{-5} moles/L and the boundary CO₂ concentration (C_{bc}) is fixed at 0.0114 moles/L (or 500 ppm assuming a CO₂ mass of 43.98 gm/moles). The boundary concentration (C_{bc}) is an intermediate value taken from CO₂ melt inclusion data (range 167–1596 ppm) of ultra – slow Gakkel spreading ridge (Shaw et al., 2010). Two different silicate melt temperature decay scenarios are considered in this study. (a) Rapid cooling where the silicate melt cools from 1200°C to 460°C at a rate of 7.4×10^{-40} C/year and (b) slow cooling where the silicate melt cools from 1200°C to 460°C at a rate of 3.7×10^{-50} C/year respectively. The cooling rates chosen in this study are within the same order of magnitude of the estimation of Spera (1980). He estimated that a granitic pluton with a radius of 10 km will cool down asymptotically from 1030°C to 700°C within 10^6 years i.e. the rate of cooling is 3.3×10^{-40} C/year.

6. *Calculation Strategy*

The system of equations using the initial and boundary conditions were solved using one dimensional finite difference method for each set of parameters. For the first time step, (1) calculate the temperature, (2) calculate the diffusivity of CO₂ using obtained temperature; (3) solve the diffusive and/or diffusive – advective equation for CO₂ transport. Following this, we calculated temperature, temperature dependent diffusivity and CO₂ concentrations at every point in the system, for the first time step. These values become the initial values for the next time step at all points of the profile, and so on.

References

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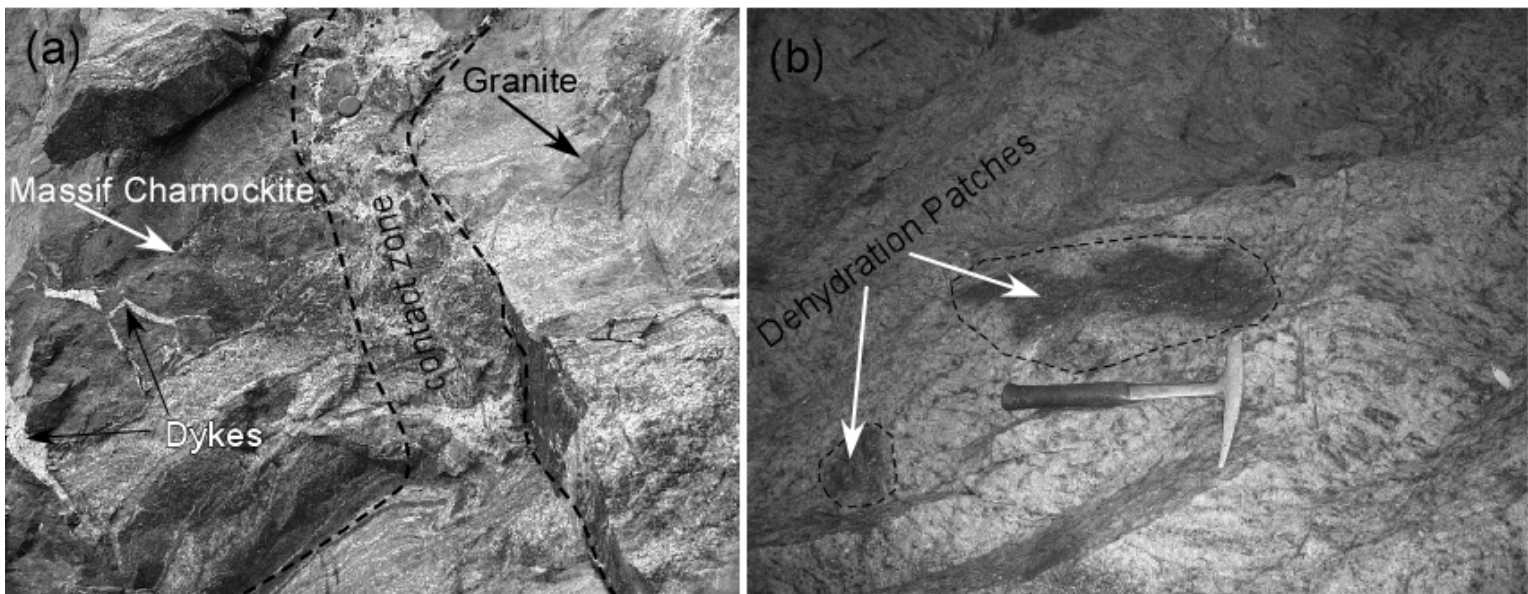


Figure DR1. (a) Massif charnockite pluton showing parallel contact with granite and cross cut by dykes of different compositions. (b) Meter to centimeter scale dehydration patches (incipient charnockite) within a granite enclave. The locations are from Central Madurai Block.

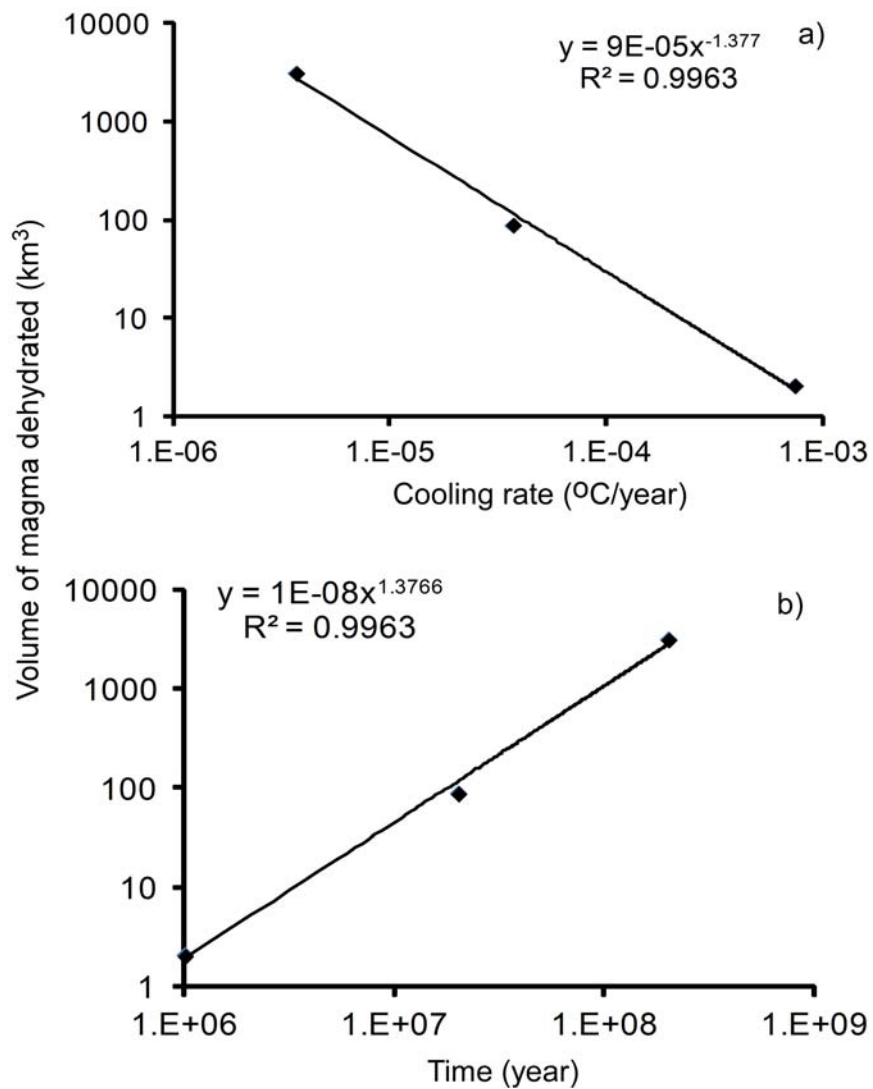


Figure DR2. Log – log plot of the parameters (a) the calculated cooling rate versus calculated dehydrated melt volume, and (b) calculated CO₂ travel time versus calculated dehydrated melt volume showing very high correlation amongst the parameters.