

Melting, Compaction and Reactive Flow: Controls on Melt Fraction and Composition Change in Crustal Mush Reservoirs

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Abstract

Changes in melt fraction and local bulk composition in high-crystallinity, crustal mush reservoirs are essential to produce the large volumes of low-crystallinity, silicic magma that are emplaced to form plutons, or erupted to surface. Heating (and cooling) is well understood and widely invoked in driving melt fraction change, but does not cause chemical differentiation because there is no separation of melt and crystals. Fractional crystallisation at high melt fraction is widely assumed to explain differentiation, but is inconsistent with the evidence that large-scale, long-term magma storage and evolution occurs in high-crystallinity mush reservoirs. Compaction has been suggested to explain melt fraction change and differentiation at low melt fraction, but compaction (and decompaction) causes simple unmixing (and mixing) of melt and solid crystals: to produce very refractory bulk composition by compaction, melt fraction must be driven down to very low values. Yet microstructural evidence demonstrating widespread compaction in crustal mush reservoirs at low melt fraction is lacking. Here we show that melt fraction change can be expressed in terms of heating/cooling and compaction, plus an additional term that we call 'reactive flow'. Similarly, composition change can be expressed in terms of compaction and reactive flow. Reactive flow changes the local bulk composition, which causes 'chemical' melting (dissolution) and freezing (precipitation), distinct from 'thermal' melting/freezing caused by changes in enthalpy. We use numerical modelling to show that the contributions of compaction and reactive flow in a crustal magma reservoir are similar in magnitude. However, reactive flow opposes melt fraction and composition changes caused by compaction when compaction occurs in a temperature gradient that increases upwards at, for example, the base of a sill intrusion, or decompaction occurs in a temperature gradient that decreases upwards at, for example, the top of a sill intrusion. If compaction causes melt fraction decrease and creates a more refractory bulk composition, then reactive flow causes melt fraction increase and a more evolved bulk composition, and vice versa. Reactive flow means that very small melt fraction is not required to produce very refractory composition in a crustal magma reservoir, consistent with the relatively scarce microstructural evidence for widespread compaction. The apparent lack of compaction in crustal magma reservoirs, as compared to other natural and engineered systems in which reaction does not occur, is also explained by the contribution of reactive flow. Reactive flow also means that melt loss in compacting regions of a crustal magma reservoir may instead be accompanied by evidence for mineral dissolution, which facilitates ongoing melt fraction loss by preserving connected melt flow paths through the mush pore space. Reactive flow in decompacting regions can also explain why interstitial mineral phases display textures that mimic those of interstitial melt.

Keywords: mush reservoir, compaction, reactive flow, differentiation, microstructure

Introduction

It is now widely, albeit not universally, accepted that long-term magma storage in the continental crust occurs in low melt fraction (high-crystallinity) 'mush reservoirs' rather than the high melt fraction (low-crystallinity) 'magma chambers' that have dominated conceptual models of magma storage and differentiation for over a century (e.g. [Bachmann and Bergantz, 2004](#); [Bachmann and Huber, 2016](#); [Bowen, 1915, 1919](#); [Cashman et al., 2017](#); [DePaolo, 1981](#); [Edmonds et al., 2019](#); [O'Hara, 1977](#)). However, recognition that mush reservoir processes play an important role in controlling magma storage and differentiation has generated a host of new questions around what those processes are, how they operate and what evidence they may

leave in the rock record. Chief among these is how high-crystallinity mush reservoirs produce chemically evolved, low-crystallinity magmas, which can leave the reservoir and ascend through the crust to be emplaced at shallower level or erupted at the surface. To understand and explain the formation of these magmas, it is necessary to understand the processes that cause, and control, changes in melt fraction and composition in crustal mush reservoirs.

Melt fraction change by heating or cooling is well understood and widely invoked, with numerous modelling studies exploring its impact in crustal magma reservoirs (e.g. [Annen et al., 2005](#); [Dufek and Bergantz, 2005](#); [Hodge, 1974](#); [Karakas et al., 2017](#); [Petford and Gallagher, 2001](#)). Yet heating and cooling alone cannot yield differentiation, because there is no physical separation of

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melt and solid crystals: the bulk composition remains constant. Recognizing this, fractional crystallisation is typically suggested to explain the formation of evolved magmas, with crystals assumed to separate rapidly from melt (e.g. Bowen, 1915, 1919; DePaolo, 1981; Keller *et al.*, 2015; O'Hara, 1977). At high melt fraction, relative flow of melt and crystals can occur by a variety of mechanisms such as crystal settling or convective fractionation (e.g. Bachmann and Bergantz, 2004). Yet differentiation in high melt fraction magma chambers is inconsistent with the evidence that large-scale, long-term magma storage and evolution typically occurs at low melt fraction (e.g. Bachmann and Huber, 2016; Cashman *et al.*, 2017; Edmonds *et al.*, 2019).

At low melt fraction, relative flow of melt and crystal matrix leading to melt fraction change and compositional evolution is widely assumed to occur by compaction and similar mechanisms such as filter pressing and gas sparging (e.g. Anderson *et al.*, 1984; Bachmann and Bergantz, 2004; McKenzie, 1984, 1985; Sisson and Bacon, 1999). Yet Holness (2018) has pointed out that microstructural evidence for crystal deformation consistent with compaction at low melt fraction is scarce in igneous rocks, at least those formed in relatively shallow magmatic systems. Consequently, it is not straightforward to explain how melt fraction and composition change in crustal mush reservoirs: heating and cooling alone do not yield composition change, while compaction yields both, but is not obviously consistent with the rock record.

Here we begin by exploring the mechanisms that change melt fraction and composition using a simple and widely recognized binary phase diagram (Fig. 1). We demonstrate the changes in melt fraction and composition that occur during the well-known processes of heating, cooling and compaction; we also demonstrate that melt fraction can change in response to the addition or removal of components (i.e. a change in local bulk composition) at fixed enthalpy. Consistent with later sections of the paper, we call this latter process 'reactive flow'. The reactive, percolative flow of melt through crystal mush has been proposed to explain observed geochemical features in magmatic systems in the continental and oceanic crust (e.g. Camejo-Harry *et al.*, 2018; Jackson *et al.*, 2018; Keller and Katz, 2016b; Keller and Suckale, 2019; Leuthold *et al.*, 2014; Lissenberg and MacLeod, 2016), building on numerous earlier studies of the mantle (e.g. Collier and Kelemen, 2010; Kelemen, 1990; Kelemen *et al.*, 1990; Kelemen *et al.*, 1995; Reiners, 1998; Rudge *et al.*, 2011; Spiegelman and Kelemen, 2003; Spiegelman *et al.*, 2001). However, its specific contribution to melt fraction and composition change in crustal mush reservoirs has not been quantified.

We then use the well-known mass conservation (continuity) statements for phases (solid and melt) and components (corresponding to a binary phase diagram) to derive mathematical descriptions for melt fraction and composition change. The general case for multiple phases and components is presented in A.1. We show that melt fraction change in our binary phase diagram can be expressed in terms of heating/cooling and compaction, consistent with the examples demonstrated using Fig. 1, plus an additional term that does not correspond to either of these processes. This additional term shows that melt fraction changes when melt and solid flow through gradients in composition; the term describes reactive flow and we provide here a clear, general mathematical definition. Similarly, we show that composition change can be expressed in terms of compaction and reactive flow; heating/cooling does not contribute.

We finish by quantifying the relative contributions of these different mechanisms to melt fraction and composition change in a crustal mush reservoir using numerical modelling and a number

of example cases. We investigate whether melt fraction change is dominated by heating/cooling, compaction or reactive flow, and why there may be a lack of evidence for compaction in the rock record.

The terms melt fraction, local composition (bulk, melt or solid) and local thermodynamic equilibrium are used throughout the paper. By 'local', we mean at the scale of a 'representative elementary volume' (REV) (e.g. Bear, 1972). The REV concept is commonly invoked in porous systems: it is small enough to be considered mathematically as a point, but large enough to contain a representative number of solid crystals and melt-filled pores. The REV here could be considered to correspond to the scale of a thin section, hand specimen or rock sample in a melting experiment. Hence, when we use the term 'local bulk composition', we mean the bulk composition that would be determined experimentally at the thin section to hand-specimen scale. Similarly, local melt or solid composition means the melt or solid composition that would be determined at this scale by, for example, microbeam analysis. Melt fraction and other properties such as temperature and enthalpy are defined at the same scale. Local thermodynamic equilibrium is assumed at this scale, such that the melt and solid phases have the same temperature, and component exchange has occurred to yield equilibrium solid and melt compositions. The assumption of local thermodynamic equilibrium allows the use of equilibrium phase diagrams to relate temperature or enthalpy to melt fraction, local bulk composition and local melt and solid compositions. We consider the consequences of relaxing this assumption in the Discussion section.

Melt fraction and composition changes in crustal magma reservoirs: insights from a simple binary phase diagram

Although melting and crystallisation of rocks, and segregation of melt from solid crystals, are familiar problems in igneous petrology, it is informative to represent these processes on a simple phase diagram. Here we consider the well-known, experimentally derived, binary phase diagram diopside–anorthite, shown in Fig. 1 in its conventional temperature-composition (T-C) format (Fig. 1a), as well as in an enthalpy-composition (H-C) format (Fig. 1b). As noted by Ussler and Glazner (1992), the (H-C) format is especially useful when discussing heat, which, unlike temperature, is a conserved quantity. We use the phase diagram in conceptual experiments to demonstrate the three processes considered here that impact melt fraction and local bulk composition in crustal mush reservoirs: heating/cooling, compaction and reactive flow.

Melt fraction change by heating/cooling Heating/cooling in the anorthite–diopside binary system

The most commonly invoked cause of melt fraction change in magmatic systems is simple heating or cooling, which corresponds to the gain or loss of enthalpy but is more typically expressed in terms of an increase or decrease in temperature (e.g. Annen *et al.*, 2005; Dufek and Bergantz, 2005; Hodge, 1974; Karakas *et al.*, 2017; Petford and Gallagher, 2001) (Fig. 1). Heat is required both to raise the temperature of the system (sensible heat) and to cause solid to liquid phase change (latent heat). The consequences of adding or removing heat can be easily demonstrated using our simple phase binary phase diagram (Fig. 1): consider 100 g of rock (an REV) described by the phase diagram, shown as case A in Fig. 1, with bulk composition An₂₀, initially at room temperature

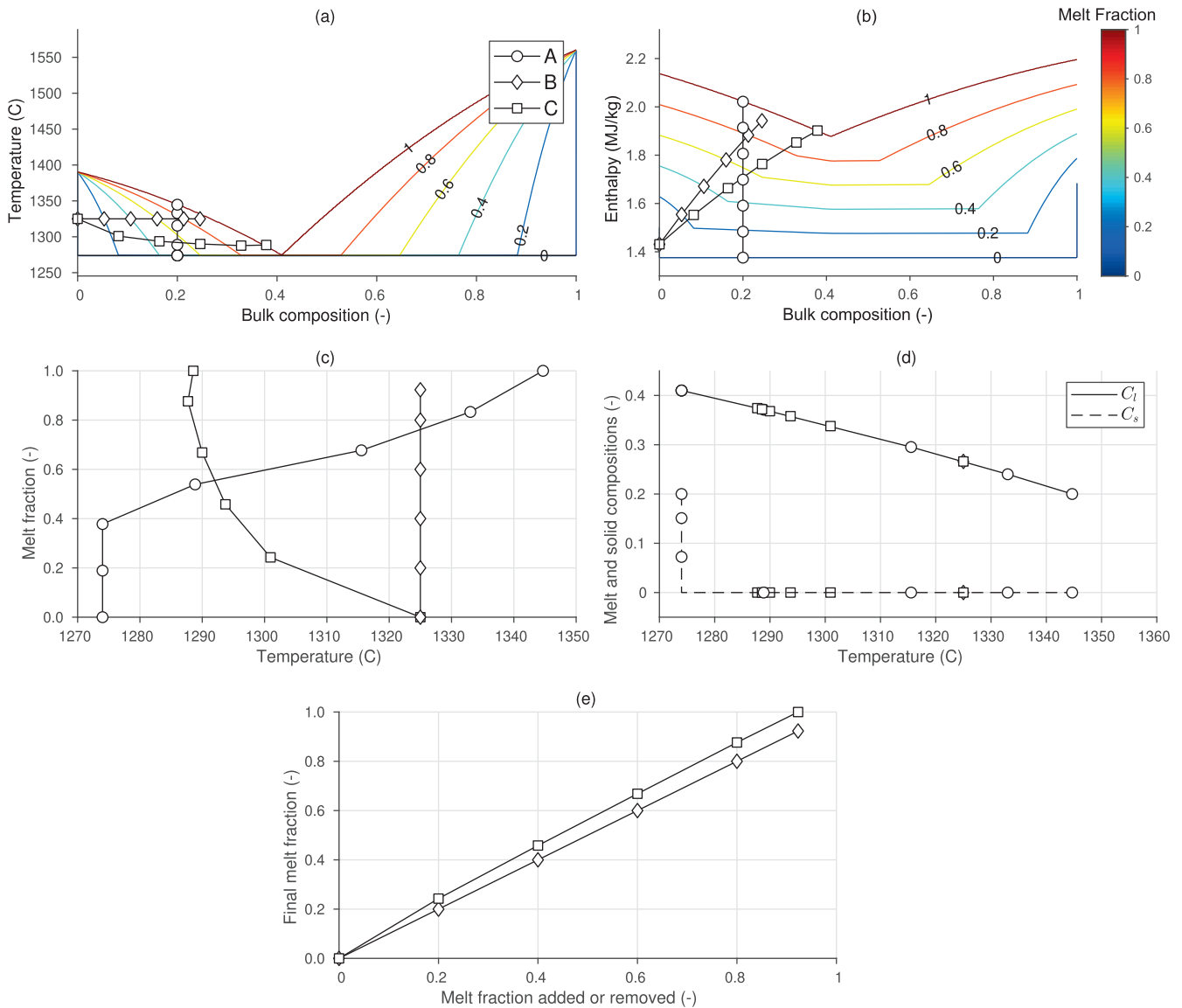


Fig. 1. Diopside–anorthite phase diagram generated using data from [Ussler and Glazner \(1992\)](#), expressing (a) temperature as a function of bulk composition; (b) enthalpy as a function of bulk composition; (c) melt fraction as a function of temperature, (d) melt and solid composition as a function of temperature and (e) final melt fraction as a function of melt fraction added to, or lost from, a conceptual experimental rock sample. Case A corresponds to pure heating/cooling; case B corresponds to pure compaction, and case C corresponds to reactive flow. Data points represent results from the conceptual experiments described in the text. Colours in (a) and (b) denote melt fraction contours. Composition $\bar{C} = 0$ corresponds to pure diopside (An0); composition $\bar{C} = 1$ corresponds to pure anorthite (An100). See Appendix B for details of the mathematical description of the phase diagram.

(20°C). A composition of An20 corresponds to 20% anorthite and 80% diopside; we equivalently express this composition as $\bar{C} = 0.2$. We place this sample in an experimental melting apparatus, and add 135.4 kJ of energy to bring the sample to temperature $T = 1274^\circ\text{C}$ (Figure 1a,b). We quench and measure the melt (glass) fraction and composition, identifying incipient (very low fraction) melt with composition An42 ($C_l = 0.42$); in this experiment, we have just reached the solidus enthalpy.

We repeat the experiment, this time adding 146.2 kJ of energy. The sample reaches the same temperature, but the melt fraction is now $\phi = 0.189$. We repeat the experiment again, adding more energy in each cycle, plotting the results on the phase diagram (Fig. 1a,b) and producing a plot of melt fraction against temperature (or enthalpy) at constant bulk composition (Fig. 1c). The fixed bulk composition means that these heating/cooling

data plot as a vertical line on Fig. 1a,b. Melt fraction change in response to heating occurs at constant temperature and melt composition, until solid anorthite crystals are exhausted; further heating then causes dissolution of diopside crystals into the melt (Fig. 1c). Melting at constant temperature demonstrates that the appropriate (conserved) quantity to describe melting is enthalpy, not temperature (e.g. Fig. 1b), as pointed out previously ([Ussler and Glazner, 1992](#)). However, temperature is more easily measured and controlled in melting experiments. The equilibrium melting process described here is reversible, such that cooling and crystallisation are simply the opposite of heating and melting. This simple thought experiment has been implemented practically in numerous studies to determine melt fraction and composition against temperature for given starting bulk rock compositions.

Heating/cooling in crustal magma reservoirs

Cooling is an inevitable cause of melt fraction reduction in all igneous rocks, which were once (partially) molten. Conversely, heating has been proposed as a mechanism for melt fraction increase in crustal magma reservoirs, in a process termed ‘thermal rejuvenation’ or ‘defrosting’ (Burgisser and Bergantz, 2011; Cooper and Kent, 2014; Szymanowski *et al.*, 2017). In this mechanism, a magma reservoir that is at low melt fraction because it is ‘cool’ or ‘cold’ is heated by intrusion of new, hot magma, causing the melt fraction in the surrounding mush to increase.

Thermal rejuvenation is widely suggested as a mechanism to create low-crystallinity magmas in mush reservoirs, often over short timescales (e.g. Bachmann and Huber, 2016; Burgisser and Bergantz, 2011; Cooper and Kent, 2014; Sliwinski *et al.*, 2017; Sparks *et al.*, 2019). However, although thermal rejuvenation may occur in some cases where hot mafic magma intrudes into cooler felsic magma (e.g. Blundy and Sparks, 1992; Wiebe, 1980) no differentiation occurs unless the melt is removed from the solid crystal residue unless the thermally rejuvenated melt is extracted from the solid residue. Differentiation (i.e. a change in local bulk composition) requires relative motion to separate melt and solid crystals (i.e. non-zero melt and solid velocities).

Melt fraction and composition change by compaction

Compaction in the anorthite–diopside binary system

Compaction is another commonly suggested cause of melt fraction change in low melt fraction magma reservoirs (e.g. Bachmann and Bergantz, 2004; Jackson *et al.*, 2018; McKenzie, 1984, 1985; Solano *et al.*, 2012) and is usually assumed to occur in response to the upwards flow of buoyant melt and the resulting deformation of the crystal matrix. Compaction occurs in many natural and engineered systems, and is generally defined as a net loss of porosity (which we take to be synonymous with melt fraction) without chemical component exchange (interphase mass transfer) from solid to liquid: examples include soils saturated with air or water (e.g. Hamza and Anderson, 2005), hydrocarbon reservoirs saturated with gas and/or oil and water (e.g. Nagel, 2001) and industrial filter presses (e.g. Tiller and Kwon, 1998). Decompression is defined by a net gain of porosity (or melt fraction) without interphase mass transfer. Compaction results from the relative motion of fluid and solid.

In the context of our simple phase diagram, compaction can be visualised in terms of a simple unmixing of a melt from its equilibrium solid residue; note we use mixing here to mean mixing of melt and solid phases, not mixing at the molecular level. Consider an experimental melting apparatus in which melt can be squeezed out of a partially molten sample by operating a mechanical press (e.g. Philpotts and Dickson, 2000). The heating system keeps the temperature constant. We place again 100 g of rock (an REV) described by our binary phase diagram in the apparatus, shown as case B in Fig. 1. The sample has initial bulk composition An_{24.5} ($\bar{C}$ = 0.245) and is initially at room temperature (20°C). We add 192.1 kJ of energy to bring the sample to a new temperature of 1325°C. We quench the sample and measure the melt fraction, solid and melt composition. We record a melt fraction of $\phi = 0.923$, and determine that the solid has pure diopside composition An₀ ($C_s = 0$) and the melt has composition An₂₇ ($C_l = 0.266$). We repeat the experiment, but now use the mechanical press to expel a mass of melt equivalent to a melt fraction of 0.12 at constant temperature before quenching and measuring again the melt fraction, and melt and solid compositions, from which we calculate the new

bulk composition of the sample. The melt and solid compositions remain unchanged from the previous experiment, but the melt fraction has dropped to $\phi = 0.8$ and the bulk composition has changed to An_{21.3} ($\bar{C} = 0.213$). We repeat the experiment again, expelling further melt in each cycle and plotting the results on the phase diagram (Fig. 1) until we are left with a very small melt fraction in the sample.

There is no change in melt or solid composition during pure compaction at constant temperature; hence, the compaction data plot as a horizontal line on Fig. 1a, and a vertical line on Fig. 1c. However, melt has a higher heat content than solid at the same temperature (the difference depends on composition, varying between 0.5 and 0.635 MJ/kg at 1325°C for the range of compositions on the left-hand side of the eutectic), so compaction redistributes heat. If we were able to measure the heat gained or lost in each experiment, we would obtain a linear relationship between melt fraction and heat content that reflects unmixing of the melt and solid as melt is expelled from the sample (Fig. 1b). The final melt fraction in the sample after each experiment simply reflects the melt lost by compaction (Fig. 1d); as we confirm later, changes in melt fraction due to compaction lie on a simple melt–solid mixing line. The gradient of the mixing line depends on the temperature of the experiment, because temperature controls melt composition. In terms of temperature, enthalpy and composition, the compaction process described here is reversible, such that decompression is simply the opposite of compaction and we could, in principle, implement a similar experiment that increased rather than decreased melt fraction to cover the full melt fraction range. However, the micro-scale distribution of melt, and the grain-scale mechanisms allowing melt fraction change within an REV, would be different during compaction and decompression.

Compaction in magma reservoirs

Compaction, as generally defined here, can occur in magma reservoirs through a variety of mechanisms. In a cooling and crystallising magma at high melt fraction (Fig. 2a), compaction occurs when crystals sediment onto a lower boundary. Decompression can likewise occur if upwards migrating melt percolating through the sedimented crystals causes the bed to become fluidised (e.g. Bergantz *et al.*, 2015; Schleicher *et al.*, 2016).

At moderate melt fraction (Fig. 2b), the sedimented crystals form an interconnected framework; compaction now requires deformation of the solid matrix, which can occur by rearrangement of the crystals without any change in crystal morphology in a process termed ‘mechanical compaction’ (Fig. 2b; Holness, 2018; Revil *et al.*, 2002). Flow of melt relative to the solid matrix can be driven by the buoyancy of the melt, or by other processes such as gas sparging or shear dilation; if there is a net loss (or gain) of porosity as a result of flow, then compaction (or decompression) has occurred, irrespective of the force(s) driving flow. In industrial filter presses, the driving force is supplied by pumps; in a coffee press, the driving force is provided by the arm of the thirsty drinker. In both cases, the solid residue compacts as the liquid is squeezed out.

At low melt fraction (Fig. 2c), compaction can only proceed if the crystal morphology changes, in a process sometimes termed ‘viscous compaction’. Crystal deformation during viscous compaction in mush reservoirs is typically assumed to occur via creep mechanisms (e.g. Holness, 2018; Rosenberg, 2001). At higher strain rates and/or lower temperatures, deformation occurs by dislocation creep, whereas at higher temperatures and/or lower strain rates, deformation occurs by diffusion creep (Cooper and

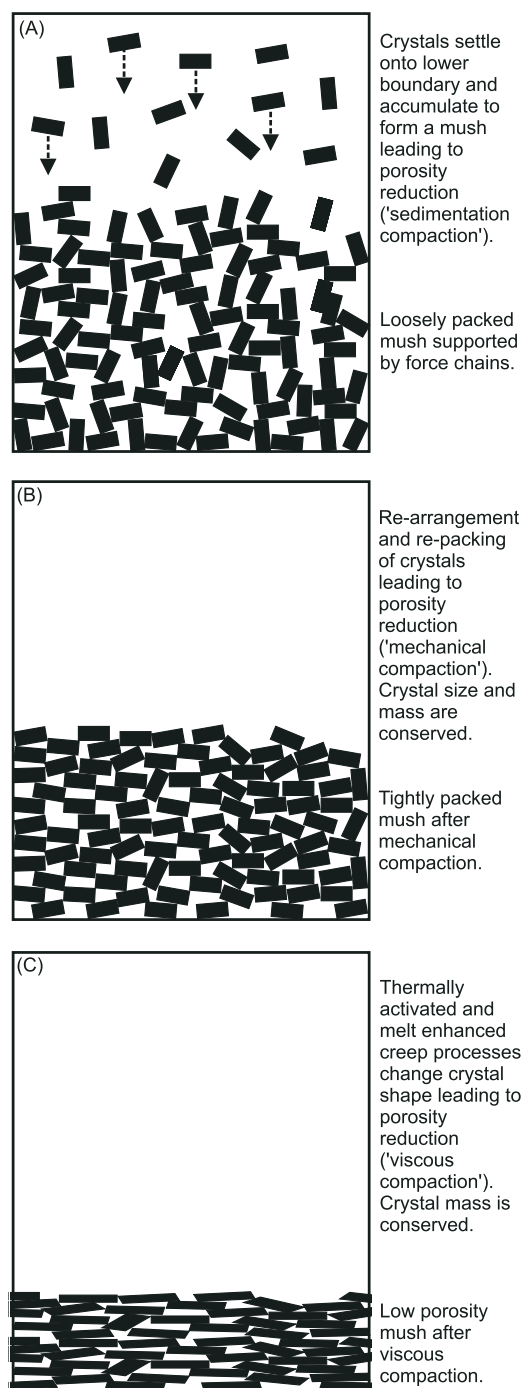


Fig. 2. Schematic illustrating compaction at (a) high melt fraction ($\phi > 0.5 - 0.7$; note these melt fraction (porosity) estimates depend strongly on factors such as crystal shape and size distribution, and crystal cohesion (e.g. Philpotts and Dickson, 2000; Revil *et al.*, 2002); see Holness (2018) for a discussion); (b) moderate melt fraction ($0.05 - 0.4 < \phi < 0.5 - 0.7$) and (c) low melt fraction ($\phi < 0.05 - 0.4$).

Kohlstedt, 1984; Dell'Angelo *et al.*, 1987). The presence of melt can facilitate diffusion creep, by providing higher diffusivity pathways for the transport of solid components from sites of high stress to sites of low stress. The prevalence of viscous compaction in crustal magmatic systems has been questioned, at least as formulated in numerical models where matrix deformation during compaction is assumed to be homogeneous and Newtonian (e.g. Bergantz, 2019; Holness, 2018).

Matrix deformation may be highly heterogeneous due to the presence of force chains: a network of contacting solid grains that transfer a large proportion of the applied stress and are largely responsible for the strength of granular materials (e.g. Bergantz *et al.*, 2015; Zhang and Kamrin, 2017). In mush reservoirs, creep mechanisms may allow the crystal morphology to change, such that the force chains relax and allow compaction to occur.

Compaction requires relative motion of melt and crystals, consistent with the requirement of relative motion to produce chemical differentiation: melt lost from the system corresponds to the evolved product of differentiation, while compacted solid corresponds to the refractory residue. Melt loss by flow relative to the solid must be accommodated by compaction to ensure conservation of mass. Yet microstructural evidence for crystal deformation consistent with compaction at low melt fraction is scarce in igneous rocks, at least those formed in relatively shallow magmatic systems (Holness, 2018). There is an apparent contradiction between the need for compaction to occur in mush reservoirs to allow melt loss during chemical differentiation, and the absence of evidence for widespread compaction in the rock record. Compaction is widely observed to occur in other earth and industrial systems, so it is curious that mush reservoirs should apparently be the exception.

Melt fraction and composition change by reactive flow

Finally, we introduce the third mechanism considered here that can change melt fraction and composition in crustal mush reservoirs. Consider a conceptual experimental melting apparatus in which melt can be added to a sample, and solid can be removed from a sample, such that the sample bulk composition changes at constant mass. Adding melt in this way is designed to mimic reactive flow, in which relative flow of melt and solid causes melt to enter and solid to leave an REV in a magma reservoir. The composition change occurs in a thermally insulated chamber so no heat is gained or lost from the surroundings, but the temperature can be measured once the new local equilibrium is established. We place 100 g of solid diopside (an REV with bulk composition An_0 ; $C = 0$) in the apparatus, shown as case C in Fig. 1, initially at room temperature (20 °C), and add 141.0 kJ of energy to bring the sample to a temperature of 1325°C. We bring a eutectic melt (composition An_{41} ; $C_1 = 0.41$) to the same temperature; eutectic melt at such high temperature is not in equilibrium with any solid composition (Fig. 1). We use the apparatus to replace a mass fraction of 0.92 of solid diopside in the sample with an equivalent mass fraction of the eutectic melt; in other words, we replace most, but not all, of the solid diopside with eutectic melt at constant mass. Note that for An_{41} at 1325°C the heat content of the melt is higher than that of the solid by 0.50 MJ/kg, so adding melt also adds heat.

We first observe the temperature response of the apparatus. Despite adding heat, we observe a significant decrease in sample temperature to 1286°C. This occurs because diopside dissolves into the eutectic melt that was added, and the latent heat required consumes sensible heat. We quench the sample and find that the melt fraction is $\phi = 1$; having added 0.92 eutectic melt fraction, the final melt fraction is higher. We repeat the experiment, adding progressively smaller amounts of eutectic melt in each cycle, recording the temperature, and then quenching and measuring the melt fraction, melt and solid compositions, from which we calculate the new bulk composition of the sample.

The decrease in temperature caused by mixing means that, unlike compaction, the reactive flow results plot with decreasing temperature as the local bulk composition (An content) increases (Fig. 1a). However, the higher heat content of the added melt means that enthalpy increases with increasing An content (Fig. 1b). The most surprising results are in Fig. 1c and 1d: in Fig. 1c, melt fraction increases with decreasing temperature, while in Fig. 1d, the final melt fraction is always higher than the added melt fraction. Both of these apparently counter-intuitive results are a consequence of reaction caused by relative motion of melt and solid into, and out of, the sample (REV); here, the reaction increases melt fraction as compared to simple mixing (or compaction), and consumes latent heat at the expense of sensible heat.

This simple conceptual experiment shows that melt fraction can change in response to changes in bulk composition at constant enthalpy, as well as the more typically invoked changes in enthalpy (temperature) at constant bulk composition. We term this process of melt fraction change 'reactive flow', because changes in local bulk composition in natural systems must be caused by flow of melt relative to matrix, and the resulting changes in melt fraction occur in response to reaction (component exchange) between melt and solid (Aharonov *et al.*, 1995; Jackson *et al.*, 2018; Keller and Katz, 2016b; Keller and Suckale, 2019; Solano *et al.*, 2014). Reactive flow is therefore distinct from compaction (case B), in which melt fraction changes without component exchange. The extra melt present in the case of reactive flow (case C) can be thought of as 'chemical melt', in contrast to the 'thermal melt' produced by heating (case A). Chemical melting is a result of changing the local bulk composition. Everyday analogies include adding salt to an icy road in winter to produce a salty liquid.

In the interests of simplicity, we have chosen to isolate these three cases and demonstrate each in a simple, two-component binary system. However, in natural systems, the processes of heating/cooling, compaction and reactive flow occur simultaneously; for example, flow can lead to both compaction and reaction, and reaction can exchange latent and sensible heat, causing heating and cooling. It is not realistic to assume isothermal, adiabatic or zero flow conditions in crustal magmatic systems. To identify and quantify the different contributions requires a mathematical description of each.

Mathematical description of melt fraction and composition change

Melt fraction and composition changes from mass and component conservation

We derive here expressions for melt fraction and composition change in terms of melt and solid composition for a two-phase (solid and melt), two-component system. The general case for multiple phases and components is presented in Appendix A.

We begin with the well-known mass conservation (continuity) equation

$$\frac{\partial \phi}{\partial t} + \nabla \cdot \phi \mathbf{u}_l = \Gamma \quad (1a)$$

$$\frac{\partial(1-\phi)}{\partial t} + \nabla \cdot (1-\phi) \mathbf{u}_s = -\Gamma \quad (1b)$$

where ϕ , $\mathbf{u}_l$ and $\mathbf{u}_s$ are melt fraction, liquid (melt) and solid (matrix) velocity, respectively. For simplicity, we have invoked the

Boussinesq approximation, neglecting density variations except when gravitational forces are considered. The Boussinesq approximation is widely used in fluid dynamics, including in models of phase change and melt flow in the crust and mantle (e.g. Jackson and Cheadle, 1998; Jackson *et al.*, 2018, 2003; Jones *et al.*, 1997; Katz, 2008; Keller and Katz, 2016b; Solano *et al.*, 2014, 2012; Turner *et al.*, 2017); here it means that we conserve volume rather than mass, which significantly simplifies the numerical solutions we show later and allows validation of the numerical solutions against analytical solutions. The volume error for the density values used here (see Appendix C) is of order 15%

The equivalent expression for component conservation is given by

$$\frac{\partial \phi C_l}{\partial t} + \nabla \cdot \phi C_l \mathbf{u}_l = \Gamma_c \quad (2a)$$

$$\frac{\partial(1-\phi)C_s}{\partial t} + \nabla \cdot (1-\phi)C_s \mathbf{u}_s = -\Gamma_c, \quad (2b)$$

where C_l and C_s are liquid and solid composition, respectively. Γ_c is the rate of component transfer from solid to liquid. We neglect Fickian diffusion of components through the melt and solid phases, recognizing that this is slow; as we show in the Discussion, component transport at length-scales greater than a few cm is dominated by advection rather than diffusion.

The source terms Γ and Γ_c depend on the chemical model, such as a phase diagram, used to relate local bulk, melt and solid composition to melt fraction, temperature and pressure (e.g. Solano *et al.*, 2014; Weatherley and Katz, 2012). Here we begin by eliminating Γ and Γ_c to derive equations for melt fraction and composition change that are independent of the chemical model. Applying the chain rule to equation 2(a), we have

$$C_l \frac{\partial \phi}{\partial t} + \phi \frac{\partial C_l}{\partial t} + C_l \nabla \cdot \phi \mathbf{u}_l + \phi \mathbf{u}_l \cdot \nabla C_l = \Gamma_c \quad (3)$$

$$\Leftrightarrow C_l \left(\frac{\partial \phi}{\partial t} + \nabla \cdot \phi \mathbf{u}_l - \Gamma \right) + \phi \frac{\partial C_l}{\partial t} + \phi \mathbf{u}_l \cdot \nabla C_l + \Gamma C_l - \Gamma_c = 0 \quad (4)$$

$$\Leftrightarrow \phi \frac{\partial C_l}{\partial t} + \phi \mathbf{u}_l \cdot \nabla C_l + \Gamma C_l - \Gamma_c = 0. \quad (5)$$

The same procedure applied to equation 2(b) yields

$$(1-\phi) \frac{\partial C_s}{\partial t} + (1-\phi) \mathbf{u}_s \cdot \nabla C_s - \Gamma C_s + \Gamma_c = 0. \quad (6)$$

Adding equation 5 and 6 to eliminate Γ_c leads to

$$\phi \frac{\partial C_l}{\partial t} + (1-\phi) \frac{\partial C_s}{\partial t} + (1-\phi) \mathbf{u}_s \cdot \nabla C_s + \phi \mathbf{u}_l \cdot \nabla C_l + \Gamma(C_l - C_s) = 0. \quad (7)$$

Re-arranging for Γ and substitution into equation 1(a), the rate of change of melt fraction can be written as

$$\begin{aligned} \frac{\partial \phi}{\partial t} = & - \underbrace{\frac{1}{C_l - C_s} [(1-\phi) \mathbf{u}_s \cdot \nabla C_s + \phi \mathbf{u}_l \cdot \nabla C_l]}_{\text{reactive flow}} - \underbrace{\nabla \cdot \phi \mathbf{u}_l}_{\text{compaction}} \\ & - \underbrace{\frac{1}{C_l - C_s} \left[\phi \frac{\partial C_l}{\partial t} + (1-\phi) \frac{\partial C_s}{\partial t} \right]}_{\text{heating/cooling}}. \end{aligned} \quad (8)$$

Applying a similar approach to bulk composition $\bar{C}$ yields

$$\frac{\partial \bar{C}}{\partial t} = - \underbrace{[(1 - \phi)\mathbf{u}_s \cdot \nabla C_s + \phi\mathbf{u}_l \cdot \nabla C_l]}_{\text{reactive flow}} - \underbrace{(C_l - C_s)\nabla \cdot \phi\mathbf{u}_l}_{\text{compaction}} \quad (9)$$

Equation (8) shows that the change in melt fraction can be expressed in terms of three different contributions (the three terms on the right hand side), while equation 9 shows that the change in bulk composition can be expressed in terms of two different contributions. We discuss the physical meaning of these terms in the next section.

When $C_l = C_s$ we have a singular point at which equation 8 cannot be applied, because phase change occurs at constant bulk, melt and solid composition and melt fraction is no longer constrained by the lever rule

$$\bar{C} = \phi C_l + (1 - \phi)C_s. \quad (10)$$

At these points, melt fraction change occurs by melting or freezing at fixed temperature and composition, and is a function only of latent heat exchange, given by

$$\frac{d\phi}{dt} = \frac{1}{L_f} \frac{dH}{dt}, \quad (11)$$

where H is the enthalpy per unit mass and L_f is the latent heat, which is assumed to be constant. Such singular points occur at specific locations in the phase space of the chemical model; for example, in a binary eutectic phase diagram when $C_l = C_s = C_{ae}$ where C_{ae} is the eutectic composition (e.g. Fig. 1), or in a binary solid solution when $C_l = C_s = 0$ or 1.

Melt fraction and composition changes in response to heating/cooling, compaction and reactive flow

To identify the physical meaning of the terms in equations (8) and (9), we can substitute the equations describing our example chemical model of a simple binary phase diagram (see Appendix B). For $T > T_s$ we obtain

$$\begin{aligned} \frac{\partial \phi}{\partial t} = & - \underbrace{\frac{2a_1\phi}{(b_1 + \sqrt{b_1^2 - 4a_1(c_1 - T)})\sqrt{b_1^2 - 4a_1(c_1 - T)}}[\mathbf{u}_l \cdot \nabla T]}_{\text{reactive flow}} \\ & - \underbrace{\nabla \cdot \phi\mathbf{u}_l}_{\text{compaction}} - \underbrace{\frac{2a_1\phi}{(b_1 + \sqrt{b_1^2 - 4a_1(c_1 - T)})\sqrt{b_1^2 - 4a_1(c_1 - T)}}\frac{\partial T}{\partial t}}_{\text{heating/cooling}} \end{aligned} \quad (12)$$

$$\begin{aligned} \frac{\partial \bar{C}}{\partial t} = & \underbrace{\frac{\phi}{\sqrt{b_1^2 - 4a_1(c_1 - T)}}[\mathbf{u}_l \cdot \nabla T]}_{\text{reactive flow}} \\ & + \underbrace{\frac{b_1 + \sqrt{b_1^2 - 4a_1(c_1 - T)}}{2a_1}\nabla \cdot \phi\mathbf{u}_l}_{\text{compaction}} \end{aligned} \quad (13)$$

The constants a_1 , b_1 and c_1 are chosen to fit the liquidus curve of the binary phase diagram to experimental data (see Appendix B).

Having substituted the equations describing our example chemical model, it becomes clear that the final (third) term on the right-hand-side (RHS) of equation (8) represents static heating/cooling (phase change with no flow of melt or solid), because it is expressed in equation (12) in terms of the time rate of change of temperature. Indeed, for a system in local equilibrium, the third term on the RHS of equation (8) represents phase change for any chemical model that describes phase composition in terms of temperature and pressure. Other chemical models used to describe phase change in the crust and mantle (e.g. Solano et al., 2014; Weatherley and Katz, 2012) yield the same interpretation. There is no corresponding term in equation (9), because phase change does not change the bulk composition: changes in melt and solid composition occur because components are exchanged between phases, but the bulk composition remains constant.

The second term on the right hand side of equation (8) shows that melt fraction changes when there is a non-zero divergence of melt (or solid) velocity, in which case there is a net accumulation or loss of melt. The corresponding term on the right hand side of equation 9 shows that bulk composition also changes when there is a non-zero divergence of melt (or solid) velocity and the melt and solid compositions are different. This term in each equation represents compaction and remains the same irrespective of the chemical model, because it involves no temporal or spatial changes in melt or solid composition (i.e. it is not associated with component exchange between phases).

The first term on the right hand side of equations (8) and (9) shows that melt fraction and local bulk composition change when the melt and/or solid have non-zero velocity and there is a gradient in composition. This term in each equation does not correspond to heating/cooling or compaction, yet is obtained from the standard expressions for conservation of mass and components. For a system in local equilibrium, the term represents reactive flow, consistent with the widely accepted physical definition of reactive flow as the situation where fluid flow drives chemical reaction (e.g. Ladd and Szymczak, 2021). When flow of melt and/or solid advects (transports) components through a gradient in composition, the system moves out of local chemical equilibrium, causing reaction that returns the system to local equilibrium. If the velocity is zero there is no flow; if the composition is uniform there is no reaction. In both cases, there is no reactive flow. Our conceptual experiment in the previous section represented reactive flow by replacing solid with non-equilibrium melt in a reaction vessel, re-equilibrating the new mixture and measuring the new melt fraction and bulk composition.

For our example binary phase diagram, the reactive flow term is expressed in terms of temperature (equations 12 and 13), because temperature dictates melt composition (see Appendix B). Moreover, there is no contribution to melt fraction or composition change from reactive flow of the solid crystals during compaction, because the solid composition is uniform when $T > T_s$ (Appendix B) so terms including solid gradients in equations (8) and (9) are zero.

Equations (8) and (9) provide direct insight into when reactive flow is important in a system in local equilibrium. For example, reactive flow is more significant in systems with steep gradients in melt and/or solid composition and/or high flow velocities, so long as local equilibrium is maintained. If composition is uniform, then there is no reactive flow; this means that there is no reactive flow during phase change of a pure substance, or in a two-component substance with the eutectic composition.

Quantifying the contribution of heating/cooling, compaction and reactive flow to melt fraction and composition change

The terms on the right hand side of equations (8) and (9) provide a framework to quantify the contributions of heating/cooling, compaction and reactive flow to melt fraction and bulk composition change. We can calculate the cumulative change in local melt fraction and bulk composition (i.e. sum the changes in melt fraction and bulk composition over time) caused by each mechanism at a given spatial location by integrating each of the terms over time. The contributions of heating/cooling, compaction and reactive flow to melt fraction change at a given time t are therefore given by

$$\Delta\phi_H = - \int \frac{1}{C_l - C_s} \left[\phi \frac{\partial C_l}{\partial t} + (1 - \phi) \frac{\partial C_s}{\partial t} \right] dt \quad (14a)$$

$$\Delta\phi_C = - \int \nabla \cdot \phi \mathbf{u}_l dt \quad (14b)$$

$$\Delta\phi_R = - \int \frac{1}{C_l - C_s} [(1 - \phi) \mathbf{u}_s \cdot \nabla C_s + \phi \mathbf{u}_l \cdot \nabla C_l] dt, \quad (14c)$$

where the subscripts H , C and R refer to melting/cooling, compaction and reactive flow respectively. The contributions to changes in local bulk composition are likewise given by

$$\Delta\bar{C}_H = 0 \quad (15a)$$

$$\Delta\bar{C}_C = - \int (C_l - C_s) \nabla \cdot \phi \mathbf{u}_l dt \quad (15b)$$

$$\Delta\bar{C}_R = \int [(1 - \phi) \mathbf{u}_s \cdot \nabla C_s + \phi \mathbf{u}_l \cdot \nabla C_l] dt. \quad (15c)$$

Although equations (8) and (9) provide a mathematical definition of melt fraction and composition change, along with a means to quantify the different contributions for our example chemical model using equations (14) and (15), the mechanisms are strongly cross-coupled in all cases of interest to crustal magma reservoir dynamics. Moreover, to deliver quantitative estimates of the contribution of each mechanism, we require a flow model to determine the phase velocities (e.g. Jackson *et al.*, 2018; Ribe, 1985; Solano *et al.*, 2014; Weatherley and Katz, 2012). We also need to ensure that we conserve energy and momentum, in addition to conserving mass and components (e.g. Jackson and Cheadle, 1998; Jackson *et al.*, 2018; Katz, 2008; Solano *et al.*, 2014; Weatherley and Katz, 2012). In the next section, we use numerical modelling to demonstrate end-member cases where each mechanism dominates, and quantify the contribution of all three mechanisms in a geologically realistic model.

Numerical modelling of melt fraction and composition changes in a two-phase, two-component system relevant to crustal magma reservoirs

Flow and transport model

The one-dimensional (1D) numerical model used here is extended from that presented by Solano *et al.* (2014) and solves the mass and component conservation equations (1) and (2) along with heat and momentum conservation, including heating/cooling, compaction and reactive flow (see Appendix C for details). In common with previous studies (e.g. Jackson *et al.*, 2018; Katz, 2008; Solano *et al.*, 2014), we conserve enthalpy and neglect irreversible processes

such as viscous dissipation and radiogenic heat balance in our energy conservation equation. Melting yields a single liquid (melt) phase so the model contains one (solid or liquid) or two (solid and liquid) phases. Each phase can contain two components.

Compaction of the solid matrix is assumed to occur by melt-enhanced diffusion creep, yielding a Newtonian rheology. Unless otherwise stated, permeability and matrix bulk and shear viscosities are functions of melt fraction; melt viscosity and melt and matrix densities are functions of composition (Appendix C). The melt is buoyant relative to the solid matrix. Material properties used in the numerical models are outlined in Table 2.

Chemical model

The chemical model uses data obtained from melting experiments at constant pressure on samples of natural basalt and dacite (Fig. 3; see Appendix B for details). We plot the experimentally determined SiO_2 content (in wt %, normalised to 100% anhydrous) as a function of temperature: melt SiO_2 content was taken directly from the published experimental glass analysis; solid SiO_2 content was determined from mass balance. Perhaps surprisingly, the data are well modelled using a binary eutectic, similar to the anorthite-diopside phase diagram used in the earlier section, but with the eutectic composition corresponding to the most evolved (highest SiO_2) composition. Melt SiO_2 content varies with temperature; solid SiO_2 remains approximately constant for a given sample, plotting as a vertical line (see Appendix B for further discussion). The experimental data demonstrate that evolved (high SiO_2) melt can be in equilibrium with refractory (low SiO_2) crystals. Application of this fixed pressure phase diagram is restricted to models of relatively limited vertical extent.

Numerical experiments

We use the numerical model to investigate two different scenarios. In each scenario, we quantify the contribution of heating/cooling, compaction and reactive flow to melt fraction and composition change at each spatial location in the model using equations (14) and (15). The first scenario is a simple column of rock, in which numerical experiments are used to demonstrate end-member cases where melt fraction and composition changes are dominated by either heating/cooling, compaction or reactive flow. This scenario lacks geologic realism, but is useful to demonstrate the key concepts; moreover, analytical solutions are obtainable for the end-member cases that we use to validate the numerical model. The second scenario is more geologically realistic; we use a numerical experiment to demonstrate changes in melt fraction and local bulk composition after intrusion of a basalt sill into solid crust of the same composition. The magma in the sill cools and crystallises, and compaction and reactive flow redistribute components.

End-member models of heating/cooling, compaction and reactive flow

The simple column of rock has length 50 m and is sealed and perfectly insulated along its sides so flow and transport only occur in the vertical direction. In each end-member case, the model is run for 100 y to show how temperature, melt fraction and composition change from the initial conditions.

Heating/cooling end-member

We first consider a rock column that is also sealed at the top and base. The column has uniform initial bulk composition ($\bar{C} = 51\% \text{ SiO}_2$), corresponding to a typical basalt (Figure. 3) and initially contains a layer of hot, pure melt at its centre ($\phi = 1$)

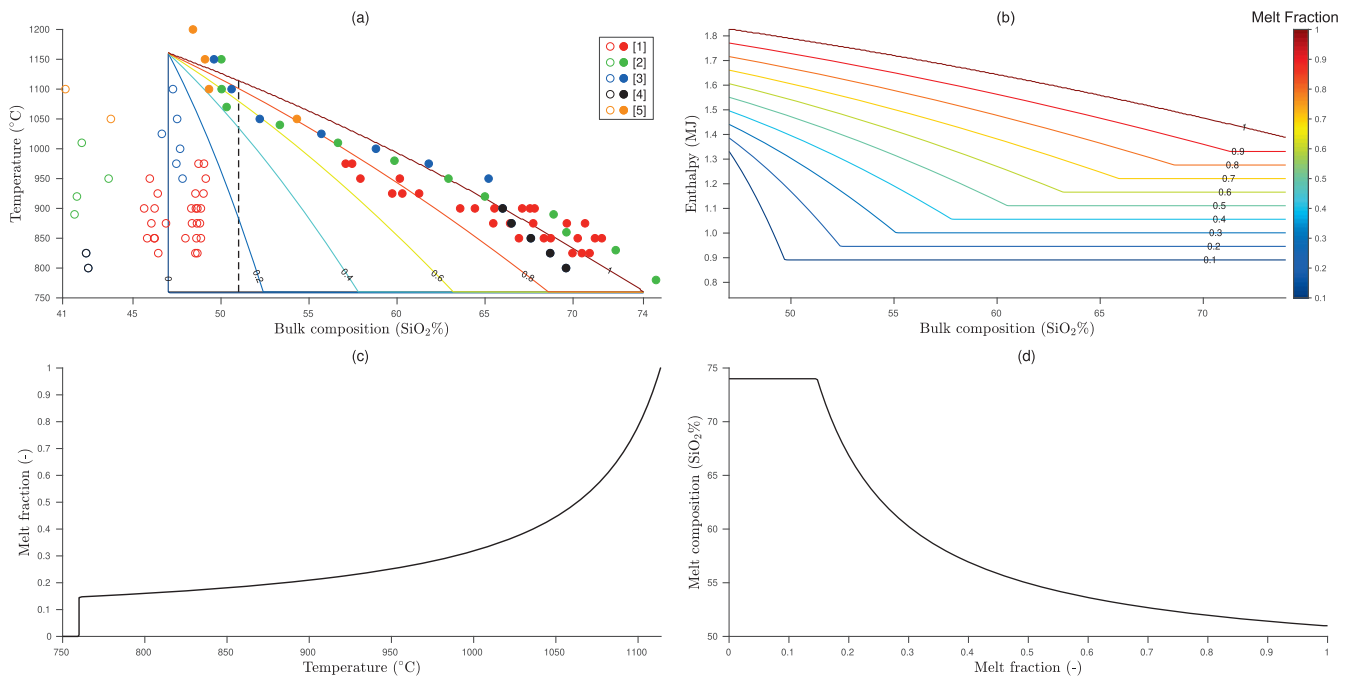


Fig. 3. Binary eutectic phase diagram, matched to data from melting experiments using samples of basalt and dacite, expressing melt fraction as a function of (a) composition (SiO₂%) and temperature; the black dashed line indicates the basalt chosen as the starting composition in our example cases; (b) composition and enthalpy; (c) shows melt fraction as a function of temperature for our chosen basalt and (d) shows melt composition (SiO₂%) as a function of melt fraction for our chosen basalt. Data points in (a) are from [1]: *Sisson et al. (2005)*; [2]: *Nandedkar et al. (2014)*; [3]: *Blatter et al. (2013)*; [4]: *Blatter et al. (2017)*; [5]: *Melekhova et al. (2015)*, with solid and hollow symbols representing melt and solid, respectively. See Appendix B for details of the parent rock samples, calculation of solid SiO₂ content and mathematical description.

with low melt fraction mush above and below ($\phi = 0.15$; Fig. 4a,e). We assume that the mush is rigid. The properties of the phase diagram have been modified in this case (see Appendix D) so that we can develop an analytical solution for melt fraction. This scenario is motivated by the numerous previous thermal models of sill intrusion (e.g. *Hodge, 1974*; *Petford and Gallagher, 2001*; *Annen et al., 2005*; *Karakas et al., 2017*) and represents the situation immediately following intrusion of hot magma into a crustal mush reservoir.

The rigid mush cannot compact, so there is no flow of melt or matrix, confirming our previous observation that in a closed system, without compaction there can be no flow. Consequently, heat transport occurs only by conduction: the melt layer cools as heat is lost to, and partially melts, the surrounding mush (Fig. 4a–d; e–h). Note our numerical solution closely matches the analytical solution, confirming that our numerical model correctly solves for conductive heat transport and associated thermal melting and freezing (Fig. 4a–d).

As predicted by the binary phase diagram (Fig. 3d), the local melt composition evolves to become more silicic as the melt layer cools (Fig. 4i–l). The local bulk composition also remains constant: changes in melt composition are associated with changes in melt fraction such that, although melt composition changes, bulk composition is fixed. As expected, changes in melt fraction are dominated by heating/cooling because there is no flow (Fig. 4m–p). Within the melt layer, changes in melt fraction are negative because the melt fraction decreases in response to cooling; above and below the melt layer, changes in melt fraction are positive because melt fraction increases in response to heating. Melt fraction changes are most pronounced at the contacts between the melt layer and the surrounding mush, because the most significant changes in temperature occur here over the duration

of the simulation. There are no changes in bulk composition, because there is no flow (Fig. 4q–t).

Compaction end-member

We next consider the same rock column that is sealed at the top and base and has initial bulk composition corresponding to basalt with $\bar{C} = 51\%$ SiO₂ (Fig. 3). The column initially contains a uniform melt fraction ($\phi = 0.3$; Fig. 5a) at uniform temperature ($T = 990$ °C; Fig. 5e). This scenario is motivated by previous studies of melt fraction change via pure compaction (e.g. *McKenzie, 1985*).

The buoyant melt migrates upwards and accumulates at the top of the sealed column, so the melt fraction locally increases as the matrix decompacts, whereas loss of melt from deeper in the column is caused by compaction (Fig. 4b–d). Similar solutions for melt fraction have been obtained in previous studies and, in the limit of constant matrix bulk and shear viscosity and constant melt shear viscosity, melt fraction in the compacting boundary layer at the base of the column can be calculated without recourse to numerical methods (*McKenzie, 1985*). The analytical solution (see Appendix D) matches closely our numerical solution (Fig. 4b–d), where we have chosen to use constant solid viscosity to facilitate comparison (Table 2). This confirms that our numerical model correctly solves for melt and solid velocity and associated melt fraction changes during compaction.

The local bulk composition at the top of the column changes to become more evolved, reflecting the accumulation of melt; likewise, the local bulk composition at the base becomes more refractory, reflecting the loss of melt (Fig. 5i–l). However, as predicted by the binary phase diagram (Fig. 3), the melt and solid compositions remain constant, because the temperature remains constant (Fig. 5i–l). Temperature remains constant (Fig. 5e–h), because there is no phase change and hence no exchange of

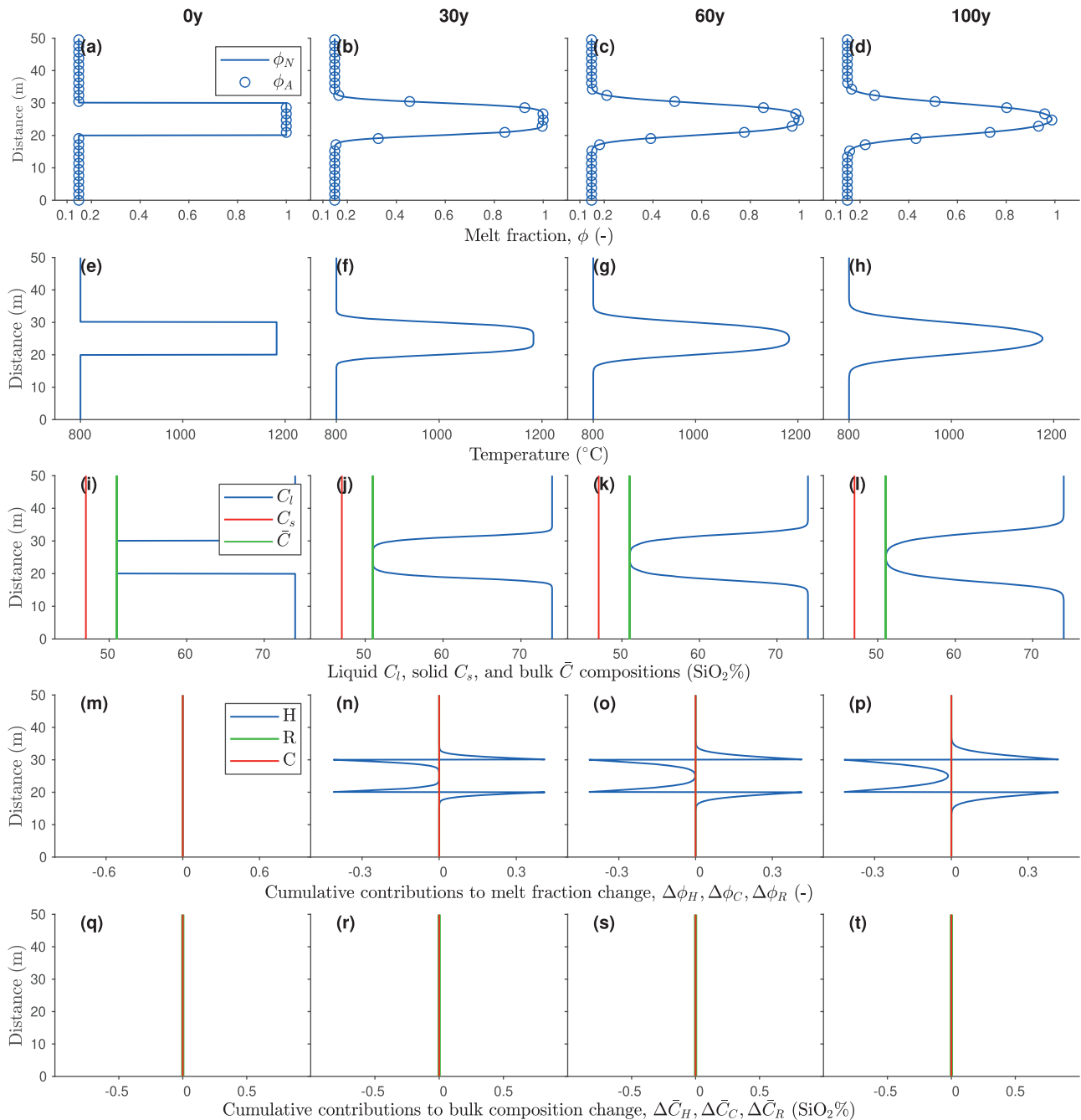


Fig. 4. Snapshots of the time evolution of a partially molten, insulated column, showing: (a–d) melt fraction, including results from our numerical model (subscript N) and equivalent analytical solutions (subscript A); (e–h) temperature; (i–l) liquid, solid and bulk composition (SiO₂%); (m–p) cumulative change in melt fraction due to heating/cooling (H), compaction (C) and reactive flow (R) and (q–t) cumulative change in bulk composition (SiO₂%). This example is designed to be dominated by heating/cooling, with a rigid matrix and no flow. Snapshots are shown at the initial condition (0 y), and then after 30 y, 60 y and 100 y.

latent heat, and is uniform so there is no net advection of sensible heat. Changes in melt fraction and local bulk composition are dominated here by compaction (Fig. 5m–t). At the top of the column, changes in melt fraction and composition are positive because the melt fraction increases and the bulk composition becomes more evolved, and vice versa.

These results confirm that compaction alone leads to chemical differentiation. The bulk composition lies on a simple mixing curve (Fig. 6), reflecting the lack of component exchange between solid and melt (Fig. 5c); this is a characteristic feature of differentiation by pure compaction (Fig. 1e). The gradient of the mixing

curve depends on temperature, because the melt composition becomes more evolved as the temperature decreases and vice versa; Fig. 6 also shows the ‘end-member’ mixing curves for the maximum ($T = 1400\text{ °C}$) and minimum ($T = 760\text{ °C}$) temperatures defined in the phase diagram (Fig. 3). If compaction dominates, melt fraction and bulk composition must plot on one of the family of mixing curves between these end-members.

Reactive flow end-member

We finish by considering the same rock column, but now impose a gradient in initial bulk composition from $\bar{C} = 57\%$ SiO₂ at the

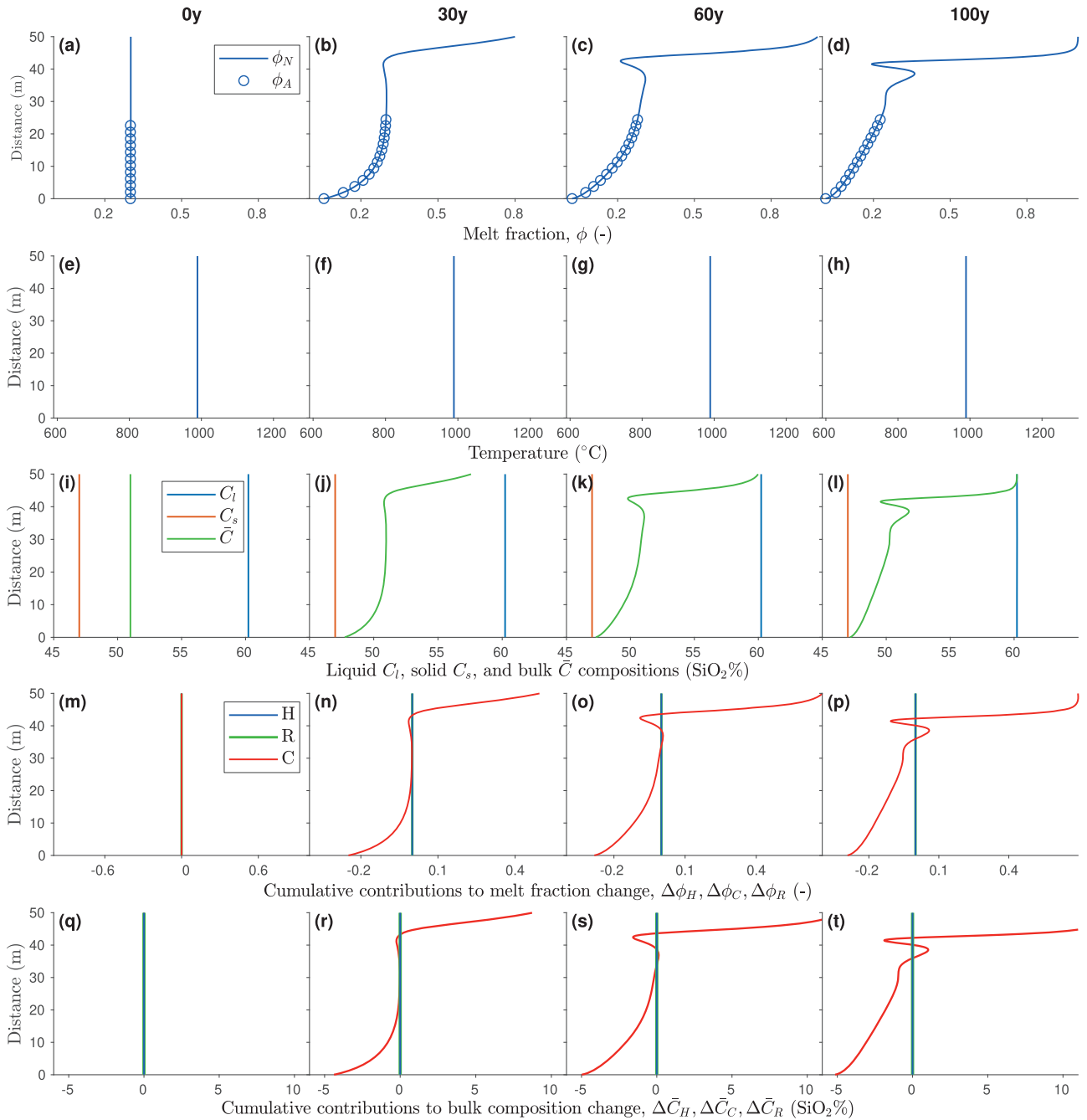


Fig. 5. Snapshots of the time evolution of a partially molten, insulated column with uniform temperature, showing (a–d) melt fraction, including results from our numerical model (subscript N) and equivalent analytical solutions (subscript A); (e–h) temperature; (i–l) liquid, solid and bulk composition ($\text{SiO}_2\%$); (m–p) cumulative change in melt fraction due to heating/cooling (H), compaction (C) and reactive flow (R) and (q–t) cumulative change in bulk composition ($\text{SiO}_2\%$). This example is designed to be dominated by compaction, with uniform temperature and a deformable matrix. Snapshots are shown at the initial condition (0 y), and then after 30 y, 60 y and 100 y.

base to $\bar{C} = 48\%$ SiO_2 at the top. We fix the temperature at the top and base such that the temperature initially increases linearly from 810°C at the base to 1100°C at the top (Fig. 7e); the corresponding initial melt fraction decreases from 0.4 at the base to 0.1 at the top (Fig. 7a). Melt composition, which is a function of both local bulk composition and temperature, initially decreases from 71% SiO_2 at the base to 52% SiO_2 at the top (Fig. 7i). The column has melt injected into the base, at constant rate and with the same composition as the initial melt at the base, causing melt to be expelled from the top of the column. This case is designed to minimize temperature changes caused by latent heat

exchange during reactive flow: the steep temperature gradient imposed across the rock column corresponds to a large heat flux. This flux dominates the comparatively small quantity of latent heat absorbed by chemical melting near the top of the column and released by chemical freezing near the base of the column. Consequently, although temperature can vary within the column, it remains approximately fixed with the same gradient as initially imposed. We assume the mush is rigid. An analytical solution for this case can be found in Appendix D and the close match confirms that our numerical model correctly solves for component transport and associated chemical melting and freezing.

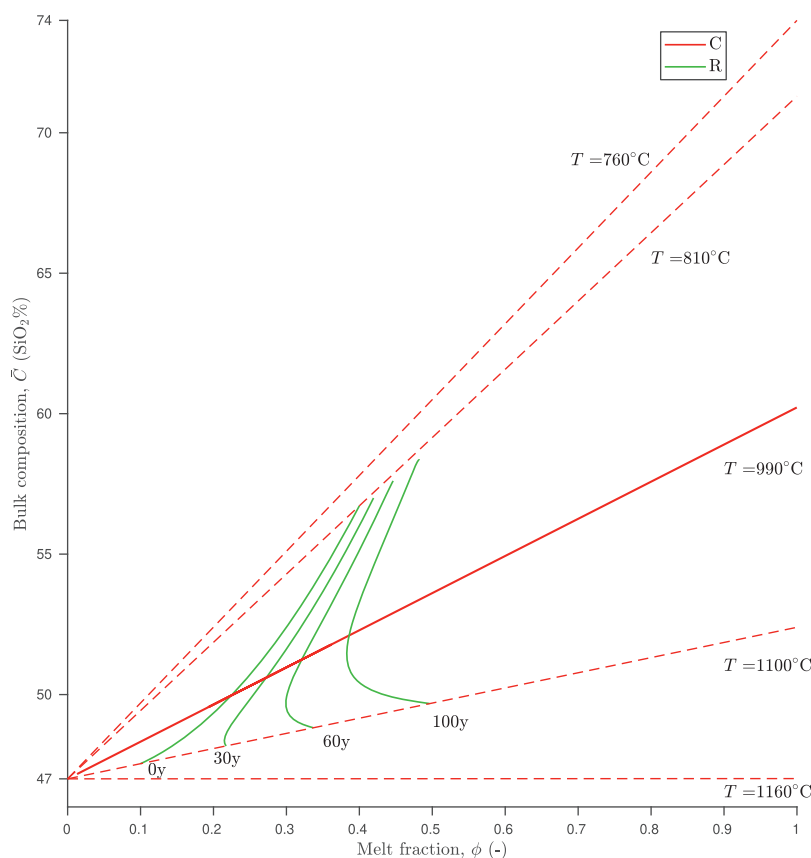


Fig. 6. Local bulk composition ($\text{SiO}_2\%$) versus melt fraction for the compaction-dominated case (solid red line) (Fig. 5) and reactive flow-dominated case (green lines) (Fig. 7). The compaction-dominated case always plots on a mixing curve; the reactive flow-dominated case deviates from the mixing curves and the relationship depends on the time step considered. Reactive flow results correspond to the snapshots at the initial condition (0 y), and then 30 y, 60 y and 100 y, shown in Fig. 7. These curves begin and end on the mixing curves that correspond to the maximum ($T = 1100^\circ\text{C}$) and minimum ($T = 810^\circ\text{C}$) temperatures in the model. Also shown are the mixing curves (red dashed lines) for the maximum ($T = 1400^\circ\text{C}$) and minimum ($T = 760^\circ\text{C}$) temperatures in the model phase diagram (Fig. 3).

As the more evolved melt injected into the base of the column flows upwards through the column, it reacts with the solid phase so the local bulk composition at the top of the column changes to become more evolved (Fig. 7i–l). The melt fraction also increases at the top of the column, such that melt fraction no longer decreases linearly with height (Fig. 7b–d). The additional melt is created by ‘chemical melting’ rather than ‘thermal melting’ that occurs in response to addition of heat. The temperature remains approximately fixed so there is no heating or cooling (Fig. 7f–h), and there is no compaction because the mush is rigid. Hence, melt fraction and composition change occurs only by reactive flow (Fig. 7m–t): reactive flow occurs because the melt flows through a composition gradient (Fig. 7i–l). This case demonstrates that reactive flow gives rise to melt fraction and composition change, independent of heating/cooling and compaction. It also demonstrates some of the non-intuitive consequences of reactive flow: here, a more evolved (rhyolitic) melt chemically melts a more refractory (basaltic) mush. Such a scenario may seem unlikely at first sight, but this is because conceptual understanding is dominated by consideration of thermal rather than chemical melting. Here, there is no thermal melting; chemical melting occurs simply because reactive flow transports components such that local equilibrium is disturbed, causing chemical reaction.

The local bulk composition no longer lies on the simple mixing curve observed for pure compaction, reflecting component

exchange between solid and melt (Fig. 6). At each time step, the curve relating melt fraction and composition starts and ends on the mixing curves that correspond to the maximum and minimum temperatures in the model, but the gradient of the reactive flow curve differs significantly from the bounding mixing curves. For more evolved bulk composition and at early times, the gradient is typically much steeper than either of the bounding mixing curves, demonstrating that the local bulk composition can be much more evolved or refractory than would be predicted by pure compaction for the same melt fraction and temperature. For more refractory bulk composition and later times, the gradient of the reactive flow curves is reversed compared to the mixing curves, with the local bulk composition becoming more evolved with decreasing, rather than increasing, melt fraction. These deviations from simple mixing are characteristic of differentiation by reactive flow. Here, the local bulk composition is typically more evolved for a given melt fraction than would be observed for pure compaction (mixing), because we inject more evolved melt into the base of the column.

Sill intrusion model

The second scenario considers the more geologically realistic case of a single basaltic sill of 100m thickness, with initial composition $\bar{C} = 51\% \text{ SiO}_2$, intruded at 10km depth into initially solid basalt of the same composition, with an initial linear geotherm of 25 K/km. The model extends vertically from surface to 20 km depth, and

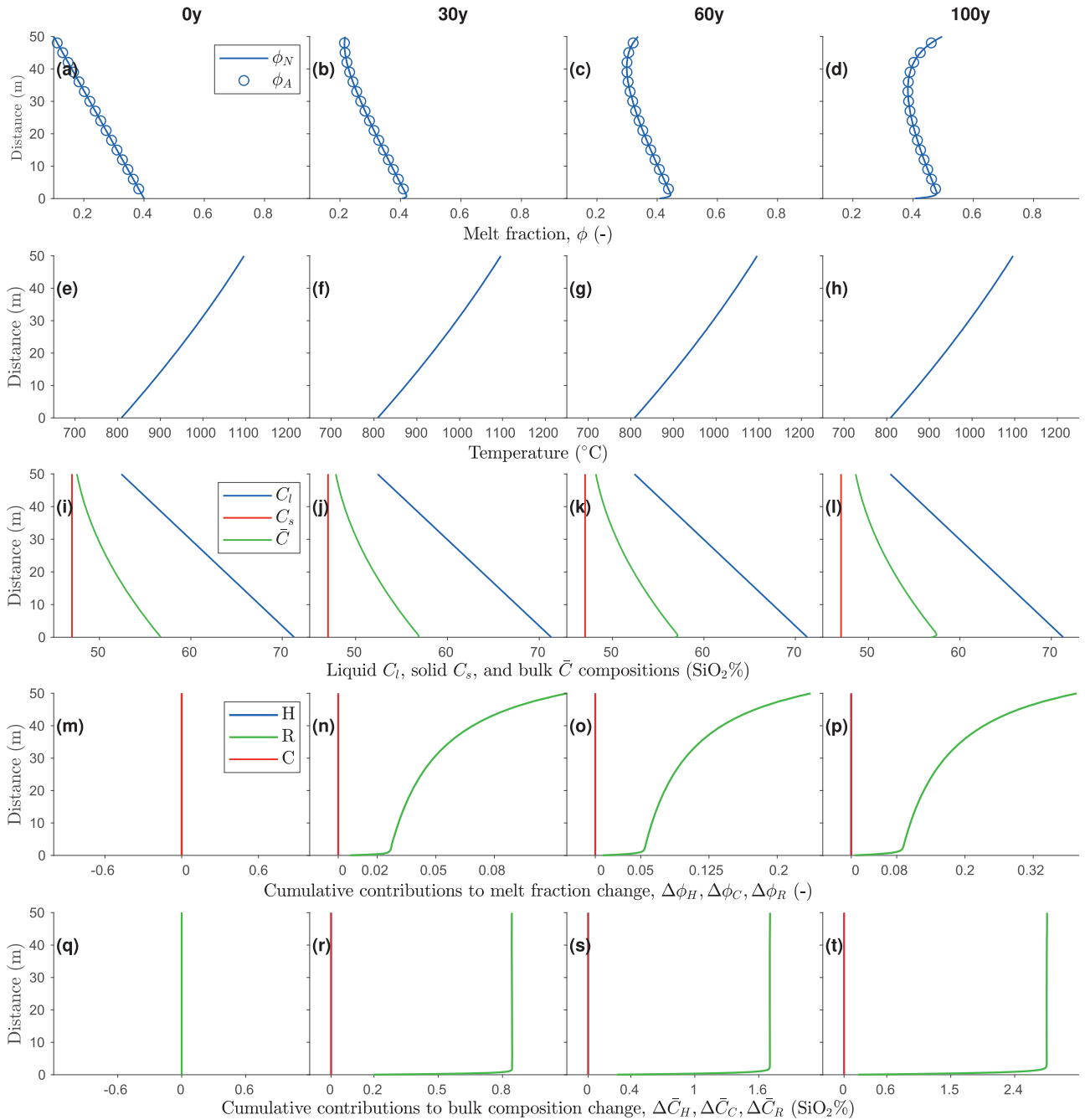


Fig. 7. Snapshots of the time evolution of a partially molten column embedded in a large domain with a fixed temperature gradient, showing (a–d) melt fraction, including results from our numerical model (subscript N) and equivalent analytical solutions (subscript A); (e–h) temperature; (i–l) liquid, solid and bulk composition (SiO₂%); (m–p) cumulative change in melt fraction due to heating/cooling (H), compaction (C) and reactive flow (R) and (q–t) cumulative change in bulk composition (SiO₂%). This example is designed to be dominated by reactive flow, with a constant temperature gradient and melt flux, and a rigid matrix. Snapshots are shown at the initial condition (0 y), and then after 30 y, 60 y and 100 y.

has boundary conditions of fixed temperature and zero velocity. The model boundaries are far from the sill intrusion, so are not reached by temperature changes. The model parameters are given in Table 2. The initial condition (0 y) corresponds to the situation immediately after intrusion, with an entirely liquid body of basalt magma surrounded by solid basalt crust (Fig. 8a); the basalt magma is initially at its liquidus (Fig. 8f). This initial condition corresponds to the sill intrusion model of Jackson *et al.* (2018).

The sill cools by conductive heat loss into the surrounding solid crust (Fig. 8f–j); temperature decreases upwards in the upper part of the sill and overlying crust, but increases upwards in the

lower part of the sill and underlying crust. An inverted geotherm is typical below sill intrusions and in the lower parts of crustal magma reservoirs, because the magma is hotter than the surrounding crust (e.g. Annen *et al.*, 2005; Hodge, 1974; Jackson *et al.*, 2018; Karakas *et al.*, 2017; Petford and Gallagher, 2001; Solano *et al.*, 2012). Cooling of the magma in the sill is associated with an overall decrease in melt fraction in response to crystallisation until the sill has entirely solidified (Fig. 8a–e). However, as the sill cools, buoyant melt migrates upwards and accumulates near the top of the sill (e.g. Fig. 8c,d), forming a layer of evolved, high melt fraction magma above a refractory residue (e.g. Fig. 8m,n). Once the sill

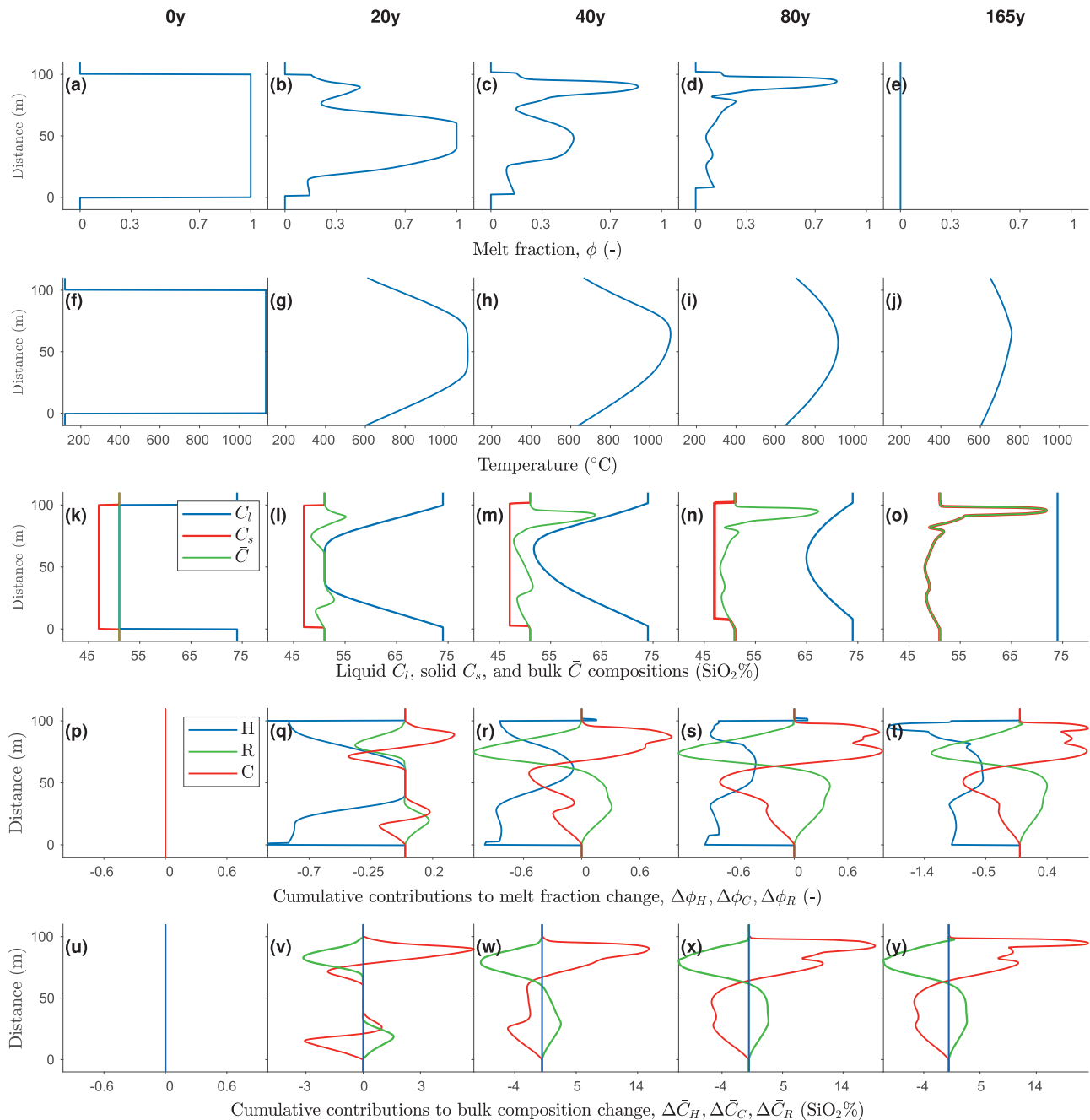


Fig. 8. Snapshots of the time evolution of a basalt sill intrusion showing (a–e) melt fraction; (f–j) temperature; (k–o) liquid, solid and bulk composition ($\text{SiO}_2\%$); (p–t) cumulative change in melt fraction due to heating/cooling (H), compaction (C) and reactive flow (R) and (u–y) cumulative change in bulk composition ($\text{SiO}_2\%$). Snapshots are shown at the initial condition (0 y), and then after 20 y, 40 y, 80 y and finally 165 y when the entire domain is solidified. The model domain extends from surface to 20 km depth; we show here only the sill intrusion and a small portion of the under- and overlying crust.

has solidified, the variations in local bulk composition induced by melt flow are preserved (Fig. 8o).

Similar variations in composition have been observed in some natural sill intrusions and demonstrated in numerical models (e.g. Latypov, 2003; Marsh, 2002; Solano et al., 2012); the mush properties (especially mush permeability and bulk viscosity) were chosen here to produce rapid and significant segregation and accumulation of melt before the sill solidified (see Table 2). Using different mush material properties such as higher viscosity or lower permeability would yield less pronounced differentiation prior to solidification. The time evolution of melt fraction and

composition in the sill are essentially dictated by the relative rates of heat and melt flow: if the former is rapid compared to the latter, then the system cools before melt flow can cause differentiation and vice versa; see Jackson and Cheadle (1998) for a discussion.

The new result here is the quantification of the relative contributions of heating/cooling, compaction and reactive flow to melt fraction and bulk composition change in the cooling sill (Fig. 8p–y). In contrast to the previous ‘end-member’ column models, all three mechanisms contribute in this more realistic example. As expected, cooling of the sill generally causes a decrease in melt fraction; heating of the surrounding rock causes a small

amount of melting, which locally increases melt fraction (e.g. Fig. 8r,s) but, overall, melt fraction change is negative in response to cooling (Fig. 8p–t; see Fig. 8t for the final contributions after the sill has solidified). Cooling has the largest impact on melt fraction at the top and base of the sill where the hot magma is juxtaposed against cold country rock (Fig. 8t). As expected, heating/cooling has no effect on bulk composition (Fig. 8u–y).

The contributions of compaction and reactive flow to melt fraction change are complex, varying both spatially and temporally. At early times, compaction creates approximately symmetrical zones of melt fraction increase (decompaction) overlying melt fraction decrease (compaction) (Fig. 8q). However, at later times, compaction broadly acts to increase melt fraction in the upper half of the sill, and decrease melt fraction in the lower half of the sill, consistent with the column model results reported in the previous section (Fig. 5n–p).

At early times, reactive flow broadly acts to enhance melt fraction changes caused by compaction (Fig. 8q) but, at later times, reactive flow opposes melt fraction changes caused by compaction, generally increasing melt fraction in the lower half of the sill, and decreasing melt fraction in the upper half (Fig. 8q–t). The contributions of compaction and reactive flow to melt fraction change are of similar magnitude. Indeed, it can be observed in Fig. 8q–t that melt fraction decrease in the lower part of the sill is dominated by cooling, because the contributions of compaction and reactive flow almost exactly cancel each other; conversely, melt fraction in the upper part of the sill only decreases at late times because cooling and reactive flow together are large enough to overwhelm compaction (Fig. 8s).

The complex interplay between cooling, compaction and reactive flow is reflected in the variation of melt fraction with depth through the sill, which, at early to mid-times, differs significantly from the simple, upwards increasing trend in melt fraction observed in compaction-dominated systems (Fig. 8b). At later times, the melt fraction trend does resemble a compaction-dominated system (Fig. 8c), but the decrease in melt fraction at the base of the sill is caused primarily by cooling, rather than compaction (Fig. 8r). Compaction acts most significantly to reduce melt fraction at the centre of the sill (Fig. 8r–t), rather than at the base as predicted by pure compaction models (cf. Fig. 5a–d).

The contributions of compaction and reactive flow to composition change are also complex. At early times in both the upper and lower parts of the sill, compaction locally increases, and decreases, SiO₂ content (Fig. 8v); however, at later times, compaction broadly acts to increase SiO₂ content in the upper half of the sill, and decrease SiO₂ content in the lower half of the sill. The upper part of the sill therefore becomes more evolved and the lower part becomes more refractory, consistent with the column model results reported in the previous section (Fig. 5j–l).

Reactive flow generally opposes composition changes caused by compaction, increasing the bulk SiO₂ content in the lower half of the sill, and decreasing the bulk SiO₂ content in the upper half (Fig. 8v–y). The contribution of compaction slightly exceeds that of reactive flow, such that the base of the sill becomes more refractory and the top becomes more evolved.

The contribution of reactive flow to melt fraction and composition change can be explained by considering the gradients in melt and solid composition (Fig. 8k–o). The solid composition remains close to uniform, so there is negligible reactive flow of the solid phase even though compaction causes solid to be transported downwards (equation 9). However, in the lower half of the sill, melt migrates upwards through a gradient in melt composition: the most evolved melt is located at the base of the sill where the

temperature is lowest, and the least evolved melt is located near the centre of the sill where the temperature is highest (Fig. 8l–n). Consequently, reactive flow delivers evolved melt from the base to the overlying mush, causing the melt fraction to increase and the composition to become more evolved, consistent with the column results reported in the previous section (Fig. 7j–l).

In the upper half of the sill, melt percolates upwards through a reversed gradient in melt composition, with the most evolved melt located at the top of the sill where the temperature is lowest (Fig. 8l–n). Consequently, reactive flow delivers less evolved melt to the overlying mush, causing the melt fraction to decrease and the bulk composition to become less evolved.

Discussion

Compaction and reactive flow in crustal magma reservoirs

The identification of the terms in equations (8) and (9) representing melting/cooling, compaction and reactive flow for our example chemical model (Fig. 3) allows their relative contributions to melt fraction and composition changes in mush reservoirs to be quantified (equations 14 and 15). Note that the term in equation (8) describing phase change by melting/cooling also includes phase change caused by change in pressure, but here we have confined ourselves to the isobaric case.

We find that the contributions of compaction and reactive flow to melt fraction change are similar in magnitude, but reactive flow typically opposes melt fraction changes caused by compaction or decompaction; i.e. if compaction acts to cause melt fraction decrease, then reactive flow acts to cause melt fraction increase, and vice versa (Fig. 8). In general, for our example chemical model, the terms defining compaction and melt reactive flow in equations 8 and 9 show that compaction and reactive flow cause opposing melt fraction changes when more evolved melt leaves a compacting region; similarly, decompaction and reactive melt flow cause opposing melt fraction changes when less evolved melt enters a decompacting region. For the case of buoyancy-driven compaction driving vertical flow in a basaltic system, compaction and reactive flow cause opposing melt fraction changes when compaction occurs in a composition gradient that decreases upwards; similarly, decompaction and reactive flow cause opposing melt fraction changes when decompaction occurs in a composition gradient that increases upwards. An upwards decrease in composition (to become more refractory) corresponds to an upwards increase in temperature (equation 13); conversely, an upwards increase in composition (to become more evolved) corresponds to an upwards decrease in temperature.

Whether reactive flow in a crustal magma reservoir causes the melt fraction locally to increase, or decrease, depends on the composition of the melt percolating into an REV of partially molten rock, which, in turn, reflects the temperature gradient. Reactive flow of melt into an REV increases the local melt fraction if more evolved melt enters a hotter REV containing less evolved interstitial melt, causing dissolution of one (or more) solid mineral(s) such that the final amount of melt is greater than the sum of the initial and the added melt (e.g. case C in Fig. 1). This corresponds to chemical melting. Conversely, if less evolved melt enters a cooler REV within which the interstitial melt is more evolved, precipitation of solid mineral(s) occurs such that the final amount of melt is less than the sum of the initial and the added melt. This corresponds to chemical crystallisation and the ‘reactive crystallisation’ of Collier and Kelemen (2010).

In crustal magma reservoirs, reactive flow therefore typically opposes melt fraction changes caused by compaction or decompression, because compaction typically occurs near the base of the reservoir where numerous thermal models have shown that the temperature typically increases upwards (e.g. [Annen et al., 2005](#); [Hodge, 1974](#); [Jackson et al., 2018](#); [Karakas et al., 2017](#); [Petford and Gallagher, 2001](#); [Solano et al., 2012](#)). Conversely, decompression typically occurs near the top of the reservoir where thermal models show that the temperature typically decreases upwards.

Evidence for compaction and reactive flow in the rock record

The view that compaction is the principal mechanism driving melt segregation and differentiation in low melt fraction crustal mush reservoirs has motivated research to identify evidence for compaction in the rock record (e.g. [Elliott et al., 1997](#); [Holness, 2018](#); [Holness et al., 2005, 2017](#); [Rudge, 2018](#)). There has been far less attention paid to reactive flow, most likely because its role in driving melt fraction change and differentiation in magma reservoirs within the continental crust has not been widely recognized. Here we have shown that reactive flow is just as important as compaction in driving changes in melt fraction and bulk composition, suggesting that we should be equally focused on identifying evidence for reactive flow.

We find that reactive flow typically causes melt fraction to increase in upwards-warming regions of a mush reservoir where compaction is causing melt fraction to decrease, which has important consequences for mush textural evolution. The contribution of reactive flow in driving melt fraction and composition change means that very small melt fractions cannot be obtained by compaction alone, even in refractory parts of a mush reservoir. This is demonstrated in the sill intrusion results: the bulk composition after 80 y in the lower half of the sill is already refractory, having almost reached the SiO_2 content of the solid phase (47%; Fig. 8n), yet the melt fraction in this part of the sill is of order 0.1–0.2 (Fig. 8d). The mixing curves show that pure compaction could produce such refractory bulk composition only by driving the melt fraction to such low values that microstructural evidence for compaction would be inevitable (dashed lines in Fig. 6). Melt fraction decrease is, in fact, almost entirely dominated by cooling in this part of the sill; melt fraction changes by compaction and reactive flow almost exactly counterbalance (Figure 9). Reactive flow opposes melt fraction decreases caused by compaction, causing chemical melting (dissolution) of the solid phase and thus maintaining melt channels for ongoing loss of melt.

These findings have important implications for how we interpret the rock record. Because compaction does not drive melt fraction to very small values even in refractory parts of the system, there will be little or no microstructural evidence for viscous compaction at low melt fraction ([Holness, 2018](#)). Rather, melt loss may be accompanied by evidence of chemical melting (dissolution) of the solid phase due to reactive flow. The role of reactive flow in driving melt fraction change can explain the relative lack of evidence for widespread compaction in the igneous rock record. Even relatively small contributions of reactive flow could be important in reducing or removing the evidence for compaction. The apparently anomalous lack of compaction in mush reservoirs as compared to other natural and engineered systems is also explained by the contribution of reactive flow: in systems such as soils and filter presses, where there is little or no component exchange between solid and liquid phases, reactive flow does not play a significant role.

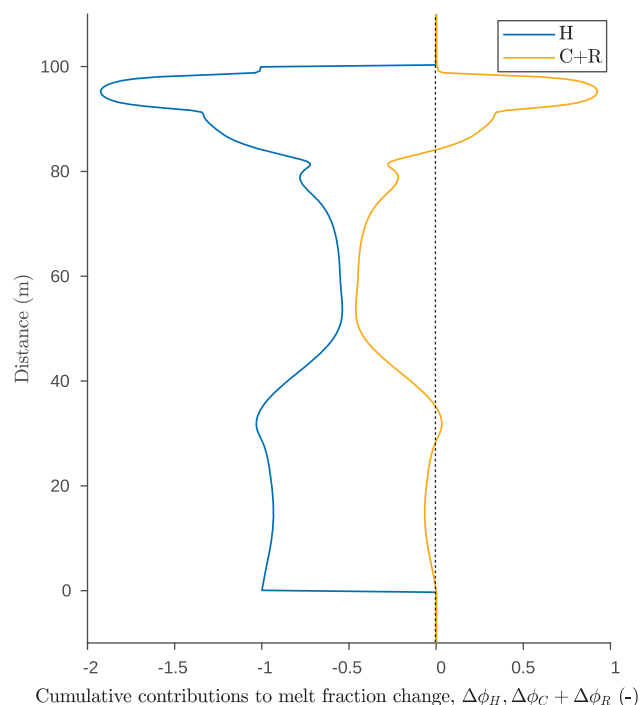


Fig. 9. Cumulative contributions to local melt fraction change in the sill model, showing the combined contributions of compaction and reactive flow (C + R) compared to heating and cooling (H). Data extracted from Fig. 8t.

Reactive flow in upwards-warming, compacting regions of a crustal mush reservoir causes dissolution of the solid phase. Figure 10 shows three examples of rock textures that may have been formed by varying degrees of melt loss during reactive flow. Samples in Fig. 10A–C are xenoliths from St. Vincent, East Caribbean volcanic arc, interpreted to represent fragments of mush reservoir entrained in carrier magma during volcanic eruptions from depths of 5–15 km in the sub-volcanic crust ([Melekhova et al., 2019](#)). The largest melt (glass) fraction is observed in Fig. 10A, where euhedral plagioclase, clinopyroxene and magnetite and rounded olivine are surrounded by glass, which represents quenched melt present in the xenolith at the time of incorporation into the carrier magma. Subsequent magma ascent likely led to vesiculation of the matrix melt; however, the original melt distribution is preserved. The mineralogy of Fig. 10B is broadly similar to Fig. 10A, but melt fraction is lower, and clinopyroxene and plagioclase show rounded crystal faces due to dissolution, consistent with a larger contribution from reactive flow. In Fig. 10C, the mineralogy is exclusively plagioclase, with a small volume fraction of interstitial glass. Almost all melt has been lost from this sample, accompanied by extensive plagioclase dissolution consistent with reactive flow. The plagioclase grains themselves show no evidence of internal deformation, such as kinking or bending of twin planes, which argues against compaction as the mechanism that reduced melt fraction to such a small value in this sample. However, grains are rounded, indicative of dissolution.

Reactive flow in upwards-cooling, decompacting regions of a crustal mush reservoir causes interstitial melt to be replaced by new crystalline material, which will either accrete to the rims of existing crystals, if it has the same composition, or precipitate as a new mineral phase, occluding the pore space. This process may account for the widespread tendency of interstitial mineral

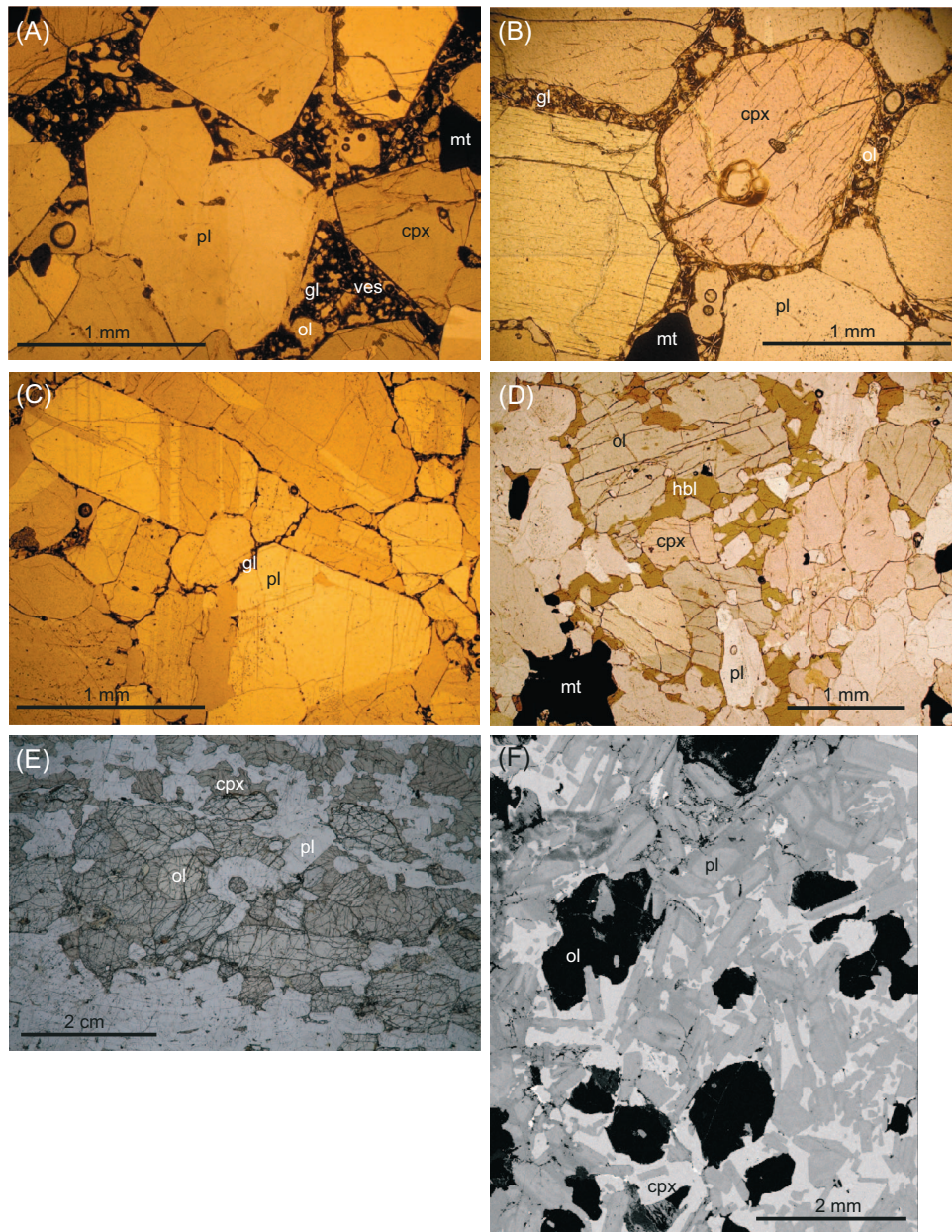


Fig. 10. Textural features of crystal mushes showing possible consequences of reactive flow. Panels (A) to (C) show the presence of partial melt; panels (D) to (F) show mono-mineralic replacement of original melt-filled pore space. (A) Photomicrograph (plane-polarised light) of olivine gabbro xenolith (sample VS34) from St. Vincent, East Caribbean volcanic arc. Euhedral plagioclase (pl), clinopyroxene (cpx) and magnetite (mt) and rounded olivine (ol) are surrounded by brown glass (gl) with vesicles (ves). The glass represents quenched melt present in the xenolith at the time of incorporation into the carrier magma. Vesiculation probably occurred on eruption, although some vesicles may have been present at the time of entrainment. Sample from Wallibou River valley (N13deg18'46.1" W61deg15'52.4"). For more information on related samples of xenoliths from St. Vincent, see [Tollan et al. \(2012\)](#). (B) Photomicrograph (plane-polarised light) of gabbro xenolith (VS11) from St. Vincent. Texture and mineralogy are broadly similar to (A), but melt fraction is lower and clinopyroxene and plagioclase show rounded crystal faces due to dissolution into melt. Interstitial glass contains abundant plagioclase microlites and vesicles formed upon decompression of the xenolith to the surface. The large clinopyroxene in the centre of the photograph contains a vesicular melt inclusion. Sample from Rabacca Dry Driver gully (N13deg19'41.6" W61deg10'23.1"). (C) Photomicrograph (plane-polarised light) of an anorthosite xenolith (VS40) from St. Vincent. Mineralogy is exclusively plagioclase with low volume fraction of interstitial vesicular glass. Locally the glass has been plucked from the slide, but remnants are visible as dark rinds decorating the rounded plagioclase grain boundaries. Almost all melt has been expelled from this sample, a process accompanied by plagioclase dissolution. Sample location as in (A). (D) Photomicrograph (plane-polarised light) of a hornblende gabbro xenolith (VS22) from St. Vincent showing pale pink-green pleochroic clinopyroxene, colourless plagioclase and opaque magnetite grains with interstitial khaki hornblende (hbl). The texture of hornblende reflects the original melt distribution, such as that shown in (B). Note the rounded and resorbed edges of clinopyroxene and plagioclase grains in contact with hornblende, testifying to reaction between interstitial melt and original mineral grains in the mush. This textural relationship indicates that the interstitial melt was hydrous. Sample is from Jacob's Well (N13deg07'31.0 W61deg10'35.1). (E) Photomicrograph (plane-polarised light) of troctolitic gabbro from Rum layered intrusion, UK showing olivine and plagioclase grains with interstitial clinopyroxene. The texture of the clinopyroxene reflects the original distribution of interstitial melt in an olivine + plagioclase mush. Note the rounded outline of olivine grains indicating reaction with the melt. Sample (11JL17B) is taken from Unit 9 of the intrusion (N56deg59'06.55 W61deg16'58.92). For further details of related samples from this locality, see [Leuthold et al. \(2014\)](#). (F) Calcium X-ray map of poikilitic gabbro from Rum layered intrusion. Darkest grey is olivine; mid-grey is plagioclase, showing concentric, normal zoning from higher to lower anorthite content; lightest grey is clinopyroxene, showing interstitial texture. Sample (D4) is taken from Unit 3 adjacent to a picrite dyke at N56deg59'3.69 W61deg16'09.07. For further details of this locality, see [Sides \(2008\)](#).]

phases to display textures that mimic those of interstitial melt, even though their monomineralic character is inconsistent with the chemical composition of any plausible melt phase. Figure 10 shows three examples of textures that may have been formed by this process. The first (Fig. 10D) shows interstitial amphibole in a gabbro xenolith, again from St. Vincent. The distribution of the amphibole mimics that of the glass in Fig. 10A and B, testifying to amphibole formation by reaction of hydrous, silicic melt with plagioclase and clinopyroxene. The amphibole is compositionally distinct from the original melt, indicating that it is the result of a reactive flow process, rather than simple cooling and freezing-in of the melt. Reaction product melt undoubtedly left the rock during amphibole formation.

The second two examples (Fig. 10E,F) show interstitial clinopyroxene in a troctolite (E) and poikilitic gabbro (F); both samples are from the Rum Layered Intrusion (UK). Here the evidence of clinopyroxene formation by dissolution–reprecipitation reactions with minerals in the mush (olivine + plagioclase) is more limited, probably because the three phases are in a cotectic rather than peritectic relationship. In Fig. 10F it is apparent from the Ca X-ray map that plagioclase grains have sodic rims in contact with clinopyroxene, suggesting that both minerals were precipitated during the reactive flow process. This texture is not evident in plane-polarised light (Fig. 10E). In both of these examples, however, we argue that the interstitial mineral phase(s) does not correspond to the melt composition; rather, the mineral(s) was deposited during reactive flow of a chemically different melt phase.

Interstitial mineral phases that pick out original melt distribution are a common feature of cumulate igneous rocks, as originally noted by Wager *et al.* (1960) when they made the distinction between an orthocumulate where ‘the products of crystallization of the intercumulus liquid, necessarily [have] the composition of the contemporary magma’ and mesocumulate, adcumulate and heteradcumulate rocks, where they demonstrably do not. Wager *et al.* (1960) concluded that ‘some new mechanism...has operated in the formation of the poikilitic crystals and we suggest that they have grown between the cumulus crystals, while they were at or near the top of the pile, as a result of diffusion of material to relatively rare crystal nuclei developed in the intercumulus liquid’. Since this process was suggested, experimental data have shown that component (Fickian) diffusion is limited even within a melt phase. As we show below, diffusive component transport over the metres to tens of meters scale proposed by Wager *et al.* (1960) is slow compared to advective transport by melt flow. Moreover, the timescales for diffusive transport would be different for different components, leading to diffusive fractionation of a type not observed in intercumulus mineral phases. We suggest that the ‘new mechanism’ sought by Wager *et al.* (1960) is percolative reactive flow, which causes components to be transported by advection through the cumulate mush.

Compaction will play a greater role in driving melt fraction loss in situations where reactive flow is relatively less important. This could occur in parts of a magma reservoir where there are relatively shallow composition gradients, corresponding to shallow temperature gradients or regions where melt composition is relatively insensitive to temperature, such that melt does not migrate through a steep gradient in composition. Compaction will also dominate if melt flow is rapid compared to chemical reaction such that local chemical equilibrium is not established. Compaction and reactive flow combine to drive melt fraction and composition change when compaction occurs in a temperature gradient that decreases upwards (or decompaction occurs in a

temperature gradient that increases upward) and these may also be special cases where evidence for compaction is preserved in the rock record.

Equation 8 shows that, in general, melt fraction and bulk composition change depends on spatial gradients in both the melt and solid composition. However, for both phase diagrams used here (Figs 1 and 3), the solid composition is constant so there is no gradient in solid composition and hence no contribution from flow of the solid phase. For all realistic phase diagrams we have tested, including phase diagrams exhibiting solid solution, the contribution of reactive flow is qualitatively similar to that shown here.

Assumption of local thermal and chemical equilibrium

We have assumed here that thermal and chemical equilibrium is maintained at the scale of an REV, which requires that the diffusive transport of heat and components between melt and solid phases is rapid compared to the advective transport of heat and components by flow. The relative rates of diffusive and advective transport are characterised by the Peclet number ($Pe = uL/D$) where u is a characteristic velocity, L is a characteristic length scale and D is a characteristic diffusivity. Smaller Pe corresponds to more significant transport by diffusion and vice versa. The assumption of local thermal equilibrium is common in subsurface reservoir models, given the relatively high thermal diffusivity (of order 10^{-7} m²/s) of silicate melt and crystals [e.g. Turcotte and Schubert, 2002]. However, the assumption of local chemical equilibrium is restricted by the relatively low component diffusivity, especially in the solid phase (of order 10^{-15} m²/s) (e.g. Zhang *et al.*, 2010).

In our chemical model (Fig. 3), the solid phase composition is close to uniform, so component diffusion in the crystals—at least of the SiO₂ component that we model—is not required to maintain local chemical equilibrium. Rather, the approach to equilibrium is governed by the rate of diffusive component transport through the melt phase (of order 10^{-11} m²/s; (Zhang *et al.*, 2010)). The characteristic length scale here is the crystal size; we require equilibration to occur as melt flows past a crystal. A suitable estimate of the characteristic velocity is provided by the Darcy separation velocity (e.g. Jackson *et al.*, 2005)

$$u_D = k \frac{(\rho_s - \rho_l)g}{\phi \mu_m} \quad (1)$$

Assuming a typical melt fraction of 0.1 (see, for example, Fig. 8), a component diffusivity of order 10^{-11} m²/s and substituting values from Table 2, we obtain Pe of order 10^{-3} to 1 depending on melt viscosity, suggesting that the assumption of local chemical equilibrium is marginal for lower values of melt viscosity and for higher values of melt fraction. The further the system deviates from local chemical equilibrium, the less important the contribution of reactive flow. Local equilibrium is favored by low melt fraction and high melt viscosity, because flow velocities are lower. Many crystals preserved in, or erupted from, crustal magmatic systems are zoned (e.g. Fig. 10F), suggesting local equilibrium is not maintained as the crystal experiences changing melt composition, bulk composition, temperature and pressure. The simple chemical model used here cannot capture crystal zonation. Accounting for reaction kinetics and chemical disequilibrium at the REV scale is a priority for the next development of our models [e.g. Keller and Katz, 2016a; Rudge *et al.*, 2011].

As mentioned in section 3, we neglect component diffusion at the macroscale (equation 2), based on the assumption that diffusion is slow compared to advection over lengthscales greater than a few tens of crystals. We argue that diffusion would be too slow compared to advection to explain the cumulate igneous textures originally noted by Wager *et al.* (1960). Calculating Pe for a lengthscale of $L = 10$ m as suggested for diffusive transfer by Wager *et al.* (1960), we obtain values of order 6 to 6×10^3 depending on melt viscosity, suggesting that our decision to neglect component diffusion at the macroscale is reasonable.

Volatiles and more complex chemical modelling

The binary phase diagram that we have adopted (Fig. 3) captures much of the important behaviour of natural rock systems in a sufficiently simple way to be implemented in our numerical models. However, natural, multicomponent petrological systems show additional complexity that is not readily observed in a simple binary. First, natural systems show solid solution among a large number of mineral phases. This modifies the form of the solidus curves. The extent of solid solutions changes with decreasing temperature such that during sub-solidus cooling, phase compositions will change. Second, melting behaviour in natural systems is rarely eutectic; peritectic melting reactions of the form $A=B+L$ (where A and B are solids and L is liquid) are common. In such cases, melt production also involves solid production, which will affect porosity. Third, natural magmatic phase relations are pressure dependent, which will lead to depth-dependent behaviour in vertically extensive magmatic systems. The Clapeyron slopes (dP/dT) of mineral-melt equilibria depend primarily on the relative volumes of melts and minerals. In dry systems, mineral melting temperatures increase with increasing pressure (positive dP/dT). In hydrous systems, mineral stability curves show more complex behaviour, including negative dP/dT in water-saturated melts. Finally, the presence of water (and other volatiles) also affects the dynamics of the system. Volatile solubilities are another pressure-dependent feature of natural magmas. As melts decompress (due to ascent) or crystallise (due to cooling) they may exsolve a free volatile phase; these processes are termed 'first' and 'second' boiling, respectively. The presence of a volatile phase will impact greatly on the flow of reactive melt through a mush reservoir (e.g. Huber and Parmigiani, 2017), as well as participating itself in chemical reactions. The strongly pressure-dependent solubility of volatiles means that their explicit inclusion will confer an extra depth-related dimension to reactive flow style and consequences. For these reasons, including a volatile phase is an important next development of our models.

For a complete description of reactive porous flow in natural crustal magmatic systems all of the above effects need to be incorporated into the underlying phase behaviour. This must be done without the requirement of greatly increased computing demands, such as those required to perform free energy minimisations at each node in the system. We consider that the phenomena listed above will affect hot zone development, but will not change fundamentally our findings. However, this claim needs to be tested with further models.

Conclusions

Reading of binary phase diagrams from a purely chemical perspective is well-established and straightforward, but viewing such diagrams through the prism of realistic physical processes reveals an unexpected wealth of complexity, much of which has

important consequences for chemical differentiation in high-crystallinity magmatic systems.

Changes in melt fraction and local bulk composition in crustal magma reservoirs can be quantified in terms of distinct contributions from different processes. Two of those considered here, heating/cooling and compaction, have been widely invoked in previous studies. Phase change by heating/cooling alone does not change bulk composition, so cannot lead to chemical differentiation. Compaction involves no phase change, as there is no exchange of components between solid and melt phases; hence, changes in bulk composition caused by compaction lie on simple linear mixing curves. Consequently, compaction can only produce very refractory compositions by driving melt fraction to very small values. Conversely, decompression can only produce very evolved compositions by driving melt fraction to very high values.

Reactive flow is the transport of components by flow causing chemical reaction and phase change. Reactive flow leads to changes in bulk composition that cause 'chemical' melting and crystallisation, distinct from the changes in enthalpy (or, as is more commonly used, temperature) that cause 'thermal' melting and crystallisation. We report here a mathematical description of the contribution that reactive flow makes to changes in melt fraction and local bulk composition, and use this to quantify the contributions of heating/cooling, compaction and reactive flow to melt fraction and bulk composition change in several scenarios relevant to crustal magma reservoirs.

We find that the contributions of compaction and reactive flow to melt fraction and composition change in crustal magma reservoirs are similar in magnitude, but reactive flow typically opposes melt fraction and composition changes caused by compaction or decompression; i.e. if compaction acts to cause melt fraction decrease and the bulk composition to become more refractory, then reactive flow acts to cause melt fraction increase and the bulk composition to become more evolved, and vice versa. Compaction and reactive flow cause opposing melt fraction and composition changes when compaction occurs in a temperature gradient that increases upwards at, for example, the base of an intrusion or mush reservoir. This is because evolved melt from the deeper, cooler part of the reservoir migrates upwards, causing the bulk composition of the hotter overlying mush to become more evolved and the melt fraction to increase as a consequence. Similarly, decompression and reactive flow cause opposing melt fraction and composition changes when decompression occurs in a temperature gradient that decreases upwards at, for example, the top of an intrusion or mush reservoir. This is because less evolved melt from the deeper, warmer part of the system migrates upwards, causing the bulk composition of the cooler overlying mush to become less evolved and the melt fraction to decrease as a consequence.

The contribution of reactive flow in driving melt fraction and composition change means that very small melt fractions are not required to generate very refractory compositions in a mush reservoir, consistent with the dearth of microstructural evidence for widespread compaction at low melt fraction in the igneous rock record. The apparently anomalous lack of compaction in mush reservoirs, as compared to other natural and engineered systems, is explained by the contribution of reactive flow. Reactive flow means that melt loss in compacting regions of a mush may be accompanied by evidence for mineral dissolution, which—perhaps counter-intuitively—facilitates melt fraction loss by preserving connected melt flow paths through the mush pore space. Reactive flow can also explain why interstitial mineral phases display textures that mimic those of interstitial melt. Interpreting

chemical differentiation and the evolution of rock microstructure in crustal mush reservoirs exclusively via the heating/cooling and compaction is misleading.

A Decomposing change in melt fraction and bulk composition

A multiphase, multicomponent system

For a system that consists of N phases and M components, the continuity equation can be written as

$$\frac{\partial \phi_i}{\partial t} + \nabla \cdot \phi_i \mathbf{u}_i = \Gamma_i, \quad i = 1, 2, \dots, N \quad (A1)$$

$$\sum_{i=1}^N \Gamma_i = 0, \quad (A2)$$

where ϕ_i and $\mathbf{u}_i$ and Γ_i are the melt fraction, velocity and the volume exchange rate of the i th phase, respectively. The component transport equation reads

$$\frac{\partial \phi_i C_i^j}{\partial t} + \nabla \cdot \phi_i \mathbf{u}_i C_i^j = \Gamma_i^j, \quad j = 1, 2, \dots, M \quad (A3)$$

$$\sum_{i=1}^N \Gamma_i^j = 0 \quad (A4)$$

$$\sum_{j=1}^M C_i^j = 1, \quad (A5)$$

where C_i^j is the j th component in i th phase and Γ_i^j is the corresponding component exchange rate. Using the chain rule on equation A3 and following the same procedure as section 3 leads to

$$\phi_i \frac{\partial C_i^j}{\partial t} + \phi_i \mathbf{u}_i \cdot \nabla C_i^j + \Gamma_i C_i^j - \Gamma_i^j = 0. \quad (A6)$$

Summing equation A6 by index i eliminates Γ_i^j , which results in

$$\sum_{i=1}^N \phi_i \frac{\partial C_i^j}{\partial t} + \phi_i \mathbf{u}_i \cdot \nabla C_i^j + \Gamma_i C_i^j = 0. \quad (A7)$$

When $M < N$, the volume exchange rate Γ_i cannot be uniquely determined. When $M > N$, an additional constraint for the relationships between components C_i^j needs to be obtained from the underlying chemical reactions, whereby the system can be reduced to the same as with N components. In this situation, equation A7 can be solved. Noticing that there are only $N - 1$ and $M - 1$ independent Γ_i and C_i^j for each i , respectively, we define

$$\mathbf{C} = \begin{pmatrix} C_1^1 & C_2^1 & \dots & C_{N-1}^1 \\ C_1^2 & C_2^2 & \dots & C_{N-1}^2 \\ \dots & \dots & \dots & \dots \\ C_1^{N-1} & C_2^{N-1} & \dots & C_{N-1}^{N-1} \end{pmatrix} \quad \frac{\partial \Phi}{\partial t} = \begin{pmatrix} \frac{\partial \phi_1}{\partial t} \\ \frac{\partial \phi_2}{\partial t} \\ \dots \\ \frac{\partial \phi_{N-1}}{\partial t} \end{pmatrix} \quad \gamma = \begin{pmatrix} \nabla \cdot \phi_1 \mathbf{u}_1 \\ \nabla \cdot \phi_2 \mathbf{u}_2 \\ \dots \\ \nabla \cdot \phi_{N-1} \mathbf{u}_{N-1} \end{pmatrix} \quad (A8a)$$

$$\alpha = \begin{pmatrix} \sum_{i=1}^{N-1} \phi_i \frac{\partial C_i^1}{\partial t} \\ \sum_{i=1}^{N-1} \phi_i \frac{\partial C_i^2}{\partial t} \\ \dots \\ \sum_{i=1}^{N-1} \phi_i \frac{\partial C_i^{N-1}}{\partial t} \end{pmatrix} \quad \beta = \begin{pmatrix} \sum_{i=1}^{N-1} \phi_i \mathbf{u}_i \cdot \nabla C_i^1 \\ \sum_{i=1}^{N-1} \phi_i \mathbf{u}_i \cdot \nabla C_i^2 \\ \dots \\ \sum_{i=1}^{N-1} \phi_i \mathbf{u}_i \cdot \nabla C_i^{N-1} \end{pmatrix}. \quad (A8b)$$

The total change in melt fraction across all phases Φ can be decomposed into

$$\frac{\partial \Phi}{\partial t} = -\gamma - \mathbf{C}^{-1} \alpha - \mathbf{C}^{-1} \beta, \quad (A9)$$

where the contributions from compaction, heating and reactive flow are given by the first, second and third terms, respectively. Following the same approach for the total change in bulk composition we obtain

$$\begin{pmatrix} \frac{\partial \bar{C}^1}{\partial t} \\ \frac{\partial \bar{C}^2}{\partial t} \\ \dots \\ \frac{\partial \bar{C}^{N-1}}{\partial t} \end{pmatrix} = -\mathbf{C} \gamma - \beta, \quad (A10)$$

where $\bar{C}^j$ is the bulk composition of the j th component, which is defined as

$$\bar{C}^j = \sum_{i=1}^N \phi_i C_i^j. \quad (A11)$$

Including variable density

We begin with the mass conservation (continuity) equation with variable density

$$\frac{\partial \rho_l \phi}{\partial t} + \nabla \cdot \rho_l \phi \mathbf{u}_l = \Gamma \quad (A12a)$$

$$\frac{\partial \rho_s (1 - \phi)}{\partial t} + \nabla \cdot \rho_s (1 - \phi) \mathbf{u}_s = -\Gamma. \quad (A12b)$$

The source term Γ on the right hand side represents the rate of mass transfer from solid to liquid. The equivalent expression for component conservation, neglecting component diffusion, is given by

$$\frac{\partial \rho_l \phi C_l}{\partial t} + \nabla \cdot \rho_l \phi C_l \mathbf{u}_l = \Gamma_c \quad (A13a)$$

$$\frac{\partial \rho_s (1 - \phi) C_s}{\partial t} + \nabla \cdot \rho_s (1 - \phi) C_s \mathbf{u}_s = -\Gamma_c, \quad (A13b)$$

where C_l and C_s are liquid and solid composition, respectively. Γ_c is the rate of component mass transfer from solid to liquid. Applying the chain rule to equation A13(a), we have

$$C_l \frac{\partial \rho_l \phi}{\partial t} + \rho_l \phi \frac{\partial C_l}{\partial t} + C_l \nabla \cdot \rho_l \phi \mathbf{u}_l + \rho_l \phi \mathbf{u}_l \cdot \nabla C_l = \Gamma_c \quad (A14)$$

$$\Leftrightarrow C_l \left(\frac{\partial \rho_l \phi}{\partial t} + \nabla \cdot \rho_l \phi \mathbf{u}_l - \Gamma \right) + \rho_l \phi \frac{\partial C_l}{\partial t} + \rho_l \phi \mathbf{u}_l \cdot \nabla C_l + \Gamma C_l - \Gamma_c = 0 \quad (A15)$$

$$\Leftrightarrow \rho_l \phi \frac{\partial C_l}{\partial t} + \rho_l \phi \mathbf{u}_l \cdot \nabla C_l + \Gamma C_l - \Gamma_c = 0. \quad (A16)$$

The same procedure applied to equation A13(b) yields

$$\rho_s (1 - \phi) \frac{\partial C_s}{\partial t} + \rho_s (1 - \phi) \mathbf{u}_s \cdot \nabla C_s - \Gamma C_s + \Gamma_c = 0. \quad (A17)$$

Adding equation A16 and A17 to eliminate Γ_c leads to

$$\rho_l \phi \frac{\partial C_l}{\partial t} + \rho_s (1 - \phi) \frac{\partial C_s}{\partial t} + \rho_s (1 - \phi) \mathbf{u}_s \cdot \nabla C_s + \rho_l \phi \mathbf{u}_l \cdot \nabla C_l + \Gamma (C_l - C_s) = 0. \quad (\text{A18})$$

Re-arranging for Γ and substitution into equation A12(a), the rate of change of melt fraction can be written as

$$\frac{\partial \rho_l \phi}{\partial t} = - \underbrace{\frac{1}{C_l - C_s} [\rho_s (1 - \phi) \mathbf{u}_s \cdot \nabla C_s + \rho_l \phi \mathbf{u}_l \cdot \nabla C_l]}_{\text{reactive flow}} - \underbrace{\nabla \cdot \rho_l \phi \mathbf{u}_l}_{\text{compaction}} - \underbrace{\frac{1}{C_l - C_s} [\rho_l \phi \frac{\partial C_l}{\partial t} + \rho_s (1 - \phi) \frac{\partial C_s}{\partial t}]}_{\text{heating/cooling}}. \quad (\text{A19})$$

Thus an extra term $-\frac{\partial \rho_l}{\partial t}$ appears when density changes are included. Since this term is independent of velocity, we suggest that density changes caused by thermal expansion or chemical reaction should be included in the heating/cooling term. Thus we have

$$\frac{\partial \phi}{\partial t} = - \underbrace{\frac{1}{(C_l - C_s)} [\frac{\rho_s}{\rho_l} (1 - \phi) \mathbf{u}_s \cdot \nabla C_s + \phi \mathbf{u}_l \cdot \nabla C_l]}_{\text{reactive flow}} - \underbrace{\frac{1}{\rho_l} \nabla \cdot \rho_l \phi \mathbf{u}_l}_{\text{compaction}} - \underbrace{\frac{1}{(C_l - C_s)} [\phi \frac{\partial C_l}{\partial t} + \frac{\rho_s}{\rho_l} (1 - \phi) \frac{\partial C_s}{\partial t}]}_{\text{heating/cooling}} - \frac{\partial \rho_l}{\rho_l \partial t}. \quad (\text{A20})$$

B Construction of the binary phase diagrams

Here we report the construction of the enthalpy-based binary eutectic phase diagrams shown in Figs 1 and 3. The phase diagram predicts the melt fraction (ϕ), solid composition (C_s) and liquid composition (C_l) for a given enthalpy per unit mass (H) and bulk composition ($\bar{C}$).

Data for the binary diopside-anorthite phase diagram (Fig. 1) were taken from [Ussler and Glazner \(1992\)](#). Data to describe the melting behaviour of basalt (Fig. 3) were obtained from melting experiments on natural samples. Silicate melts are thermodynamically complex solutions whose description requires complex models, both to capture the inherent non-ideality of the melt, but also the saturation surfaces of the many different minerals (and their equally complex solutions) with which such liquids may be in equilibrium. There are a number of thermodynamic models to describe the phase equilibria of silicate melts, of which the MELTS family of algorithms is the most widely used (e.g. [Ghiorso and Sack, 1995](#)). We could couple MELTS (or a similar model) with our flow and transport code to compute the phase equilibria as a function of temperature, pressure and bulk composition. This is a robust approach, but extremely numerically intensive as a phase equilibrium calculation is required at every discretized location in space (N_x) and time (N_t): the total number of calculations is $N_x \times N_t$. Here, N_x is typically of order 10^3 , and N_t is of order 10^6 , so a billion phase equilibria calculations would be necessary even in the small, one-dimensional numerical models reported here.

An alternative approach, adopted previously by [Jackson et al. \(2018\)](#), is to capture the phase behaviour using a simple binary

eutectic phase diagram, tuned to match the experimental data. Here we refine their approach by considering both solid and liquid composition, and calibrating the phase diagram topology using selected phase equilibrium experiments on hydrous magmas of the type intruded into crustal mush reservoirs. Although such experimental data are very extensive, we have focussed our attention on experimental studies of basalt with an initial water content of ca. 2.5 wt %, over a sufficiently large temperature range that residual liquids extend to the solidus. By this means, we reduce the potential for inter-laboratory variability. We also limit ourselves to experiments where the composition and mass fraction of melt have been determined, enabling direct calculation of the solid composition from the bulk composition of the experimental charge. Note that calculation of bulk solid composition via mass balance confers much greater uncertainty in composition (or order 1–5 wt% SiO₂) than for the liquid (glass) analysis itself (of order 0.2–0.8 wt% SiO₂), due to propagation of errors through all analysed phases and through the mass balance.

The selected dataset of 42 experiments is given in Table 3, showing the range in pressure, temperature and melt composition. Three of the experiments used basalt; one used dacite. Although the basalt starting material was always water-undersaturated (2.5 ± 0.5 wt % H₂O), the melts approach water saturation as crystallisation proceeds, because the crystallising assemblage is H₂O-poor. In a true binary eutectic, the phase diagram topology is independent of the starting material composition, including H₂O. Because of the considerable effect that dissolved H₂O has on phase relations, especially the liquidus temperature, we need to take this into account when constructing our phase diagram. To this end, we include the dacite experimental dataset of [Blatter et al. \(2013\)](#) alongside the basalt datasets. Although [Blatter et al. \(2013\)](#) report a large number of experiments, we use only those with 9 wt% H₂O in the starting material, as this most closely replicates the H₂O content in residual melts from the basalt experiments at ca. 900 °C, corresponding to the highest temperature experiment of [Blatter et al. \(2013\)](#). The seamless integration of data from the basalt and dacite experiments satisfies the assumption of binary eutectic behaviour.

Our resulting phase diagram (Fig. 3) is characterised by a convex upwards liquidus that extends from basalt (ca. 48 wt % SiO₂) to rhyolite (ca. 72 wt% SiO₂). The solidus data, albeit more scattered than the liquidus data, have almost constant composition (i.e. they plot on a vertical line in Fig. 3A). This behaviour arises because the liquidus mineral assemblage changes progressively with temperature, from olivine and pyroxene-dominated assemblages at high temperature, to plagioclase and amphibole at lower temperature, therefore maintaining an overall near-constant SiO₂ content for the solidus, until quartz saturates at low temperature, which leads to a sharp bend in the solidus. We have not attempted to capture in detail this near-solidus behaviour.

All experiments used to construct the phase diagram are of equilibrium crystallisation type, wherein the bulk composition is the same in each experimental run. The phase diagram should, however, be applicable to fractional crystallisation experiments. To test our approach, we compare the model phase diagram topology against the 0.7 GPa fractional crystallisation experiments of [Nandedkar et al. \(2014\)](#), wherein the initial basalt bulk composition (with 2.3 wt % H₂O) was modified sequentially down temperature to match the liquid composition from a higher temperature experiment. The melt and solid compositions from 15 of their 17 experiments, from 1150 to 780 °C, are an

Table 1: Parameters used to construct the binary phase diagrams; see Appendix B for details. Data for Fig. 1 were obtained by fitting the liquidus in both the temperature and enthalpy plots shown in Ussler and Glazner (1992). The three values of latent heat correspond to pure anorthite (An100), the eutectic composition (An41) and pure diopside (An0), respectively. Data for Fig. 3 are representative of basalt (see Jackson et al. (2018)). All numerical simulations use the phase diagram shown in Fig. 3. The simplified phase diagram was used to obtain the analytical solution of the pure heating/cooling case (Case I) in D

Symbol	Description	Fig. 1	Fig. 3	Units
c_p	heat capacity	1080	1100	$J \cdot kg^{-1} \cdot ^\circ C$
L_f	latent heat	510, 500, 635	550	$KJ \cdot kg^{-1}$
a_1, b_1, c_1	left branch coefficients	-268.3, -172.9, 1390	-99.6, -300.4, 1160	-
a_2, b_2, c_2	right branch coefficients	-303.3, 912.4, 950.9	0, 0, 0	-
e	eutectic point	0.41	1	-
Symbol	Description	Simplified phase diagram		Units
c_p	heat capacity	3000		$J \cdot kg^{-1} \cdot ^\circ C$
p	simplified phase diagram coefficient	0.015		-
q	simplified phase diagram coefficient	-11		-

excellent match to our model liquidus and solidus, respectively, at temperatures above 750 °C; we do not include their 730 and 700 °C experiments as the melt SiO₂ content is considerably higher (ca. 78 wt%, anhydrous) than found in most natural granites of the type we seek to model. The close match of the Nandedkar et al. (2014) melts and those predicted from our simple binary phase diagram lends further support to our approach. The good match to both fractional and equilibrium crystallisation experiments means that we can confidently use this phase diagram to explore not only simple crystallisation scenarios of specific starting compositions, but also reactive flow whereby the bulk composition is repeatedly modified by addition and loss of melt.

We create a mathematical model that fits the experimental data for both phase diagrams (Figs 1 and 3), using a uniform solidus temperature, and a second order polynomial to describe the liquidus temperature, given by (Solano et al., 2014)

$$T(C_s) = T_s \quad (B1a)$$

$$T(C_l) = a_k C_l^2 + b_k C_l + c_k, \quad (B1b)$$

where a_k, b_k, c_k are fitting parameters, and $k = 1$ when $\bar{C} \leq C_e$ and $k = 2$ when $\bar{C} > C_e$.

The anorthite–diopside binary (Fig. 1) exhibits two branches of the liquidus, one on each side of the eutectic composition C_e , and we fit the data by adjusting the value of C_e and the fitting parameters a, b and c (see Table 1 for the values used). The bulk composition $\bar{C}$ varies from zero to one, with zero corresponding to An0 and one corresponding to An100.

The binary phase diagram representing basalt (Fig. 3) exhibits a single branch, with $C_e = 1$; the values of the fitting parameters are given in Table 1. The bulk composition $\bar{C}$ varies from zero to one, with $\bar{C} = 0$ corresponding to the minimum SiO₂ = 47 wt % and $\bar{C} = 1$ corresponding to the maximum SiO₂ = 74 wt %. Figure 3 expresses bulk composition in terms of SiO₂ wt %.

Melt fraction is related to bulk, liquid and solid compositions by the lever rule (equation 10) and enthalpy per unit mass H is related to temperature by

$$H = c_p T + L_f \phi. \quad (B2)$$

Substituting equations B1 and 11 into equation 10, yields leads a third order polynomial in ϕ that can be solved analytically to

give the melt fraction as a function of temperature and bulk composition when the temperature is between the solidus and liquidus. During melting at the eutectic, ϕ can be obtained using equation B2.

We can now express melt fraction as a function of enthalpy and bulk composition as

$$\phi(H, \bar{C}) = \begin{cases} 1 & H_l \leq H \\ \frac{H}{L_f} - \frac{c_p}{L_f} \left(\frac{c_k + ((2a_k(\frac{\bar{C}-C_e+C_e\phi}{\phi}) + b_k)^2 + b_k^2)}{4a_k} \right) & H_e \leq H < H_l \\ \beta(\bar{C}) \frac{H-H_e}{H_e-H_s} & H_s \leq H < H_e \\ 0 & H < H_s, \end{cases} \quad (B3)$$

where

$$\beta(\bar{C}) = \begin{cases} \frac{\bar{C}}{e} & \bar{C} \leq e \\ \frac{1-\bar{C}}{1-e} & \bar{C} > e. \end{cases} \quad (B4)$$

The enthalpy liquidus (H_l), eutectic enthalpy (H_e) and enthalpy solidus (H_s) are given by

$$\begin{aligned} H_l &= c_p(a_k \bar{C}^2 + b_k \bar{C} + c_k) + L_f, \quad k = 1, 2 \\ H_e &= c_p T_s + L_f \beta(\bar{C}) \\ H_s &= c_p T_s, \end{aligned} \quad (B5a)$$

where T_s is the solidus temperature.

Re-arranging equation B1, we can express the solid and liquid composition as a function of temperature between the solidus and liquidus temperatures. Using ϕ obtained from equation B3, C_l and C_s are calculated using

$$C_l(H, \bar{C}) = \begin{cases} \bar{C} & H_l \leq H \\ \frac{-b_1 - \sqrt{b_1^2 - 4a_1(c_1 - T)}}{2a_1} & \bar{C} < C_e \\ \frac{-b_2 + \sqrt{b_2^2 - 4a_2(c_2 - T)}}{2a_2} & \bar{C} > C_e \\ C_e & H_e \leq H < H_l \\ 0 & H_s \leq H < H_e \\ 0 & H < H_s \end{cases} \quad (B6)$$

Table 2: Dynamic parameters used for the numerical simulations. All parameters except the density data in the sill intrusion case are adopted from Jackson *et al.* (2018)

Symbol	Description	Case I	Case II	Case III	Sill intrusion	Units
k_t	thermal conductivity	3	-	-	3	$\text{W} \cdot ^\circ\text{C} \cdot \text{m}^{-1}$
η_0	reference matrix shear viscosity	-	10^{13}	-	$10^{12.5}$	$\text{Pa} \cdot \text{s}$
$\mu_{\min}$	least evolved melt shear viscosity	-	10^2	-	1	$\text{Pa} \cdot \text{s}$
$\mu_{\max}$	most evolved melt shear viscosity	-	10^2	-	10^5	$\text{Pa} \cdot \text{s}$
d	grain size	-	2.75×10^{-3}	-	2×10^{-3}	m
B	permeability exponent	-	3	-	3	-
A	permeability constant	-	125	-	300	-
β	bulk viscosity exponent	-	0	-	0.5	-
$\rho_{l\min}$	density evolved melt	2600	2500	-	2200	$\text{kg} \cdot \text{m}^{-3}$
$\rho_{l\max}$	density refractory melt	2600	2500	-	2600	$\text{kg} \cdot \text{m}^{-3}$
$\rho_{s\min}$	density evolved solid	2600	2800	-	2600	$\text{kg} \cdot \text{m}^{-3}$
$\rho_{s\max}$	density refractory solid	2600	2800	-	3000	$\text{kg} \cdot \text{m}^{-3}$

Table 3: Starting composition used for fractional crystallisation experiments ^aComposition of starting materials; SiO_2 is normalised to 100 ^b Number of experiments use

source	SiO_2^a (wt %)	H_2O^a (wt %)	P (GPa)	$T_{\max}$ ($^\circ\text{C}$)	$T_{\min}$ ($^\circ\text{C}$)	n ^b
Sisson <i>et al.</i> (2005)	51	2.3	0.7	975	825	27
Blatter <i>et al.</i> (2013)	50	2.1	0.7	1150	950	7
Blatter <i>et al.</i> (2017)	66	9	0.7	900	800	5
Melekhova <i>et al.</i> (2015)	47	2.3	0.7	1200	1000	5

$$C_s(H, \bar{C}) = \begin{cases} 0 & H_l \leq H \\ 0 & \bar{C} > C_e \\ 1 & \bar{C} \leq C_e \\ \frac{\bar{C} - \phi_e}{1 - \phi} & H_e \leq H < H_l \\ \bar{C} & H_s \leq H < H_e \\ & H < H_s. \end{cases} \quad (\text{B7})$$

C Numerical model

Governing equations

A one dimensional (1D), two-phase, two-component numerical model is developed to simulate heat, mass and component transport in a crustal mush reservoir and quantify the contributions of heating/cooling, compaction and reactive flow to melt fraction and composition change.

Ignoring the pressure difference and surface tension between the two phases and invoking the Boussinesq approximation, the momentum conservation equation can be expressed as (Bercovici *et al.*, 2001; Jackson and Cheadle, 1998; McKenzie, 1984; Solano *et al.*, 2014)

$$-\phi \frac{\partial}{\partial z} \left[(1 - \phi) \left(\zeta_s + \frac{4}{3} \mu_s \right) \frac{\partial u_s}{\partial z} \right] - \phi(1 - \phi)(\rho_l - \rho_s)g + c(u_l - u_s) = 0, \quad (\text{C1})$$

where ϕ , $\mathbf{u}$, ρ , μ and ζ denote melt fraction, velocity, density, shear and bulk viscosity, respectively, g is the acceleration due to gravity and coefficient c describes the coupling effect between the two phases. Subscripts s and l denote solid and liquid (melt) phases. We report the modelling of density, viscosity and the

coupling coefficient in the next subsection. Note that equation 44 was originally derived for low melt fraction where solid stresses restrict compaction of the solid phase and the relative flow of melt and solid follows Darcy's Law. Here we account for high melt fraction domains following the approach of Keller and Suckale (2019), in which the material properties are suitably adjusted to account for crystal setting through a magma. We describe the approach in the next section.

Mass conservation is described by the continuity equation, which, in 1D, is simply given by (Jackson and Cheadle, 1998)

$$(1 - \phi)u_s + \phi u_l = 0. \quad (\text{C2})$$

Conservation of energy is described using the enthalpy formulation (Katz, 2008; Solano *et al.*, 2014)

$$\frac{\partial H}{\partial t} - L_f \frac{\partial(1 - \phi)u_s}{\partial z} = \frac{k_t}{\rho} \frac{\partial^2 T}{\partial z^2}, \quad (\text{C3})$$

where k_t is the thermal conductivity.

Using the relationship between enthalpy and temperature (equation 38), temperature can be eliminated from equation C3 and the energy equation becomes

$$\frac{\partial H}{\partial t} - \frac{k_t}{\rho c_p} \frac{\partial^2 H}{\partial z^2} = L_f \frac{\partial(1 - \phi)u_s}{\partial z} - \frac{k_t L_f}{\rho c_p} \frac{\partial^2 \phi}{\partial z^2}. \quad (\text{C4})$$

Conservation of components is given by

$$\frac{\partial \bar{C}}{\partial t} + \frac{\partial \phi C_l u_l}{\partial z} + \frac{\partial(1 - \phi)C_s u_s}{\partial z} = 0, \quad (\text{C5})$$

where Fickian diffusion of components is small and is neglected here (Solano et al., 2014).

The final part of the model is the binary phase diagram described in Appendix B, which gives melt fraction, and liquid and solid compositions, as a function of enthalpy and bulk composition.

Equations C1, C2, C4, C5 and the phase diagram (Appendix B) form a closed system that can be solved numerically in the following way. First, equations C1 and C2 are solved with a guessed melt fraction ϕ to obtain the velocity field u_l and u_s . Then, enthalpy and composition transport, equations C4 and C5, are solved to obtain enthalpy H and bulk composition $\bar{C}$. Finally, using the obtained enthalpy and bulk composition, liquid and solid compositions C_l and C_m and melt fraction ϕ are obtained from the phase diagram (Appendix B). An iteration follows until all variables are converged. At the end of each time step, contributions to melt fraction and bulk composition change from heating/cooling, compaction and reactive flow are calculated using equations 8 and 9.

Melt and solid viscosity, density and the coupling term

We assume the viscosity of the solid phase varies with melt fraction, using a simple power-law relationship given by (e.g. Connolly and Podlachikov, 1998)

$$\zeta_s + \frac{4}{3}\mu_s = \mu_0\phi^{-\beta}, \quad (C6)$$

where μ_0 is a reference viscosity and β is a constant that typically ranges between 0 and 1 (Table 2).

Liquid (melt) viscosity is related to the liquid composition C_l (which corresponds to the SiO_2 content normalized to vary between 0 and 1) by (Jackson et al., 2018)

$$\mu_l = 10^{(\mu_{\max} - \mu_{\min})C_l + \mu_{\min}}, \quad (C7)$$

where $\mu_{\min}$ and $\mu_{\max}$ are the viscosities of the least and most evolved melt compositions, respectively (Table 2).

The densities of the liquid and solid phases vary as a function of composition

$$\rho_s = C_s\rho_{s\max} + (1 - C_s)\rho_{s\min} \quad (C8a)$$

$$\rho_l = C_l\rho_{l\max} + (1 - C_l)\rho_{l\min}, \quad (C8b)$$

where $\rho_{s\min}$, $\rho_{l\min}$, $\rho_{s\max}$ and $\rho_{l\max}$ are the densities of the least and most evolved solid and liquid compositions, respectively (Table 2).

The coefficient c in equation C1 describes the coupling between the solid and liquid phases and is chosen to ensure that the governing equations follow the desired physics at low and high melt fraction.

At low melt fraction, the relative flow of liquid and solid phases is described by Darcy's Law, so the coefficient c is related to the permeability k (Bercovici et al., 2001; Keller and Suckale, 2019)

$$c(\phi) = \frac{\mu_f\phi^2}{k(\phi)} \quad \phi \leq 0.2. \quad (C9)$$

Permeability is here related to melt fraction (porosity) using the Kozeny–Carmen relationship, which can be simplified for small

porosity to (Wark et al., 2003)

$$k(\phi) = \frac{d^2\phi^B}{A} \quad \phi \leq 0.2, \quad (C10)$$

where d is the grain size and A and B are adjustable constants. Coefficient c at low melt fraction is then given by

$$c(\phi) = \frac{A\mu_f\phi^{(2-B)}}{d^2} \quad \phi \leq 0.2. \quad (C11)$$

At high melt fraction, we assume the separation of liquid and solid phases is described by hindered-Stokes settling, in which case c is given by (Keller and Suckale, 2019)

$$c(\phi) = \frac{\mu_f}{d^2}(1 - \phi)\phi^{-5} \quad \phi \geq 0.6. \quad (C12)$$

At intermediate melt fraction, a smooth linking function is used to connect equations C11 and C12:

$$c(\phi) = \frac{A\mu_f\phi^{(2-B)}}{d^2}(1 - \mathcal{H}(\phi)) + \frac{\mu_f}{d^2}(1 - \phi)\phi^{-5}\mathcal{H}(\phi) \quad 0.2 < \phi < 0.6, \quad (C13)$$

where

$$\mathcal{H}(x) = \frac{e^{-2/x}}{e^{-2/x} + e^{-2/(1-x)}} \quad (C14a)$$

$$x = \frac{\phi - 0.2}{0.6 - 0.2}. \quad (C14b)$$

In this way, we scale the coupling between the end-members of porous media flow and hindered settling (Fig. 11).

D Analytical solutions and validation of the code

Analytical solution for heating/cooling (pure thermal diffusion) (Fig. 4)

To validate the numerical method outlined in Appendix C when heating/cooling dominate (i.e. there is no flow and heat transport occurs only by conduction), we adopted a simplified phase diagram for which an analytical solution of the simplified system of equations can be obtained. We relate liquid and solid composition to temperature using (cf. equations B7 and B8)

$$C_l = \frac{1}{pT + q} \quad (D1)$$

$$C_s = 0. \quad (D2)$$

With zero velocity (no advection), the governing equations simplify to

$$\frac{\partial H}{\partial t} = \frac{k_t}{\rho} \nabla^2 T, \quad (D3)$$

and the lever rule (equation 10). As there is no advection or diffusion of components, the bulk composition $\bar{C}$ is constant.

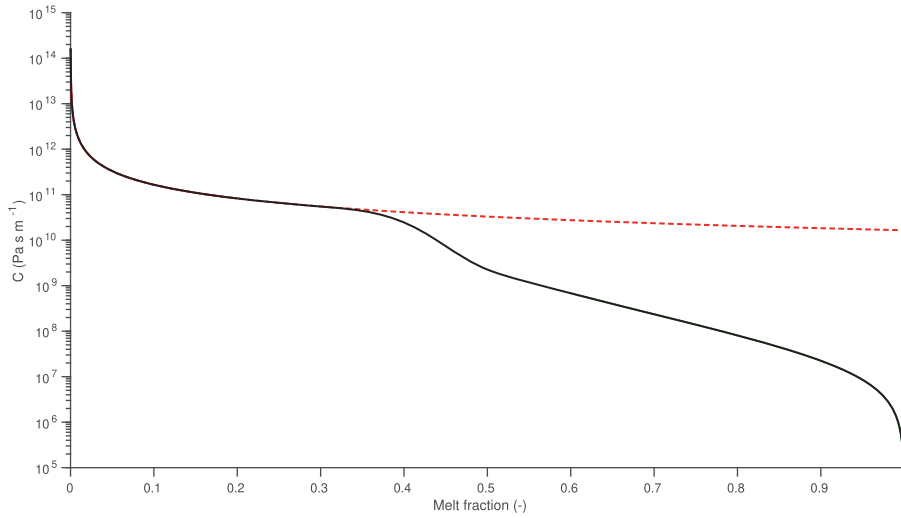


Fig. 11. Coefficient c as a function of melt fraction, calculated using the data reported in Table 2. The solid red line shows the melt fraction range where porous media flow applies ($\phi < 0.2$); the green line shows the melt fraction range where we assume hindered-Stokes settling ($\phi > 0.6$). The black line shows the value of c used here and spans the entire melt fraction range.

Substituting equations D1, D2, 10, into D3 leads to

$$K = \phi^B, \quad (\text{D12})$$

$$\frac{\partial T}{\partial t} = \frac{k_t}{\rho(c_p + \bar{C}_p L_f)} \nabla^2 T \quad (\text{D4})$$

with boundary conditions:

$$\phi = \bar{C}(pT + q), \quad (\text{D5})$$

$$u_s = u_l = 0 \quad \text{at} \quad z = 0 \quad (\text{D13})$$

which can be solved analytically. With the insulating boundary conditions used in case I, melt fraction ϕ , assuming a uniform initial melt fraction ϕ_0 corresponding to a specified initial temperature, is given by

$$\frac{\partial u_s}{\partial s} = 0 \quad \text{at} \quad z = z_m(t). \quad (\text{D14})$$

Equations (66)–(71) are non-dimensionalised using the compaction length and compaction time

$$\phi = \bar{C}p \sum_{i=1}^{\infty} E_i(x) \cos\left(\frac{\pi i}{L}x\right) e^{-D(\pi i/L)^2 t} + \bar{C}q, \quad (\text{D6})$$

$$\delta_c = \left[\frac{\phi_0^B d^2 / a(\xi + 4\eta/3)}{\mu} \right]^{1/2} \quad (\text{D15})$$

where L is the length of the domain and

$$\tau_0 = \left[\frac{\mu(\xi + 4\eta/3)}{\phi_0^B d^2 / a(1 - \phi_0) \Delta \rho g} \right]^{1/2}. \quad (\text{D16})$$

$$E_i(x) = \int_0^L \frac{1}{L} \cos\left(\frac{\pi i x}{L}\right) \phi_0(x) dx \quad (\text{D7})$$

$$D = \frac{k_t}{\rho(c_p + \bar{C}_p L_f)}. \quad (\text{D8})$$

The temperature T can be obtained from equation 62.

Semianalytical solution for compaction (Fig. 5)

To validate the numerical method outlined in Appendix C when compaction dominates (i.e. temperature remains uniform and constant, so there is no component exchange) we use the solution of Richter and McKenzie (1984) for compaction of a system with uniform initial melt fraction ϕ_0 . In dimensionless form, the solution is given by

$$\frac{\partial \phi}{\partial t} - \frac{\partial}{\partial z} [(1/\phi_0 - \phi)u_s] = 0 \quad (\text{D9})$$

$$u_l = -\frac{(1 - \phi_0 \phi)u_s}{\phi_0 \phi} \quad (\text{D10})$$

$$\frac{\partial^2 u_s}{\partial z^2} - \frac{1}{K} u_s - \frac{(1 - \phi_0 \phi)}{\phi_0 \phi} = 0 \quad (\text{D11})$$

Analytical solution for reactive flow (Fig. 7)

To validate the numerical method outlined in Appendix C when reactive flow dominates (i.e. temperature remains fixed and the matrix is rigid so there is no compaction), we assume the melt Darcy velocity is constant and the solid velocity is zero, so

$$\phi u_l \equiv F \quad (\text{D17})$$

$$u_s = 0. \quad (\text{D18})$$

Because the temperature is fixed, the liquid and solid compositions are fixed. From equation 8, the change in melt fraction reads

$$\frac{\partial \phi}{\partial t} = -\frac{1}{C_l - C_s} [(1 - \phi)u_s \cdot \nabla C_s + \phi u_l \cdot \nabla C_l] \quad (\text{D19})$$

$$= -\frac{1}{C_l} [F \cdot \nabla C_l], \quad (\text{D20})$$

which leads to

$$\phi = \phi_0 - \frac{1}{C_l} [F \cdot \nabla C_l] t, \quad (\text{D21})$$

where ϕ_0 is the initial melt fraction at that location and t is the elapsed time.

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