SCIENCE CHINA
Earth Sciences

• REVIEW •

February 2018 Vol.61 No.2: 190–204
https://doi.org/10.1007/s11430-016-9120-y

Crystal fractionation of granitic magma during its non-transport processes: A physics-based perspective

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Received October 29, 2016; accepted October 9, 2017; published online January 2, 2018

Abstract Granitic continental crust distinguishes the Earth from other planets in the Solar System. Consequently, for understanding terrestrial continent development, it is of great significance to investigate the formation and evolution of granite. Crystal fractionation is one of principal magma evolution mechanisms. Nevertheless, it is controversial whether crystal fractionation can effectively proceed in felsic magma systems because of the high viscosity and non-Newtonian behavior associated with granitic magmas. In this paper, we focus on the physical processes and evaluate the role of crystal fractionation in the evolution of granitic magmas during non-transport processes, i.e., in magma chambers and after emplacement. Based on physical calculations and analyses, we suggest that general mineral particles can settle only at tiny speed ($\sim10^{-9}$–10^{-7} m s^{-1}) in a granitic magma body due to high viscosity of the magma; however, the cumulating can be interrupted with convection in magma chambers, and the components of magma chambers will tend to be homogeneous. Magma convection ceases once the magma chamber develops into a mush (crystallinity, F>~40–50%). The interstitial melts can be extracted by hindered settling and compaction, accumulating gradually and forming a highly silicic melt layer. The high silica melts can further evolve into high-silica granite or high-silica rhyolite. At various crystallinities, multiple rejuvenation of the mush and the following magma intrusion may generate a granite complex with various components. While one special type of granites, represented by the South China lithium- and fluoride- rich granite, has lower viscosity and solidus relative to general granitic magmas, and may form vertical zonation in mineral-assemblage and composition through crystal fractionation. Similar fabrics in general intrusions that show various components on small lengthscales are not the result of gravitational settling. Rather, the flowage differentiation may play a key role. In general, granitic magma can undergo effective crystal fractionation; high-silica granite and volcanics with highly fractionated characteristics may be the products of crystal fractionation of felsic magmas, and many granitoids may be cumulates.

Keywords Granite, Crystal fractionation, Magma convection, Layering structure, Mush model, Highly fractionated granite, Granite complex, Li-F granite

Citation: Chen C, Ding X, Li R, Zhang W Q, Ouyang D J, Yang L, Sun W D. 2018. Crystal fractionation of granitic magma during its non-transport processes: A physics-based perspective. Science China Earth Sciences, 61: 190–204, https://doi.org/10.1007/s11430-016-9120-y

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1. Intruduction

Granite is one of the most important components of the Earth's continent, which makes the Earth distinguished from other planets in Solar System (Campbell and Taylor, 1983; Taylor and McLennan, 1985; Wu et al., 2007). The formation and evolution of granite is always a hot topic. Numerous studies show that crystal fractionation plays a significant role in magma evolution, and is one of the major mechanisms driving magmatic differentiation (Wager and Brown, 1968; Lee and Bachmann, 2014). By removing crystallized minerals with different components from magmas, magmas are fractionated and evolved (Becker, 1897; Wu et al., 2017). According to different action principles, various mechanisms of crystal fractionation have been proposed including hindered settling (Davis and Acrivos, 1985), compaction (McKenzie, 1984, 1985), convective fractionation (Sparks et al., 1984) and flowage differentiation (Bhattacharji and Smith, 1964; Barrière, 1976).

Actually, the above mentioned conceptions of crystal fractionation mechanism mostly come from the knowledge of mafic magmas. Nevertheless, it is still vigorously debated whether crystal fractionation can effectively work in granitic magma systems. Since granitic magma is more silicious than mafic magma and there are several orders of magnitude differences in viscosity between both, some researchers hold negative views on the possibility of crystal fractionation of granitic magma (Reid et al., 1993; Pitcher, 1997; Zhang et al., 2007; Zhang, 2012; Clemens et al., 2010; Clemens and Stevens, 2012; Gualda et al., 2012). For example, Zhang et al. (2007) and Zhang (2012) suggested that high viscosity of granitic magma as well as the similar density between crystallized minerals and melts would hinder separation of particles from melts. In field investigations, granite is rarely associated with accumulative rocks, whereas mafic intrusions are frequently accompanied by cumulates, plausibly suggesting that it is less possible for granitic magma to differentiate by crystal fractionation (Zhang et al., 2007). However, one thing should be noted that there may exist a significant difference in the form of cumulates between felsic magma and mafic magma due to their diversity in viscosity. It is thus questionable to identify cumulates of granite based on criterions of residues of mafic magma.

Besides, it is still obscure that which phenomena in felsic plutons are associated with crystal fractionation. For example, it is highly controversial whether the layering structure and K-feldspar cumulates structure, which show variety in components on even small lengthscales, are formed through crystal fractionation (e.g., Clarke and Clarke, 1998; Glazner et al., 2008; Streck and Grunder, 2008; Solgadi and Sawyer, 2008). And there is an enduring argument focusing on whether various components in granitic plutons are caused by crystal fractionation or not (Bateman and Chappell, 1979; Tindle and Pearce, 1981; Coleman et al., 2004; Glazner et al., 2004). Therefore, finding exact petrological evidences is the key to supporting crystal fractionation in granitic magma. Recently, more and more studies indicate that there seem to be a close genetic relation between volcanics and plutons (Hildreth, 2004; Bachmann and Bergantz, 2004; Lundstrom and Glazner, 2016), and rhyolite and high-silica granite are likely derived from granitic magma by crystal fractionation (Bachmann et al., 2007; Lee and Morton, 2015; Lee et al., 2015). Additionally, some special granite (such as lithium- and fluoride- rich granite, Li-F granite) are often closely related to ore deposit formation, showing a co-existence relation with moderately evolved granites in filed (Yin et al., 1995; Zhu et al., 2001). These special granites provide further insights into crystal fractionation in granitic magma.

In this paper, based on the state of magma aggregation, we divide magma evolution into two stages, namely magma transport processes and non-transport processes respectively. Magma transport refers to the migration of magma through some certain magmatic channels from sources to magma chambers and from chambers to the magma intrusion positions; while the non-transport processes include the magma activities in chambers and the gradual cooling in periods of post-intrusion. Obviously, different physical processes occur in both processes. The former is mainly controlled by magma flow in magma channels, while the latter is more complex and generally responsible for differentiation in granite compositions. To simplify, this paper is mainly focused on the non-transport processes that are of greater concern in general researches. The interference caused by the magma differentiation during the transport processes is required to be removed. On the issue of crystal fractionation of granitic magma, previous studies focus more from the perspectives of geochemical and field geological inversion analysis, but their implications are highly debated (Zhang et al., 2007). More crucially, the granitic magma with a certain degree of crystallinity is a non-Newtonian fluid with high viscosity, which is the root that causes researchers suspicious of crystal fractionation in granitic magma (Ma, 1989; Zhang et al., 2007). Therefore, we choose the physical view to review the crystal fractionation of granitic magma during non-transport processes. This paper mainly tries to answer two questions: (1) Can crystal fractionation work effectively in felsic magma systems? (2) If do, which geological phenomena are caused by the crystal fractionation of granitic magma? The key physical factors in these processes and the relationship between crystal fractionation and the related geological phenomena will be discussed below.

2. Physical properties of granitic magma chambers and mineral behaviors in chambers

2.1 Settling behavior of crystal particles in Newtonian fluid and felsic magma (non-Newtonian fluid)

Relative displacements between particles and melts as well as effective aggregation among particles are the key points for crystal fractionation to effectively work (Wu et al., 2017). Therefore, getting knowledge of the behaviors of mineral crystals in magma is critical to understand mechanisms of crystal fractionation. The movement of particles in melts is closely associated with viscosity, crystallinity, fluxion and other physical factors of magma. Among these factors, viscosity is mainly controlled by magma compositions and temperature, which is one of significant differences in physical properties between basaltic and granitic magma. Generally, the viscosity of basaltic and granitic magma is largely distinguished. For example, H_2O-rich, crystal-poor, and high-silica magmas are with viscosity of ~$10^{4.5}$ Pa s (Scaillet et al., 1998; Clemens and Petford, 1999), while the viscosity of basaltic magmas is ~ 10^2 Pa s (Murase and McBirney, 1973), showing nearly three orders of magnitude difference between both. With differentiation, magmas become richer in Si and more viscous. For example, in the processes of cooling and solidification of andesite and dacite magmas (SiO_2=60–70 wt%), the residual melts (i.e., the matrix glass corresponding to crystals) will evolve into high silica rhyolitic compositions due to intensive crystallization of minerals (Maughan et al., 2002; Sparks et al., 2008). Therefore, it is necessary to understand the behavior of granite rock-forming minerals in felsic magma.

Crystal particles in the Newtonian fluid and non-Newtonian fluid system will follow different physical laws respectively (Ma, 1987, 1989). Briefly, Newtonian fluid is a fluid where shear stress is linearly proportional to shear deformation rate at any point, and the fluid that does not satisfy this condition is a non-Newtonian fluid; non-Newtonian fluid need to overcome yield strength to produce deformation (Ma, 1987, 1989; Street et al., 1996). Previous studies have found that crystallinity has a significant effect on fluid properties of melts. In the experimental study by Martin and Nokes (1988), they observed that the settling behavior of crystals in a fluid system at particle concentration of <0.01 wt% satisfies the law of motion under Newtonian fluids. Koyaguchi et al. (1990) observed the movements of particles in slightly more dense suspensions (0.3 wt%~0.1 vol.%). Through their observation, the motion behavior of particles in such a system changed significantly and the fluid exhibited non-Newtonian fluid properties.

In a static Newtonian fluid system at particle concentration of <0.01 wt%, a simple force analysis of crystal particles (the crystal particles discussed in this paper are assumed to be spherical) (Figure 1) shows that crystal particles will be subjected to gravity and buoyancy when still. If gravity is greater or less than buoyancy, the particles will sink or float. Then the crystals begin to move and the magmas drag crystals. Drag force is opposite to the direction of particle motion, and the

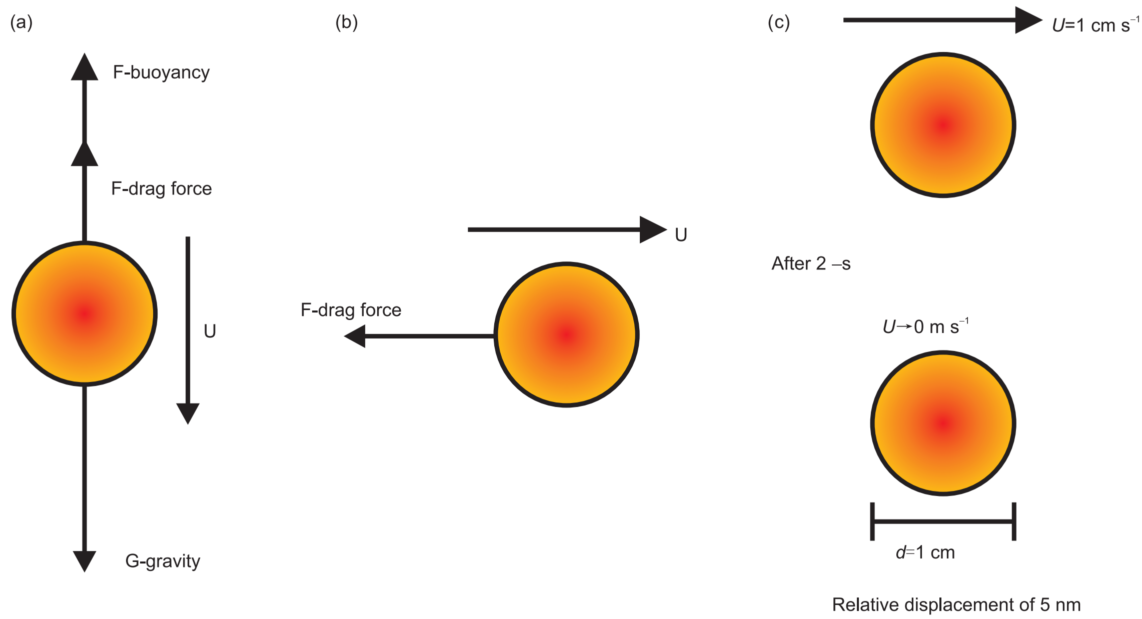

Figure 1 Schematic diagram of movement and force of mineral particles in magma. (a) The force analysis of a settling particle with greater density than melts in static magma; (b) the force analysis of the moving particle in magma (ignoring the gravity); (c) assuming that a feldspar or quartz crystal with a diameter of 1 cm at 1 cm s^{-1} relative to the rhyolite magma, the particle will tend to still after 2 μs, and the relative displacement between the particle and melts is the one millionth of mineral radius. Thus we can speculate that in flowing magma, the minerals will be carried by magma and difficult to settle.

magnitude of drag force is positively related to particle velocity (U). The particles will do acceleration until forces of crystals are balanced, and thereafter the particles stay at a constant speed. Stokes drag force (F_{Stokes}) is given by:

$$F_{\mathrm{Stokes}} = -6\pi\mu r U, \quad (1)$$

(Street et al., 1996); according to the force balance, the Stokes settling speed (U_{Stokes}):

$$U_{\mathrm{Stokes}} = \frac{2r^2 g\Delta\rho}{9\mu}, \quad (2)$$

where r is radius of the crystal particle, g is the gravitational acceleration, $\Delta\rho$ is the density contrast between crystal and melt, and μ represents the dynamic viscosity of the melt. Assuming that a radius of 2 mm quartz or feldspar particle (density of about 2600 kg m^{-3}) sinks in rhyolitic melt with crystal concentrations <0.01 wt%, density ~2300 kg m^{-3}, and the viscosity of $10^{4.5}$ Pa s, the calculated settling velocity is 8.43×10^{-8} m s^{-1} (i.e. 2.66 m yr^{-1}).

For monodisperse particles in a static, non-dilute suspension (>1 vol. %), due to the existence of yield strength, we correct the settling velocity of particles with a correction factor. The settling velocity can be expressed as:

$$U_{\mathrm{hs}} = U_{\mathrm{Stokes}} \cdot f(c), \quad (3)$$

where U_{hs} is the settling velocity, with

$$f(c) = \frac{(1-c)^2}{(1+c^{1/3})^{[5c/3(1-c)]}}, \quad (4)$$

where $f(c)$ is the correction factor, c is the crystal fraction (Barnea and Mizrahi, 1973). As Figure 2 shows, as the crystallinity increases, $f(c)$ and U_{hs} decrease sharply. When the crystallinity is greater than 0.6, the correction factor is less than 3.47×10^{-2} and the particles tend to stand still. The settling velocity formula of the monodisperse particles provides a general relationship between settling velocity and crystallinity. The polydispersed suspensions are more matchable with the realistic situation of magmas, but the settling behavior under the polydisperse system is more complicated and rarely known (Davis and Acrivos, 1985).

Based on the eqs. (2)–(4), it is known that the most important physical factors for crystal settling are radius of particles and viscosity of magmas. In the equations, r can range from micron- to centimeter- scale, and its impact on velocity is further amplified with square calculation; and the difference in viscosity of different component magmas can be up to 13 orders of magnitude (10^1–10^{14} Pa s, Murase and McBirney, 1973). In contrast, the density contrast between minerals and magmas of various magma systems is generally several hundred kg m^{-3} with the same order of magnitude, inducing limited influence on the settling velocity of crystals.

As stated above, when the particles > 1 vol.%, the rhyolitic melt behaves as a non-Newtonian fluid with a high viscosity. Even in such a system, crystal particles can still undergo

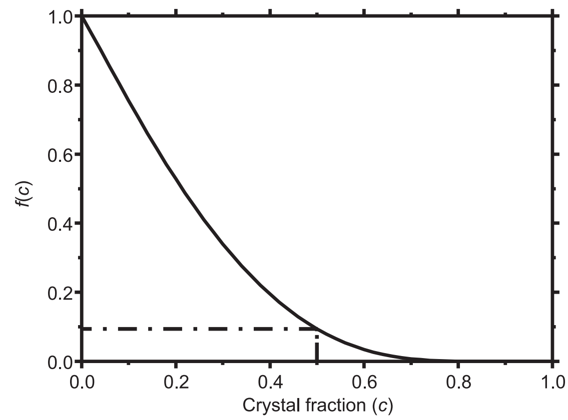

Figure 2 The evolution diagram of empirical correction factor $f(c)$ (Barnea and Mizrahi, 1973) with the crystal fraction (c). When crystal fraction (c) increases, the crystal settling velocity $U_{\mathrm{hs}}=U_{\mathrm{Stokes}}f(c)$ will drop sharply. The settling velocity will fall by about one order of magnitude at c value of 0.5. Modified from Bachmann and Bergantz (2004).

significant gravitational settling, with a precondition that settling time is sufficient and the magma chamber remains relatively static. However, the effect of gravity differentiation is related to many factors such as the shape of magma chambers, the magma chamber lifespan, the convective state of magmas, the crystallization sequence and the difference of settling velocity between different species of minerals. Therefore, crystal fractionation of granitic magma cannot be simply equivalently analyzed with basaltic magma. It is worth noting that the above-mentioned system is attached to one premise-a static magma body. It can be seen from the eq. (3) that, when the crystallinity is at 0.5, the settling velocity of quartz or feldspar particles is about 7.961×10^{-9} m s^{-1} (i.e. 0.251 m yr^{-1}). At such a small settling velocity, even weak magma convection may cause great disturbance to settling of crystals. Thus, the effective gravity-driven separation of minerals from melts requires magma to remain static. In most cases, the convective state of the magma chamber and the motion characteristics of crystals in the flowing magma have a decisive influence on the crystal distribution in chambers.

2.2 The behavior of crystals in the flowing viscous magma

As discussed above, the kinematic behavior of the crystal in magma is associated with gravity, buoyancy, and drag force. In addition, the flow state of magma is equally critical to the motion of particles. Next, we will discuss the behavior of crystal particles in intermediate-felsic magma by following the calculation of Glazner (2014).

It is assumed that thrust is applied to a spherical particle in felsic magma and removed immediately after the particle reaches the velocity of U. The simple force analysis shows that, in the case of neglecting gravity, the particle will decelerate under the drag force of magma until the movement stops (Figure 1c). By the eq. (1) and Newton's second law

$$F = -6\pi\mu rU = ma. \quad (5)$$

According to the calculus meaning of the acceleration

$$a = kU = \frac{dU}{dt}, \quad (6)$$

where $k = -6\pi\mu r / m$. Integrate (6)

$$U = U_0 e^{kt}, \quad (7)$$

in which U_0 is initial velocity and t=–k^{-1}. The sliding distance of the crystal is obtained by the following equation

$$d = \int_0^{\infty} U\,dt = U_0 t. \quad (8)$$

Suppose we apply thrust to a 1 cm diameter quartz particle to a velocity of 1 cm s^{-1} and then remove the thrust. According to the above formula, it can be calculated that the sliding distance d=5 nm (one millionth of the radius of the quartz particle) and the quartz particle will fall to 1% of the initial velocity within 2 μs (Glazner, 2014). The calculation results provide a very critical message for understanding the movement of crystals in felsic magma—When the magma flows at large flow velocity (compared to hindered settling velocity in Section 2.1 calculation), the crystal particles and melts can hardly produce relative displacement. In other words, with the presence of magma flow, the crystal particles are less likely to gravitationally settle but forced to be accompanied with magma to convect.

2.3 Magmatic convection in evolutionary stages of a magma chamber

As previously mentioned, the settling velocity of crystals in static magma chambers is very weak, only to the order magnitude of 10^{-9} m s^{-1}, while the convection of magma seems to be up to ~0.01–0.1 m s^{-1} (Sparks et al., 1984); The high viscosity of rhyolitic magma makes it difficult for particles to slide relative to the flowing magma. As a result, the distribution of crystals in a convective magma chamber is mainly controlled by the convective state of magma.

Some studies suggest that the release of latent heat makes the thermal gradient of magma chambers small, causing convection to be difficult to trigger (Brandeis and Marsh, 1989; Marsh, 1989a, 1989b, 1996). Most researchers' work, however, supports the existence of convection in the early stages of magma chambers prior to extensive crystallization (Ma et al., 1994; Bachmann and Bergantz, 2008a; Huber et al., 2009; Bea, 2010; Gutiérrez et al., 2013). For a granitic magma chamber under a closed system, the magma convection can be divided into two stages, namely, the pre-mush and the mush (Bea, 2010; Bachmann and Bergantz, 2004). Crystallinity is the main basis for dividing the convection stages. It is generally believed that there is convection in magma chambers when the crystallinity is at less than 0.5, and when the crystallinity is greater than the critical point (~0.5), the magma chambers will exist in the form of crystalline congee (Mush), and the magma convection capacity will be lost (Ma, 1994; Bachmann and Bergantz, 2004; Bea, 2010). In this chapter, we mainly discuss the process of convection in the pre-mush stage, and the magmatic differentiation of the mush stage will be discussed in the following chapter.

In the case where the crystallinity of the magma chamber is less than 0.5, magma convection occurs due to the presence of thermal gradient and negative gravity gradient (Grout, 1918), causing the magma composition to be more homogeneous to a certain extent (Whitney and Stormer, 1985; Oldenburg et al., 1989; Lindsay et al., 2001; Christiansen, 2005). Some of researches argue that convection leads to magmatic stratification (Chen and Turner, 1980; McBirney, 1980; McBirney et al., 1985; Bergantz and Ni, 1999; Jellinek and Kerr, 1999; Jellinek et al., 1999). At the edge of the magma chamber and the top of the solidification front, due to contact with the surrounding wallrock and faster cooling rate, these areas are of higher crystallinity than other parts in the chamber and reach the mush stage earlier. At the same time, the density of mush increases and a negative gravity gradient occurs. When gravity is unstable, the more viscous solidification front and the edge are settling, triggering convection. Subsequently, the surrounding parts with lower crystallinity and higher temperature are floating to fill the space. The cycle seemingly is endless until the magma chamber cools adequately so that the solidification front cannot trigger gravity instability. Therefore, convection caused by gravity instability cannot make the magma completely homogeneous. If there were no chemical diffusion in magma, the first sinking parts only reach the higher crystallinity earlier than the magmas that are later filled into the solidification front. And there should be no difference in the composition of magmas when the magma body is solidified. In fact, there is a material exchange between the magmas. However, considering the high viscosity (> 10^4 Pa s) of the granitic magma, material exchange is limited in the absence of convective mixing; the sinking solidification front will have largely temporal and spatial contact with the other major magmas in the chamber during the settling, so the settling materials in this process should tend to be consistent with the compositions of the other main magmas. Therefore, we believe that convection hardly leads the regular stratification of the magma chamber. Huber et al. (2009) simulated the convection in the magma chambers, showing that magma will be homogeneous in the early convection. For the stage where the magma convection gradually weakens, they suggest that mushification can promote magma chambers to be more homogeneous. At the same time, the intrusion of hot magma can rejuvenate mush and recover convection, so that magma compositions can be further homogeneous. Finally, we note that many granites are monotonous intermediates (MI; Hildreth, 1981), which exhibit significant homogeneity at diagenetic temperature, crystallinity and components (Bateman and Chappell, 1979; Whitney and Stormer, 1985; Bachmann et al., 2002). And the single phase zone in a complex is also existed as MI. Therefore, we argue that the convection causes the composition of the magma chamber to be homogeneous; additionally, prior to the mush stage, particles can hardly settle and melt and minerals cannot be effectively separated due to existence of convection.

2.4 Longevity of granitic magma chambers

Whether there is sufficient time for magma to effectively experience crystal fractionation and evolve is determined by the longevity of granitic magma chambers. The lifespan of a magma chamber depends on the increase and decrease of heat in the magma system. Heat loss is mainly related to the factors such as magma convection, heat conduction, and thermal physical properties of wallrock, and heat uptake is mainly from the supply of high temperature magma and latent heat release during crystal crystallization (Gelman et al., 2013; Marsh, 1981). Here we do not discuss the longevity of magma chambers in detail. Bachmann (2011) had a very comprehensive review on this subject. Numerous studies have shown that the curing rate of magma chambers in upper-middle crust is very fast; the temperature of small intrusions can be cooled below solidus in thousands years, and even for large intrusions, the curing time will not be more than hundreds of thousand years. Overall, the lifespan of magma chambers is ~10^3–10^5 yr (Pitcher, 1997; Petford et al., 2000; Glazner et al., 2004; Stimac et al., 2001; Cooper and Kent, 2014). For example, the thermal evolution simulation of a 5 km thick tabular intrusion by Lee et al. (2015) shows that even if the intrusion is emplaced in middle crust (background temperature ~400°C) with neglecting the hydrothermal circulation between the magma body and wallrock, the magma chamber will drop below Zr solubility temperature of zircon and solidus in 1 Myr under all different bulk water contents (Figure 3). Based on previous studies, we limit the age of zircons recorded in a magma chamber to 1 Myr.

3. Applicability of crystal fractionation in interpreting geological phenomena in granite

3.1 Pseudo-sedimentary layering in intrusions

After understanding the movement characteristics of mineral crystals in granitic magma, we might as well to review some of phenomena in granitic plutons that are generally considered to be associated with crystal fractionation. For instance, the layer structure which is often observed in granitoid (Figure 4a and b), it is generally found at the edge of plutons. In traditional views, this layering has been used to be analogous with sediment erosion and deposition (e.g. Gilbert, 1906; Emeleus, 1963; Smith, 1975; Clarke and Clarke, 1998; Solgadi and Sawyer, 2008; Barbey, 2009; Gaweda and Szopa, 2011), and some studies ascribe to shear segregation (e.g. Wilshire, 1969; Barrière, 1976; Bateman, 1992). However, as mentioned above, when magma flows, crystal particles will move in accompany with magma, and cannot settle and be separated from magma. Perturbation from flow cannot form such layering by gravity sorting (Glazner, 2014), which is determined by high viscosity of granitic magma.

Although layering and related structures cannot be completely analogous to sedimentary layers, it is still unable to neglect the potential role of flow and compaction in magmatic differentiation (Clemens, 2015). For example, the scroll-like structures in Figure 4c are most likely to be formed by shear triggered by flow velocity contrast (Clemens, 2015). Based on force analysis, it is found that, in the magma where velocity contrast exists, shear force on minerals is always perpen-

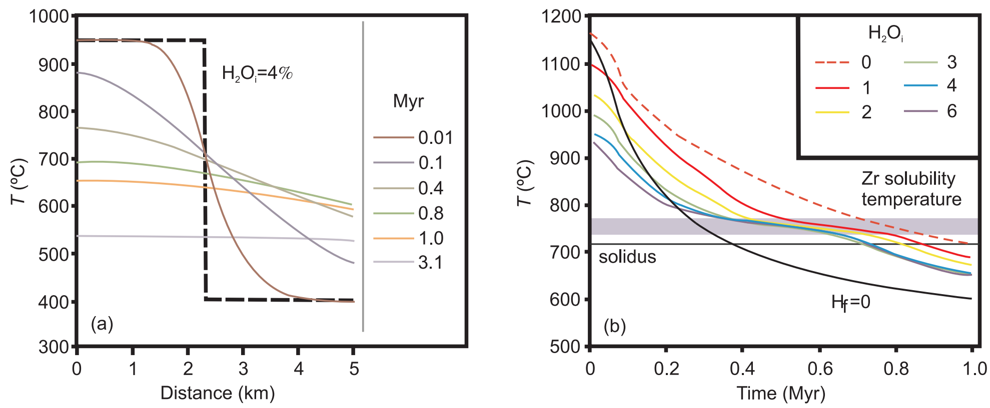

Figure 3 (a) Thermal evolution of a 5 km thick tabular intrusion intruding into wallrock with a background temperature of 400°C. Initial temperature of the magma is assumed to correspond to its liquidus, and the magma component is assumed to be a tonalitic melt with 4 wt% water. The temperature profiles at different times are shown in the figure. The thermal simulation takes into account the heat transfer under the latent heat release of crystallization, but the convection factor is neglected. The dotted line represents initial situation. (b) Temperature of the center of the magma chamber is a function of time at the different total water contents of systems (assuming a closed system). Initial temperature of the magma corresponds to the liquidus at each water content. The lines of different colors represent simulations of different water content systems. All models assume that the total latent heat release is a certain value. The black curve represents the unrealistic scenario where the total latent heat is 0. Modified from Lee et al. (2015).

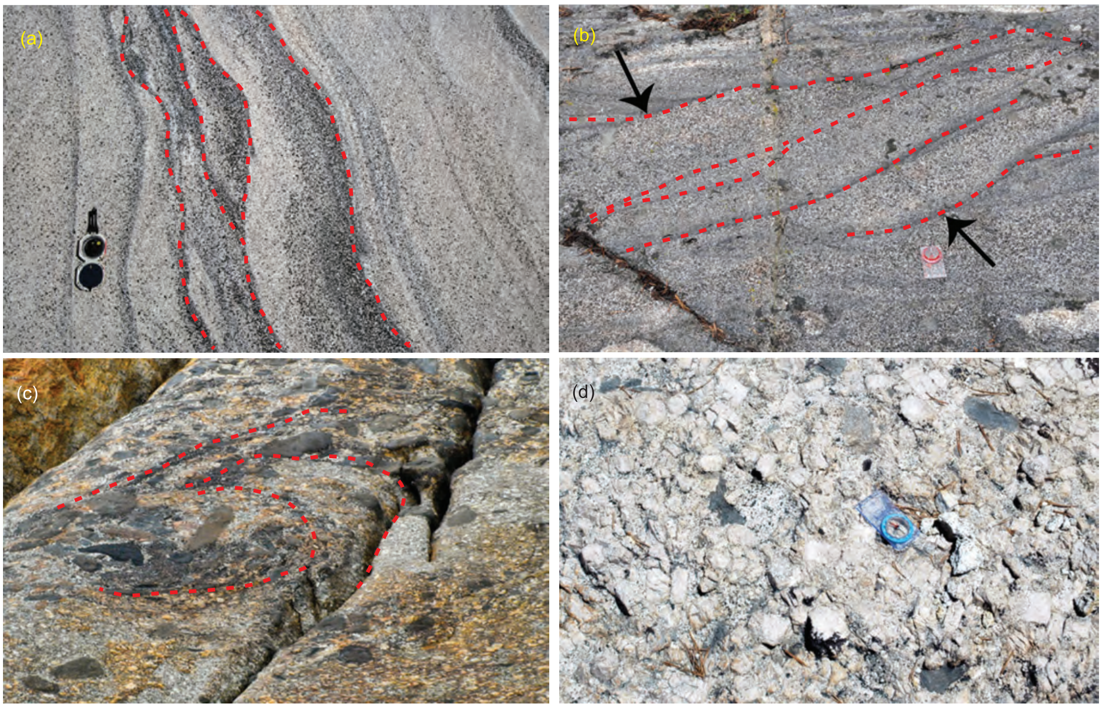

Figure 4 (a) Pseudo-sedimentary layering in granodiorite. Half Dome granodiorite, Tenaya Lake , California (USA). Compass is 7 cm wide (Photo from Glazner, 2014). (b) Truncated layering in Cretaceous granodiorite, in Rock Creek, eastern Sierra Nevada, California. Long side of compass is 9.5 cm (Photo from Glazner, 2014). (c) Scroll-like structures, showing local concentrations of giant K-feldspar phenocrysts, elongate enclaves and biotite, garnet, plagioclase and other minerals. In an S-type porphyritic monzogranite of the Wilsons Promontory batholith of southeastern Australia (Wallis, 1981) (Photo from Clemens, 2015). (d) K-feldspar megacrysts in granodiorite developed in Yosemite National Park, California, USA. Megacrysts are 5–10 cm in long dimension and make up 60–80% area of the outcrop in this area. Compass diameter is 5 cm (Photo from Glazner, 2015).

dicular to velocity, and always coexists with velocity contrast. Therefore, shear force can be a continuous force, and the resulting movement cannot be disturbed by magma flow. Consequently, such scroll-like structures can be formed by flow differentiation, which is not contradictory to theoretical analysis and is a reasonable genetic hypothesis. So, we believe that such scroll-like structures are products of local crystal concentrations and should be the product of flow differentiation.

In theory, some phenomena can be formed by crystal fractionation. However, further studies found that these conclusions may still have omissions. For example, Figure 4d shows K-feldspar megacrysts in granitic rocks found in the Yosemite National Park, California, USA. Previous studies suggested that these megacrysts probably are cumulates by gravity settling during early saturation (e.g. Gilbert, 1906; Žák and Paterson, 2005). By the eqs. (2)–(4), a 10 cm diameter megacryst settles faster about 10000 times than a feldspar with general size (diameter ~1 mm), which is equivalent to the magma viscosity drop of 3–4 orders of magnitude. Thus, from the physical perspective, it seems that these K-feldspar megacrysts indeed are cumulates. However, recently, further work suggests that megacrystic granites possibly occur at late crystallization due to thermal buffering by recharge magmas (See Johnson and Glazner, 2010; Glazner and Johnson, 2013). We believe that the genesis of such structures as scroll-like structures and K-feldspar megacrysts should be cautiously interpreted from both forward modeling and inversion. Even from a physical point of view, it is reasonable to explain, with crystal fractionation, such structures. Further work is still needed to support or eliminate these hypotheses. In flowing magma, as implied by calculations of Glazner (2014), there is almost no relative movement between crystals and melt in the direction of inertia, and movement of crystals is mainly controlled by the flowing magma. Such structures as layering in intermediate to felsic intrusions should be analyzed from the perspective of non-gravitational differentiation, such as flow differentiation. It needs to be clear that, in flowing magma, minerals cannot inertially move relative to magma, which does not mean that crystals and magmas cannot move relative to each other. For example, the crystal is continuously subjected to gravity in a relatively static magma chamber, and minerals can still have a significant displacement relative to melts at a certain period of time; besides, in the magma with flow velocity contrast, the relative displacement between particles and melt can also occur under the effects of shear stress.

3.2 Crystal fractionation in magma chambers with high crystallinity-Mush Model

Based on the analyses of physical properties and particle kinematics of granitic magma chambers with various crystallinity, it is suggested that particles can be separated from melts by such gravity-driven mechanisms as hindered settling and compaction. The "extracted" melts can develop into high-silica granite or rhyolite, while the crystal-rich residues (mineral crystals+interstitial residual melt) may be solidified into granite (Gelman et al., 2014). This process of crystal fractionation is named as Mush Model, which is beneath the growing attention from researchers in recent years. Since 2004, there have been great increasing studies from various perspectives, such as physical process calculations, computer simulations, and geochemistry inversions, to validate and understand this geological process (e.g., Hildreth, 2004; Bachmann and Bergantz, 2004, 2008a, 2008b, 2008c; Gelman et al., 2014; Lee et al., 2015; Lee and Morton, 2015; Forni et al., 2016).

The Mush model is mainly used to explain the genesis of crystal-poor rhyolites (Bachmann and Bergantz, 2004, 2008c; Hildreth, 2004; Streck and Grunder, 2008), high-silica granite (Lee and Morton, 2015; Lee et al., 2015) and the volcanic-plutonic connection (Bachmann et al., 2007; Lipman and Bachmann, 2014; Keller et al., 2015). Bachmann et al. (2007) have comprehensively reviewed the pros and cons of this model. Here we will focus on the main physical process of the model:

With minerals crystallized, crystal fraction of magma bodies is gradually increased. When crystallinity is greater than 0.4, the interstitial intermediate-felsic melt will gradually evolve into highly silicic components (Bachmann and Bergantz, 2004; Bachmann et al., 2002). When crystallinity of magma chambers increases to 0.5, magma convection will cease and particles in mushes will form permeable frameworks. By means of hindered settling, compaction, melts can be "pumped" to the top of the magma chamber, forming a high-silica melt layer (Bachmann and Bergantz, 2004; Bea, 2010). If the mush is disturbed, the high-silica melt layer may intrude, erupt, and eventually form rhyolites. High-silica granite (highly fractionated) can be generated if the high silica melt is slowly solidified in the crust. Then, when the crystallinity of residues reaches a critical point (~0.7), the crystal-melt separation ceases, and magma will lose its mechanical activity and tend to solidify (Bachmann and Bergantz, 2004; Bea, 2010; Hildreth, 2004) (Figure 5). The so-called hindered settling, as shown in previous calculations, is a movement process where crystal particles can sink at small velocity after magma convection stops. Compaction is another physical mechanism that promotes crystal-melt separation. With the influence of mineral crystal self-weight, the interstitial melt in mushes can be extruded from bottom to top (it is in 10^4–10^6 yr that the particles with radius of 2–3 mm can produce a 500 m-lengthscale separation of minerals and melts by compaction, Bachmann

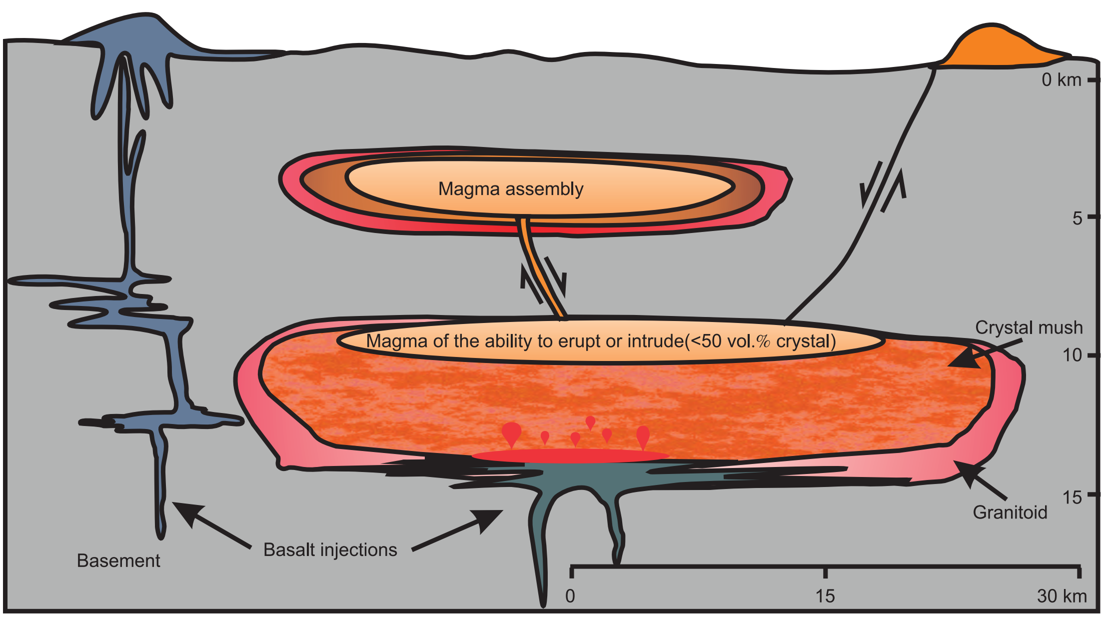

Figure 5 Schematic diagram of Mush Model (based on Bachmann and Bergantz (2008b) and Castro (2013)). When the magma chamber reaches the crystallinity threshold, magma convection is stopped. In a static mush chamber, due to the lack of disturbance of convection, the interstitial melt can be separated from minerals through hindered settling and compaction by gravity. The high-silica and crystal-poor melt layer will be formed at the top of the magma chamber. If erupted is the magma with capacity to erupt/intrude, the corresponding rhyolite can be formed; if melt is trapped at the top of the magma chamber or intruded into crust, it can form high-silica granite showing highly fractionated. Additionally, magmatic recharge is a common phenomenon. The solidified mush will be remelted partly with the penetration of hot magmas. In following processes, the magma chamber will undergo partial remelting, contamination with upwelling magmas and wallrock, followed by cooling, hindered settling and compaction again. After that, compositions of the high-silica melt layer must evolve and change. In addition, magma recharge may also trigger the top melts to intrude. Compositions extracted from mushes of various evolution are bound to exist differences.

and Bergantz,2004).

Assuming that there is a radius of 2–3 mm quartz or feldspar (about 2600 kg m^{-3}), the sinking distance is recorded when crystallinity of the magma chamber is 0.5. Due to the crystallization of minerals, melts evolves into rhyolite with density of ~2300 kg m^{-3}. It is calculated that it will take 10^3–10^4 yr for particles to settle down 500 m, which is less than the longevity of silicic mushes (10^3–10^5 yr, Cooper and Kent, 2014; Bachmann and Bergantz, 2004; Lee et al., 2015). The crystallinity is set to be a constant. In fact, the crystallinity of cumulates will increase gradually with the extraction of melts. According to eqs. (3) and (4), settling velocity of particles will be reduced. Suppose that the settling occurs in a large magma chamber (larger than 10^3 km^2) of 5 km thick. If particles settle for 0.5 km, the upper part of the magma chamber can gather 500 km^3 rhyolitic melts, and the crystallinity of the magma chamber can rise from 0.5 to 0.556. As shown in Figure 2, the calculation with a constant crystallinity of 0.5 has little effect on the calculation results, and the order of time scale is still 10^3–10^4 yr. As shown in Figure 6, within the viscosity range of rhyolitic magma ($10^{4.5}$–$10^{5.5}$ Pa s, Scaillet et al., 1998), minerals at various viscosities are all capable of settling enough before the magma chamber loses vitality completely, forming a sufficient amount of high silica melts. Therefore, the Mush model is feasible in the perspective of physical theory.

However, further work for Mush model is still needed to make it validated and perfect. Here, we would like to emphasize that the degree of crystal-melt separation may be different in kinds of tectonic settings. For example, magmatic recharge (Davidson and Tepley, 1997; Schmitt et al., 2001; Tepley et al., 2000; Peate et al., 2008; Wark et al., 2007; Forni et al., 2016) can motivate melt-crystal separation in the long term; however, abrupt development of faults will interrupt the separation. Under various conditions, the evolution of mushes is different, and the resulting geochemical characteristics of cumulated granite and associated top melts will also vary (Figure 5).

3.3 Component variations in composite intrusions: the product of evolved magma chamber?

Some large plutons have 10^2–10^4 m-scale distinctive component zoning, which has long been interpreted to be caused by crystal fractionation (Bateman and Chappell, 1979; Tindle and Pearce, 1981; Sisson and Moore, 1994) or magma mixing (Kistler et al., 1986; Frost and Mahood, 1987). The premise of these research ideas is that such pluton must evolve from the same batch of magmas, i.e., the pluton is directly formed via evolution of one magma body. Recently, based on the studies of high-precision chronology investigation of zoned plutonic suites, it is found that the emplacement timescale of many suites is far beyond time span of evolution of single magma chamber calculated by thermal simulations. This contradiction provides new clues and ideas for understanding the generation of variety in mineral, composition, and structure in large intrusions (e.g., Glazner et al., 2004; Miller et al., 2007; Scoates and Friedman, 2008; Scoates and Wall, 2015).

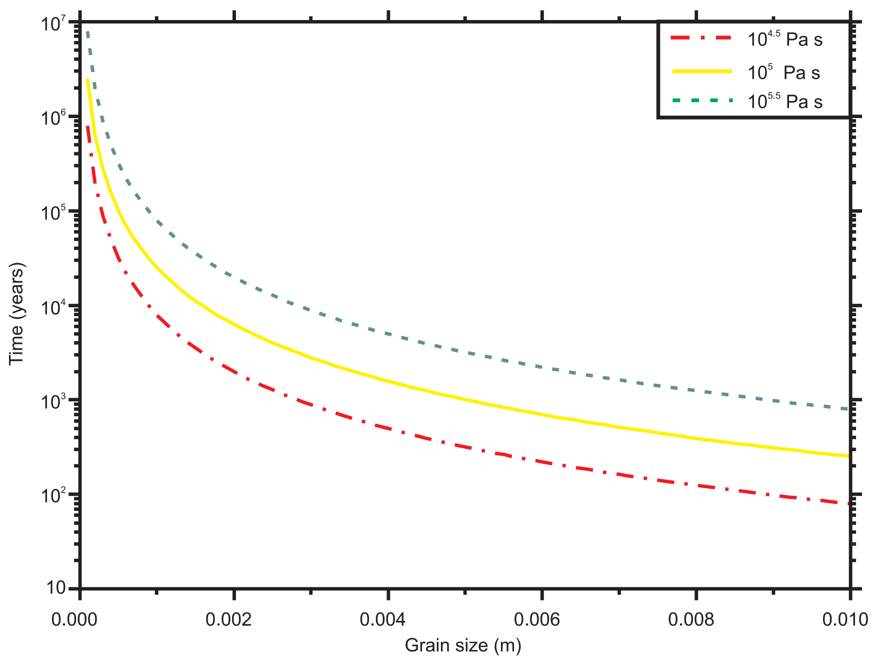

Figure 6 The relationship between crystal radius and time required for crystals (e.g., feldspar, quartz, with a density of 2600 kg m^{-3}) to settle 500 m in silicic mushes (with crystal fraction of 0.5, a melt density of 2300 kg m^{-3}). In the figure, magma viscosities are $10^{4.5}$, 10^5, and $10^{5.5}$ Pa s, respectively. The shadow is the general interval range of time needed to solidify for single-stage magma.

Taking the famous Tuolumne Intrusive Suite, Nevada, USA, for example, its zircon U-Pb ages range from 85 to 95 Ma, and even an individual pluton, Half Dome Granodiorite, had accumulated and solidified over 3 Myr (Coleman et al., 2004; Glazner et al., 2004; Memeti et al., 2010). As discussed above, 1 Myr is essentially the upper limit of zircon U-Pb age span in single injections of magma. Thus, the age interval of Tuolumne Intrusive Suite up to 10 Myr indicates that these intrusions cannot be an evolution-product of an individual magma. Coleman et al. (2004) argued that the various compositions of this suit are not caused by crystal fractionation. We suggest that such a large time span essentially indicates that the suit is not the product of a solidified chamber but a multistage magmatic assemblage; for such plutons, to discuss their origins with crystal fractionation have no premise of discussion. Recently, more studies find that many granitic rocks have the same chronologic variation as the Tuolumne rocks (Matzel et al., 2006), and even many granitic plutons had assembled more than 10 Myr, such as the Indosinian-Yanshanian complexs, which are widely distributed in southern China (Ding et al., 2015). Ding et al. (2015) applied the remobilized Mush model to explain the origins of these complex rocks. That is, when suffering from magmatic-tectonic events, the mushes within deep will rejuvenate with high-silica melts aggregating at the top of mushes. Due to buoyancy, the high-silica melts will then penetrate the magma body and intrudes the overlying crust (Burgisser and Bergantz, 2011). In these processes, the remobilized mush may experience magma mixing, contamination, hindered settling, and so on, resulting in various compositions of intrusions of different stages. Due to frequent magmatic-tectonic events, this rejuvenation may persistently occur. And corresponding magmatic emplacements will also occur in the form of surging or pulsating; additionally, the crystallization age should also be coupled with magmatic-tectonic events (Figure 5). Therefore, more detailed chronological studies of important intrusions are valuable research directions (Coleman et al., 2016), and perhaps bring breakthroughs in solving crucial problems such as granitic magma emplacement and evolution.

3.4 Possibility of forming vertical zonation by crystal fractionation in South China Li-F granite

Based on the analyses of crystal settling and convection in magma chambers, we argue that the probability of forming component and mineral assemblage zonation merely by gravity settling is negligible due to the small settling velocity and the presence of magma convection. However, in the South China, there is a special type of granite with vertically compositional and mineral assemblage zonation (Figure 7), such as the Yashan pluton in Jiangxi Province, the Jianfengling pluton in Hunan Province and the Jiamu pluton in Guangxi Province (Yin et al., 1995; Zhu et al., 2001). Furthermore, the crystalline time span is <1 Myr (in the case of the Yashan pluton, the $^{206}Pb/^{238}U$ ages of the medium-coarse grained protolithionite-muscovite granite and lepidolite granite are 150.1±1.0 Ma and 150.2±1.4 Ma, respectively, Yang et al., 2014), and in the range of age error it cannot be excluded as the result of evolution of the same period magma. This type of pluton has high volatile and network-modifying ion contents such as fluorine, phosphorus and Li^+, which decrease melt viscosity and solidus significantly (Mysen et al., 1981; Xiong et al., 1996, 1999; Manning, 1981; Dingwell and Mysen, 1985; Sirbescu and Nabelek, 2003). The detailed and systematical review of the Li-F granite from geology, petrology, mineralogy and geochemical characteristics have been discussed and summarized by Wu et al. (2017). For the formation mechanism of Li-F granite, the previous views include hydrothermal alteration and magmatic crystal fractionation (Zhu et al., 2002). We do not intend to elaborate on the focus of this dispute. In this chapter, through simple simulations we mainly clarify the particularity of the crystal fractionation process in South China Li-F granite. That is, after intrusion, there is a possibility that component and mineral assemblage zonation occurs by gravity settling.

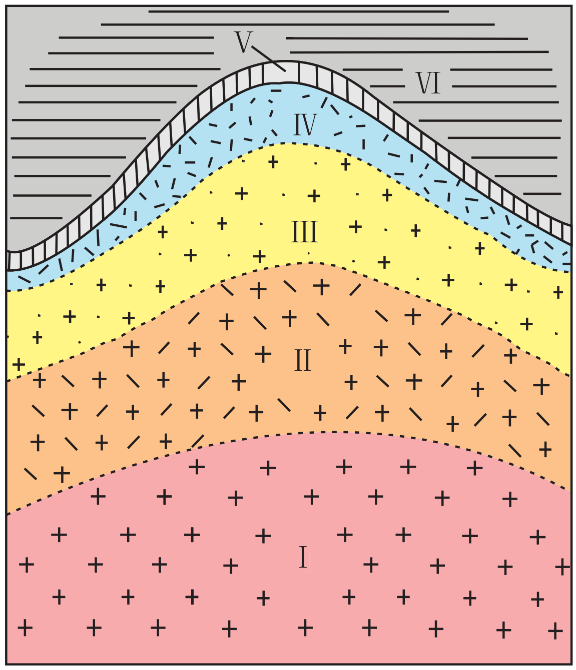

Figure 7 Schematic diagram showing vertical zonation of Li-F granite. I. two-mica granite zone; II. leucocratic granite zone; III. topaz-lepidolite-albite granite zone; IV. greisen zone; V. K-feldspar stockscheider pegmatite; VI. wallrock. Modified from Zhu et al. (2002).

Previous experiments and calculations show that fluorine can effectively reduce the viscosity of magmas, and with decreasing temperature, the increase in the viscosity of the fluorine-rich melt is less than the fluorine-poor melt (Dingwell et al., 1985; Mysen and Virgo, 1985). Dingwell et al. (1985) measured the effect of F on the viscosity of Na_2O-Al_2O_3-SiO_2 system. Based on previous data, it was found that when the temperature of magma decreased from ~1600°C to ~1000°C, the viscosities in all systems increased; for the albite melt, the viscosity of the system with 5.8 wt% F compared to 0 wt% F decreased by about one order of magnitude at 1400°C and dropped by about 2 to 3 orders of magnitude at 1000°C. Because Dingwell et al. (1985) did not experiment under lower temperatures, we speculate, depending on experimental trend, that 5.8 wt% F will reduce the magma viscosity by at least three orders of magnitude when magma temperature is dropped below 1000°C.

We could visually compare viscosities of generally granitic magma, the felsic magma containing 2 wt% F and basaltic magma and the settling velocity in three types of magma. With minerals crystallized, the melt of granitic magma evolves into rhyolitic liquid, and water in melt is continuously enriched. The viscosity of rhyolitic melt with 4 wt% H_2O is $10^{4.5}$ Pa s. Similarly, with the quartz, feldspar and other F-poor mineral crystallization, F contents in melt of Li-F granite magma is increased gradually. According to the above experimental results, we conservatively estimate that under the condition of ~800°C, the viscosity of Li-F granitic magma compared to general granitic magma falls about two orders of magnitude (~$10^{2.5}$ Pa s). Basaltic magma viscosity is within 10–100 Pa s (Murase and McBirney, 1973), and we take $10^{1.5}$ Pa s in order to facilitate comparison. It is assumed that there are three crystal particles with radius of 2 mm in such three systems with crystallinity of 0.5, and the density contrast is 300 kg m^{-3}. Bring parameters into eq. (3), it can be calculated that the settling velocity in general granitic magma is 7.961×10^{-9} m s^{-1}(i.e. 0.251 m yr^{-1}), in Li-F granite magma is 7.961×10^{-7} m s^{-1} (i.e. 25.1 m yr^{-1}), and in basaltic magma is 7.961×10^{-6} m s^{-1} (i.e. 251 m yr^{-1}) (Figure 8). It is due to high contents of volatile components and network-modifying ions that the settling viscosity of Li-F granite magma can be several orders of magnitude lower than general granitic magma, and by settling, the vertical stratification of minerals and components can form on small lengthscales (e.g. topaz- lepidolite granite in Yashan pluton, 41.5 m, Li and Huang, 2013). However, compared with basaltic magma, the viscosity of Li-F granite magma is still higher, and settling velocity is still smaller. In addition, there are various crystallization sequences in different magma systems, which makes Li-F granite with visible mineral assemblage stratification, but not form obvious mineral cumulates as mafic magma. In general granite magmatic

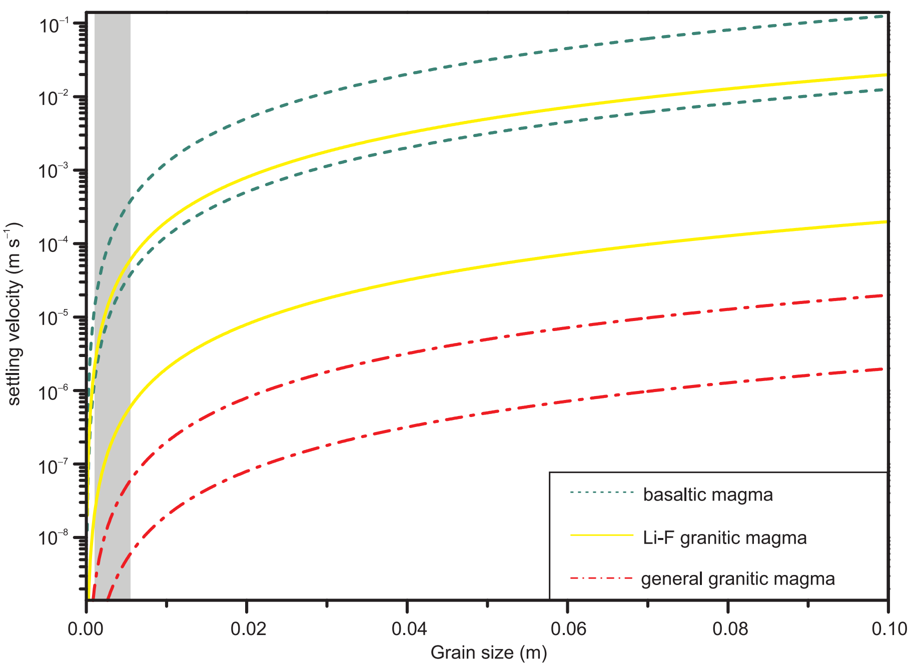

Figure 8 The diagram showing settling velocity versus grain size in several discussed types of magmas. Green curve representing basaltic magma, yellow curve representing Li-F granitic magma, and red referring to general granitic magma. The interval of the same color curves is the variation of the particle settling velocity in the corresponding system. There are significant differences in the viscosity of different magmatic systems, and the settling velocity of minerals is mainly controlled by the viscosity of systems. When the granitic magma is enriched in lithium, fluoride and other such components, the magma viscosity decreases significantly and the settling velocity increases by orders of magnitude, even overlapped with those in basaltic magma systems. While the settling velocity of particles in general granite magma is significantly slower. The shaded portion represents the particle size range of most crystals in nature. Calculation parameters: the density contrast ($\Delta\rho$) between basic magma and crystals approximately at 600 kg m^{-3} in our estimates, the range of viscosity (μ) approximately from 10 to 100 Pa s (Murase and McBirney, 1973); in general granitic magma, $\Delta\rho$ approximately at 300 kg m^{-3}, the range of μ approximately from $10^{4.5}$ to $10^{5.5}$ Pa s (Scaillet et al., 1998); in Li-F granite magma, $\Delta\rho$ approximately at 300 kg m^{-3} and the range of viscosity approximately from $10^{1.5}$ to $10^{3.5}$ Pa s. Three types of systems are studied all with the crystallinity (c) at 0.5.

system, there is no significant change in the ratios of Zr/Hf, Nb/Ta, Y/Ho (Green, 1995). While Li-F granite may undergo extensive crystal fractionation, causing low Zr/Hf ratios in bulk rock and zircon (Bau, 1996; Breiter et al., 2014). Of course, it is one possibility that this type of geochemical features is generated by the late magmatic hydrotherm (Su et al., 2015; He et al., 2015).

Overall, the viscosity of Li-F granite magmas is several orders of magnitude lower than general granitic magma, which leads to faster settling velocity and possible occurring of vertical stratification through crystal fractionation in Li-F granitic magmas.

4. Conclusions

(1) Crystal fractionation of granitic magma can occur effectively. Crystal fractionation during non-transport processes of granitic magma may be more reflected in the formation of rhyolite, high-silica granite (with distinct highly fractionated characteristics) and cumulated granite as well as in crustal evolution. Crystal fractionation contributes little to compositional variation in single granitic pluton. For "Granites and Granites", namely the diversity of granite, we should pay more attention to the influence of Mush processes on compositions of intrusions, the magma differentiation during magma transport processes, and the difference in magmatic source region.

(2) Granitic magma is generally difficult to form vertical zonation by gravity differentiation. However, the lithium- and fluoride- rich granite in South China has the possibility of forming vertical component zoning by crystal fractionation. This is because Li and F can lower magma viscosity and facilitate rapid particle settling.

(3) The causes of small-lengthscale component variation in plutons need to be further studied. However, it is clear that the effect of gravity differentiation in formation of such "pseudo-sedimentary layering" is weak; flow differentiation is not contradictory to physical analyses and maybe act. Urgently, more quantitative research is needed to constrain the effect of flow differentiation.

Noting that an increasingly heated argument on the state of magma chambers (Coleman et al., 2016; Lundstrom and Glazner, 2016; Wilson and Charlier, 2016; Clemens and Stevens, 2016), the following is added: If granitic magma is emplaced via amalgamation of many small discrete bodies with high initial crystallinity, magma chambers will directly evolve from mush stage; depending on the initial crystallinity, the degree of mush evolution is various. This has no effect on conclusions in this paper.

Acknowledgements *We are grateful to Prof. Fuyuan Wu for providing valuable comments and suggestions during the writing process. We would also like to thank Prof. Zhenhua Zhao, Prof. Qiang Wang for their contributions and insightful discussions. The paper was significantly improved by reviews by Prof. Changqian Ma and Prof. Xiaolei Wang. This work was supported by the National Key R&D Program of China (Grant Nos. 2016YFC0600204 & 2016YFC0600408), the National Natural Science Foundation of China (Grant Nos. 41421062 & 41372005).*

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