

1           **Magma differentiation and contamination: Constraints from**  
2                                   **experimental and field evidences**

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19

*Magma Differentiation and Contamination*21 **Abstract**

22 Differentiation and contamination of silicic magmas are common phenomena  
23 characterizing the granite batholiths and large igneous provinces that build up most of  
24 the continental crust. Although they can be identified by means of geochemical relations  
25 of igneous rocks exposed in the continents, the mechanisms allowing magmas to  
26 undergo the necessary crystal–liquid separation and digestion of country rocks for  
27 differentiation and contamination are poorly constrained. In this paper we show two  
28 independent approaches that are essential to understand fractionation and contamination  
29 of magmas. These are (1) the study and interpretation of field relations in exposed deep  
30 sections of batholiths, and (2) the results of laboratory experiments carried out at  
31 middle–upper crust pressure. Experiments support that fractionation is intrinsic to  
32 crystallization of water-bearing magmas in thermal boundary layers created at the  
33 sidewalls of ascent conduits and walls of magma chambers. Gravitational collapse and  
34 fluid migration are processes identified in experimental capsules. Similarly, reaction  
35 experiments in mixed capsules support reactive bulk assimilation as a plausible  
36 mechanism that is compatible with field and petrographic observations in contaminated  
37 granitic rocks.

38 **1 Introduction**

39 Magmatic fractionation (closed system) and contamination (open system) are common  
40 processes involved in the overall differentiation of igneous rocks in the continental crust  
41 [Bowen, 1928]. The two processes can act jointly in magma chambers and conduits.  
42 Many silicic ( $\text{SiO}_2 > 53$  wt%) igneous rocks, in particular those formed in active  
43 continental margins, can be modeled geochemically as resulting from a combination of  
44 assimilation and fractional crystallization (AFC) [Bohrson and Spera, 2001; DePaolo,  
45 1981]. However, the relative contribution of each process and the mechanisms of  
46 operation in magmas remain unconstrained.  
47 Many igneous rocks appearing in the continental crust, and particularly those richer in  
48  $\text{SiO}_2$ , contain isotopic signatures indicating contamination with older crustal rocks  
49 [Allègre and Ben Othman, 1980; Hawkesworth and Kemp, 2006; Kemp et al., 2007;  
50 McCulloch and Wasserburg, 1978]. Contamination may be acquired either during

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51 ascent and emplacement in the crust, or may be inherited from an already crustal-  
 52 contaminated source. Trace elements and isotopic ratios, which are regularly used to  
 53 make AFC modeling, are unable to discern between the two processes. Fortunately, the  
 54 major element compositions of melts are dependent on the composition of the solid  
 55 saturation assemblage, which is in turn imposed by intensive variables in a given  
 56 system, in such a way that the composition of melts in closed systems will follow  
 57 cotectic lines that can be determined by means of phase equilibrium experiments and  
 58 thermodynamic modeling. Comparisons between rocks and experimental liquids may  
 59 help to distinguish open from closed magmatic systems. It is expected that open system  
 60 processes may introduce characteristic departures in the composition of rocks from that  
 61 of cotectic liquids. In a theoretical case, rocks belonging to a magma fractionation series  
 62 will represent liquids extracted at any time in the course of crystallization. These are the  
 63 so-called *liquid lines of descent* (LLD) [Bowen, 1928]. However, in practice, rock series  
 64 may depart from the ideal composition of LLDs, even in case that the system is closed.  
 65 The reason is that the extracted liquid fraction may carry magmatic crystals in  
 66 suspension from a magma chamber or may drag crystals from the consolidated parts of  
 67 the chamber or conduits in the way upward. These are self-contaminated liquids (Fig.  
 68 1). Similarly, the crystal rich residue left after a partial extraction of melt becomes a  
 69 new magma system that neatly departs from the cotectic. This is a very common case in  
 70 granitic rocks, in which a residual liquid escaped in the course of crystallization. These  
 71 are called *disguised cumulates* [Lee and Morton, 2015], as a cumulate texture is not  
 72 recognized while an off-cotectic composition is identified. Magmatic differentiation by  
 73 crystal fractionation is possible if either a liquid fraction is removed from the  
 74 crystallizing magma or a fraction of crystals is separated away from the magma. Both  
 75 processes differ significantly from each other and can operate under different circumstances.  
 76 The virtual absence of monomineralic layering in silica-rich calc-alkaline systems,  
 77 indicate that crystal settling is not a dominant mechanism.  
 78 Understanding how crystals and liquids are separated in crustal magmas requires special  
 79 attention to physical and chemical features of crystallizing magmas. Most crustal  
 80 magmas are characterized by high silica contents. The implication is that viscosity is  
 81 much higher than that of basaltic magmas, making crystal–liquid separation difficult to  
 82 achieve [Glazner, 2014]. However, many lines of evidence, mostly supplied by  
 83 geological and geochemical relations, point to an effective fractionation in nearly closed  
 84 magma systems. Solution to this paradox has been addressed by several approaches,

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85 including numerical modeling [*Bachmann and Bergantz, 2004; Bachmann and Huber,*  
86 *2016; Burgisser and Bergantz, 2011; Gelman et al., 2014*], analog experiments  
87 [*Michioka and Sumita, 2005; Shibano et al., 2012; 2013*] and experiments with silicate  
88 melts at high pressure and high temperature (*Masotta et al. [2012]; Huang et al. [2009];*  
89 *Rodríguez and Castro [2017]*). Magma crystallization in a thermal boundary layer  
90 (TBL), created for instance at the walls of magma chambers and conduits [*Rodríguez*  
91 *and Castro, 2017*], is the most plausible mechanisms that contributes to separation of  
92 liquids from crystals within a solidification front [*Marsh, 2002*].

93 Because assimilation is usually considered an energy-consuming process, its role in  
94 accounting for significant differentiation of igneous rocks has been questioned [*Bowen,*  
95 *1922; Glazner, 2007; Thompson et al., 2002*]. Energy balance is applied in terms of  
96 xenolith melting and, thus, assimilation is considered as a particular case of magma  
97 mixing [*Thompson et al., 2002*] between melts from the xenoliths and the intruding  
98 magma. Also limiting is the assumed low temperature for the country rocks to be  
99 assimilated [*Glazner, 2007*]. However, the reach of assimilation, far from being a self-  
100 limiting process, can be enlarged in cases of hot country rocks and repeated intrusions  
101 of magma [*Glazner, 2007*]. In addition, country rock xenoliths can be disaggregated  
102 mechanically by inducing melting at low melt fractions, contributing to the so-called  
103 *reactive bulk assimilation* [*Beard et al., 2005*] in which energy consume is minimized.  
104 The observation of contaminated rocks over kilometeric extensions of plutonic intrusions  
105 from the Variscan Gredos batholith in Central Spain, which were emplaced in repeated  
106 layers into migmatitic metasediments [*Díaz-Alvarado et al., 2011*], points to country  
107 rock assimilation as an efficient mechanism that contributes to differentiation of  
108 magmas in the continental crust. These areas provide relevant field relations that may  
109 help to understand the intricacies of assimilation. A summary of these relations is  
110 shown in this paper.

111 In regard of the mechanisms of assimilation, essential questions are: How is in detail the  
112 process of assimilation? How do exotic elements incorporate to the magmas? What is  
113 the scope of assimilation in nature? Answering these questions require a knowledge of  
114 the process. Although some geochemical features may be indicative of magmatic  
115 assimilation, the study of field relations between igneous intrusions and country rocks  
116 in deep-seated plutons is essential to reveal the mechanisms of magma–host  
117 interactions. Even in field-based examples, the intricacies of the processes of magma–  
118 host interaction leading to contamination are poorly constrained. Experiments are useful

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119 to set limits to possible processes involving chemical equilibrium and disequilibrium.  
 120 Integration of petrological phase equilibrium relations and field-based studies are  
 121 essential to discern between competing processes. Furthermore, the identification of  
 122 paleotectonic environments through the geochemistry of magmas requires a wholly  
 123 understanding of petrogenetic processes, contamination being a very common one and,  
 124 at the same time, the most difficult to reveal. At this purpose, we present new field  
 125 description from the Gredos batholith (Central Spain) that are interpreted in the light of  
 126 evidences from relevant laboratory experiments on magma differentiation and crustal  
 127 contamination

128 **2 Geological and Geochemical Inferences on Fractionation**

129 On the basis of geochemical studies, granites of the Cordilleran batholiths are linked to  
 130 fractionation from an intermediate magma precursor of andesitic to basaltic andesitic  
 131 composition (e.g., Sierra Nevada batholith [Lee *et al.*, 2006]; Patagonian batholith  
 132 [Castro *et al.*, 2019; Castro *et al.*, 2011; Pankhurst *et al.*, 1999]). Experimental phase  
 133 equilibria [Castro, 2019 and refs herein] also point to a similar way, adopting an  
 134 intermediate precursor as the parental magma to batholiths. Even in intracontinental  
 135 calc-alkaline batholiths (e.g., Caledonian Newer granites and Variscan batholiths of  
 136 Iberia), whose origin points to fluid-assisted melting of the lower crust (secondary I-  
 137 type granites) [Castro, 2019], a fractionation trend from tonalites and quartz-diorites to  
 138 granodiorites and granites is observed. Curved patterns in Harker diagrams are  
 139 characteristic of cotectic variations and, hence, of liquid fractionation from one or  
 140 various parental magmas. By contrast, rectilinear patterns are considered as indicative  
 141 of magma mixing or contamination, as they result from a mechanical mixing between  
 142 two systems, namely magma–host or magma–magma. However, mixing of fractionated  
 143 liquids with their cognate crystals may also produce rectilinear patterns while the  
 144 system is closed to external contaminants [Bea *et al.*, 2005]. Figure 1a shows internal  
 145 mixing relations between liquid and crystals in the MgO–CaO diagram, which can be  
 146 taken as a proxy of cotectic variations in an ample variety of mafic and intermediate  
 147 systems, including the calc-alkaline series. The two components, MgO and CaO, are  
 148 preferentially partitioned into the solid saturating assemblage, which is dominated by  
 149 Pl+Cpx or Pl–Amp (mineral abbreviations after Whitney and Evans [2010]) along a  
 150 wide temperature interval from near-liquidus to near-solidus conditions. Commonly,

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151 rocks fractionated in closed systems within the continental crust evolve following the  
152 curved patterns dictated by the thermodynamic cotectic. However, rock series evolving  
153 in a closed system may plot outside the cotectic, as internal mixing between fractionated  
154 liquid and crystals from the cumulate is also possible (Fig. 1a). In sum, the geochemical  
155 inferences on fractionation must be taken with care. Only curved patterns are indicative  
156 of fractionation, as they are governed by thermodynamic cotectic variations. A better  
157 way to delimitate patterns of fractionation from those of assimilation is by plotting  
158 rocks on triangular diagrams using a multicomponent space projected onto the plane  
159 Orthopyroxene–Orthoclase–Anorthite (Fig. 1b).

160 In calc-alkaline plutonic systems, layered monomineralic cumulates are rare.  
161 Paradoxically, plutons are formed by rocks that display nearly-cotectic variations. Thus,  
162 where are the cumulates from which fractionated liquids were extracted? The  
163 explanation is that fractionation proceeds by expulsion of liquid from a crystallizing  
164 magma, and not by separation of crystals from the magma. The resulting cumulates are  
165 hardly identified by textures or compositions. Many diorites and gabbros of the calc-  
166 alkaline plutonic associations are “disguished” cumulates [Lee and Morton, 2015]. That  
167 is, they represent crystals aggregates, or mushes, that lost a residual liquid in the course  
168 of crystallization. For this reason, rocks of intermediate composition (diorites and  
169 quartz-diorites) from batholithic associations are scattered in MgO–silica and MgO–  
170 CaO diagrams. These cumulate-like diorites share the same scattered region of lower  
171 crust granulites, pointing to fractionation as an overall process responsible for the  
172 differentiation of the lower and upper continental crust [Castro *et al.*, 2013].

### 173 **3 Mechanisms of Liquid–Crystal Separation**

174 Although fractionation by liquid–crystal separation is a necessary process to account for  
175 geochemical (e.g., cotectic variations in closed systems) and geological (e.g., zoned  
176 plutons) observations, the mechanisms of such a physical separation remain debated.  
177 Liquid expulsion from a crystallizing aggregate is a preferred mechanism in silicic  
178 magmas in which, individual crystal separation by gravity settling is impeded by the  
179 high viscosity of melts and the low density contrast between melt and crystals [Brandeis  
180 and Jaupart, 1986]. However, gravity compaction and expulsion of liquid may be  
181 encountered in the crystal-rich mush formed atop of solidification fronts. Also, the  
182 interstitial melt trapped in the mush may undergo water saturation leading to boiling and

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183 vesiculation that may force deformation and expulsion of the liquid from a crystalline  
 184 aggregate. The two processes, gravitational collapse and fluid-assisted filter pressing are  
 185 analyzed here.

186 3.1 Gravitational Collapse and Compaction

187 In spite of the scarce theoretical support to an efficient process of crystal–liquid  
 188 separation that allows large volumes of silicic magmas to be available in the continental  
 189 crust in short periods of time [*Bachmann and Huber, 2018*], it is a fact that fractionation  
 190 is identified on the basis of geochemical nearly-cotectic trends [*Castro, 2013; Castro,*  
 191 *2019*], continuous variations in zoned intrusions and large volcanic eruptions of silicic  
 192 (rhyolitic) magmas [*Lipman, 1988*]. The two main approaches to understand the  
 193 mechanisms of silicic magma fractionation, namely analog modeling and experimental,  
 194 have failed to account for the generation of fractionated liquids in the required volumes  
 195 and at the necessary rate, in a time span shorter than the cooling time in the upper crust.  
 196 Mechanical interaction of falling particles (crystals) in silicic magmas can occur even at  
 197 low crystal fractions, leading to a “hindered” settling processes with the implication of  
 198 slow rates of melt extraction [*Bachmann and Bergantz, 2004; Bachmann and Huber,*  
 199 *2018*]. An alternative mechanism is compaction of a crystal-rich matrix, which is  
 200 considered as effective in increasing the rate of melt extraction [*McKenzie, 1984*].  
 201 Compaction is a common phenomenon in long-duration (> 10 days) experimental runs.  
 202 We discuss below evidences from analog materials and high pressure-high temperature  
 203 experiments carried out in presence of temperature gradients. These may shed light on  
 204 the relative importance of gravity settling and compaction in magma chambers.

205 3.1.1 Analog Experiments and Modeling

206 Separation of liquid and crystals in the magma chambers was explained in the  
 207 conceptual and analytical model of *Marsh [1988]* as due to convection beneath the  
 208 *capture front*, defined as the surface separating the rigid crust and crystal mush layers  
 209 (crystallinity larger than 25%) from a crystal suspension zone. According to that model,  
 210 the resulting convection pattern includes crystal-laden plumes falling from the  
 211 suspension zone to the deeper parts of the chamber. The analog experiments of  
 212 *Michioka and Sumita [2005]* simulated a solidifying magma chamber by means of an  
 213 experimental cell consisting of a thin particle layer (glass beads) at the top, overlying a

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214 thick liquid layer (glycerine solution or silicone oil). A limited zone, located at the  
215 interface between the particle-rich and the liquid-rich layers, became unstable, forming  
216 descending plumes, thereby presenting an experimental confirmation of the convection  
217 model of *Marsh* [1988]. The similar analog experiments of *Shibano et al.* [2012]  
218 extended those results to the case of a thick particle-rich layer and found that the  
219 downwelling crystal-laden plumes actually come from a dilated boundary layer located  
220 beneath a granular layer whose particles are in a jammed state. Descent of the plumes  
221 caused a cellular convection pattern within the liquid layer, which eroded the dilated  
222 boundary layer. This mechanism differs from those of compaction, Stokes settling,  
223 hindered settling, and Rayleigh–Taylor instabilities, and permits the upward migration  
224 of the liquid layer, becoming a potentially efficient process of melt transport within  
225 magma chambers. Interestingly, some of the experiments performed by *Shibano et al.*  
226 [2012] do not rule out the activity of permeable flow (compaction and hindered settling)  
227 as a secondary mechanism for liquid transport, particularly when the granular layer  
228 slides downwards as a whole, allowing the generation of a liquid-rich layer at its top.  
229 This mechanism is akin to that described by *Marsh* [2002] to explain the presence of  
230 large silicic lenses in the upper part of mafic intrusions. Finally, *Shibano et al.* [2013]  
231 advanced in the analysis of magma chamber processes simulating roof melting by  
232 means of experimental cells filled with wax and glass beads. Those experiments, which  
233 do not preclude the effects of crystallization within the magma chamber, are able to  
234 explain the generation of rhythmic layering at the bottom of the chamber, and showed  
235 that magma ascent can be a cyclical and intermittent process.

236

## 237 3.1.2 Compaction Experiments at High Pressure–High Temperature

238 Crystal accumulation is commonly observed in experimental capsules in long-duration  
239 runs of several days, overcoming the limitations imposed by the small size of crystals  
240 and the expected high viscosity of silica-rich liquids. This phenomenon allows us to  
241 simulate experimentally the role of gravity compaction and expulsion of an interstitial  
242 liquid from a crystal-rich mush and to compare the results with magmatic differentiation  
243 series. In this way, geological inferences and mechanical analysis on a possible collapse  
244 of the partially crystallized solidification front atop of magma chambers [*Marsh*, 2002],  
245 were confirmed experimentally by *Masotta et al.*, [2012] and contrasted with  
246 petrological relations of mush fragments (crystal-rich enclaves) in volcanic rocks



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247 [Masotta *et al.*, 2016]. Another interesting experiment of crystallization, possibly  
 248 accompanied by compaction, was carried out by Huang *et al.* [2009] using a natural  
 249 andesite (the AGV USGS standard [Flanagan, 1967]) as starting material with added  
 250 water, crystallizing within a thermal gradient of 600 °C. The favored interpretation was  
 251 differentiation by ion migration in response to the thermal gradient [Huang *et al.*, 2009].  
 252 However, the presence of monomineralic crystal layers at the bottom of the capsule  
 253 indicates that compaction was an effective mechanism of differentiation by crystal–  
 254 liquid separation in that experiment. It is interesting to mention that experiments with a  
 255 dry andesite in a thermal gradient (ca. 300 °C) produced no significant differences in  
 256 composition along the capsule, while marked differences were found in capsules with  
 257 the same starting material with added water [Rodríguez and Castro, 2017]. Thus, the  
 258 cause of element fractionation must be found in the presence of water and not in the  
 259 thermal gradient. One of the experiemnts reported by Rodríguez and Castro [2017] was  
 260 carried out with the above mentioned AGV andesite within a 30°C/mm gradient at 5  
 261 kbar in the classical vertical position of the piston–cylinder. In this arrangement,  
 262 identical to the experiment by Huang *et al.* [2009], the thermal gradient within the  
 263 capsule acts in the same direction than gravity. This vertical experiment was carried out  
 264 as a benchmark run (Run CRV2; Rodríguez and Castro [2017]) to compare with  
 265 horizontally arranged runs of the same study in which, thermal gradient and gravity are  
 266 orthogonal (see next section). After comparison, fractionation was less effective  
 267 compared with horizontal runs as crystal–liquid separation is controlled by gravity  
 268 compaction and not by exsolution of a strongly fractionated (rich in silica and alkalis)  
 269 fluid phase within the solidification front (see below). Nevertheless, we found  
 270 interesting relations in that vertical run that merit the attention here. By contrast with the  
 271 other thermal gradient experiments, in which temperature remains constant at the hot  
 272 spot, in the run CRV2 [Rodríguez and Castro, 2017] a dynamic thermal gradient is  
 273 imposed following a programmed cooling ramp of 0.6 °C/hour, representing a more  
 274 realistic scenario of a cooling magma chamber or dike. A half polished section of the  
 275 vertical run CRV2 is compared with a horizontal run crystallized under identical  
 276 conditions but with the gravity vector arranged orthogonal to the gradient temperature  
 277 vector (Fig. 2). It can be observed that crystals are mostly concentrated at the bottom of  
 278 the capsule (Fig. 2b) compared with the horizontal run (Fig. 2a). Because both  
 279 experiments were set at the same initial conditions and both were slowly cooled at the  
 280 rate 0.6 °C/hour during 309 hours, the only explanation for the observed differences is

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281 compaction and liquid expulsion from the cumulate in the vertical run. Moreover, a thin  
282 monomineralic carapace of Amp, the *liquidus* phase of this water-rich system, is broken  
283 and collapses down leaving free space near the walls allowing interstitial liquid to scape  
284 upwards (Fig. 2e). The upper layer, containing tiny magnetite crystals, is possibly  
285 formed during intrusion of upwards moving liquid plumes. The composition of glass  
286 (quenched melt) along the capsule is fractionated. In the crystal-free zone, glasses are  
287 richer in SiO<sub>2</sub> and K<sub>2</sub>O, and poorer in CaO (Fig. 2d) compared with the original  
288 composition of the AGV andesite [Rodríguez and Castro, 2017]. The constant  
289 composition of glasses in the crystal-free zone, within a strong thermal gradient of ~30  
290 °C/mm (see green curve in Fig. 2b) precludes a Soret effect [Huang et al., 2009] as the  
291 cause of liquid fractionation. By contrast, these results reinforce the role of gravitational  
292 instability as an efficient mechanism to produce liquid–crystal separation. In summary,  
293 application of a dynamic thermal gradient enhances fractionation of the bulk magma  
294 system accompanied by compaction of the crystal-rich mush formed at the solidification  
295 front. Moreover, the results of other compaction experiments [Huang et al., 2009;  
296 Masotta et al., 2016] are totally comparable as they are characterized by expulsion from  
297 the mush zone (i.e., the side walls of conduits and/or magma chambers) of a  
298 fractionated liquid that mixed with the pristine liquid ahead of the front leading to  
299 fractionation of the whole system.

## 300 3.2 Crystallization in a Vertical (Non-Gravitational) Thermal Boundary Layer (TBL)

301 The mechanism of compaction and gravitational collapse of a crystal-rich mush requires  
302 that the thermal gradient that creates the solidification front is closely parallel to the  
303 gravity vector. This condition is satisfied in both the roof and bottom of magma  
304 chambers for which, most mechanical models have been developed [Bachmann and  
305 Bergantz, 2004; Lake, 2013; Marsh, 2002]. However, the processes of magma  
306 crystallization at the vertical walls of magma chambers and the sidewalls of ascent  
307 conduits have received less attention [Humphreys and Holness, 2010; Namur et al.,  
308 2013]. The case of vertical conduits are relevant as most intermediate magmas (e.g.,  
309 calc-alkaline batholiths) that feed plutons at the upper crust have traveled tens of km  
310 from the source region of melt segregation at the lower crust or the upper mantle. In  
311 case of horizontal thermal gradients, as the sidewalls of conduits, the alternative to  
312 hindered settling and gravitational collapse is liquid expulsion by *gas-driven filter*  
313 *pressing* [Pistone et al., 2015].

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314 3.2.1 Effects of Volatile Exsolution

315 *Rodríguez and Castro* [2017] demonstrated experimentally that *gas-driven filter*  
 316 *pressing* is a mechanism able to operate in a water-bearing magma crystallizing in a  
 317 thermal boundary layer (TBL) in which, a continuous variation in the crystal fraction or  
 318 crystallinity ( $X_c = \text{crystals}/\text{crystals} + \text{liquid}$ ) from all-solid ( $X_c = 1$ ) to all-liquid ( $X_c = 0$ ) is  
 319 established. Other properties, as magma viscosity and strength, also changes across the  
 320 TBL, making the walls of magma conduits and chambers places of special relevance in  
 321 the generation of textural heterogeneities in magmas [*Fernández and Castro*, 2018]. In  
 322 this section, only the petrological consequences of gas expulsion and vesiculation by  
 323 second boiling are explored on the basis of laboratory experiments [*Rodríguez and*  
 324 *Castro*, 2017].

325 The principle is that any water-bearing liquid, the general case of calc-alkaline magma  
 326 systems, will reach saturation by second boiling in the course of crystallization, in the  
 327 way as water is partitioned into the remaining liquid, reaching saturation at a given state  
 328 of crystallinity. The value of  $X_c$  for boiling depends on the initial water content (in wt%  
 329  $H_2O$ ) of the magma ( $W_0$ ) and the water solubility at the pressure of crystallization  
 330  $[W_s]_{(P)}$ . The fraction of water-saturated liquid ( $X_{sl}$ ) is given by the expression:

331  
 332 
$$X_{sl} = W_0 / [W_s]_{(P)} \tag{1}$$

333  
 334 Water solubility is strongly dependent on pressure and weakly on temperature  
 335 [*Burnham*, 1979]. A relation between pressure and solubility is obtained by second  
 336 order polynomial regression of the Burnham's solubility curve for granite liquids  
 337 [*Castro*, 2013]:

338  
 339 
$$W_s = - 0.27 P^2 + 3.54P + 0.42; \text{ (for } P < 6 \text{ kbar)} \tag{2}$$

340  
 341 By substitution in Eq. (1) we get the empirical relation:

342  
 343 
$$X_{sl} = W_0 / - 0.27 P^2 + 3.54P + 0.42; \text{ (for } P < 6 \text{ kbar)} \tag{3}$$

344  
 345 From this relation it is possible to know the critical crystallinity ( $X_{cc} = 1 - X_{sl}$ ); That is,  
 346 the crystal fraction at which the remaining liquid reaches saturation. The value of  $X_{cc}$

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347 depends on the pressure of crystallization and the initial water content of magmas (Fig.  
348 3). The effectiveness of boiling and vesiculation in promoting liquid expulsion and  
349 fractionation depends on the rheological state of the partially crystallized magma at the  
350 time of water saturation. At relatively high crystallinity ( $X_{cc} > 0.7$ ), deformation of the  
351 rigid crust is impeded by the crystal interlocking structure of the magma. Formation of  
352 tension gashes can be formed at this state [Fernández and Castro, 2018]. Many aplites  
353 and pegmatites in granite plutons are true *degassing* structures. These form dikes and  
354 irregular pods in which the contacts with the host granite are at the scale of crystals,  
355 denoting that the host was a crystal-rich magma, and not a solid rock, at the time of  
356 fracturing and fluid segregation. It can be expected that fluids expelled out via fractures  
357 (dikes) from the rigid crust of a solidification front, will carry strongly fractionated  
358 components that may mix and/or dissolve into the liquid-rich area ahead of the  
359 saturation front. Such a mechanism of “fluid migration” is very efficient in granitic  
360 magmas giving rise to zoned intrusions and fractionated cupolas atop of plutons. This  
361 principle is the basis for crystallization experiments of a water-bearing magma in a  
362 thermal gradient (see below).

363 Depending on the initial water content and the pressure of crystallization, the magma  
364 can reach water saturation at varied crystal contents and, thus, varied rheological states  
365 within the saturation front can be found [Rodríguez and Castro, 2017; Fernández and  
366 Castro, 2018]. The most favorable state is that of a deformable mush in which the  
367 formation of bubbles can push liquid away of the crystal framework by promoting  
368 compaction of the crystal aggregate [cf. Bachmann and Huber, 2018]. In the case of the  
369 sidewalls of conduits, shear deformation of the solidification front may favor liquid  
370 expulsion by compaction of the mush. Many flow structures with high concentration of  
371 K-feldspar crystals can be explained by this mechanism. Outside the mush zone, in the  
372 suspension zone ( $X_c < 0.25$ ), flow is controlled by the liquid phase as crystals are  
373 “floating” in the liquid with scarce mechanical interactions. Moreover, water saturation  
374 can only be reached in the suspension zone in anomalous cases of high initial water  
375 contents and very low pressure (Fig. 3). For a magma to reach saturation at  $X_c < 0.5$  at  
376 the pressure of 3 kbar, the initial water content must be higher than 6 wt% H<sub>2</sub>O.  
377 The most favorable mechanical conditions for water saturation and vesiculation to  
378 effectively promote the expulsion of liquid from the crystalline framework, are found  
379 within the rigid crust ( $X_{cc} > 0.55$ ), within a critical zone of  $X_{cc}$  from 0.6 to 0.7, for  
380 magmas with initial water content  $W_0 > 3$  wt% [Pistone et al., 2015]. These critical

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381 conditions can be encountered within a wide range of pressure from 3 to 6 kbar (Fig. 3).  
 382 For shallower conditions ( $P < 2$  kbar), low initial water contents ( $W_0 < 3$  wt%) are  
 383 required to get  $X_{cc}$  within the critical zone of rigid crust of a solidification front. In sum,  
 384 the rheological state of the magma must be deformable at the time of melt water  
 385 saturation to promote liquid expulsion. At values of  $X_{cc} > 0.7$  the rigid crust can  
 386 experience hydraulic fracturing and segregation of a water-saturated melt.

387 3.2.2 Magma Splitting in a TBL: An Experimental Finding with Implications

388 The sidewalls of magma ascent conduits represent large transient interfaces along which  
 389 magma loses heat to the surrounding host with the consequent generation of thermal  
 390 boundary layers (TBL) along several tens km from the region of magma segregation to  
 391 the final level of emplacement. It is inferred that these large TBL structures play an  
 392 important role in magma differentiation during ascent; particularly in the case of water-  
 393 bearing magmas as water saturation will be encountered within the TBL. All  
 394 phenomena referred to above in the previous section can operate along the conduits.  
 395 The main inference for differentiation in TBL at the conduits comes from the presence  
 396 of autoliths in granite (*sensu lato*) plutons. It has been demonstrated that most mafic  
 397 microgranular enclaves characterizing calc-alkaline batholiths are true autoliths and not  
 398 fragments from synplutonic dikes [Paterson *et al.*, 2016; Žák and Paterson, 2010].  
 399 Autoliths represent eroded fragments from different parts of the TBL (chilled margins)  
 400 of conduits. All textural observations in autoliths, as the fine-grain size, the presence of  
 401 resorbed crystals of plagioclase, the presence of double enclaves, together with the  
 402 observed geochemical and isotopic features [Rodríguez and Castro, 2019], are  
 403 supporting such an interpretation. A mechanical analysis of the rheology of conduits at  
 404 the TBL accounts satisfactorily for the observed field relations, shape and size of  
 405 autoliths [Fernández and Castro, 2018].  
 406 Rodríguez and Castro [2017] carried out experiments to simulate the crystallization of a  
 407 water-bearing magma in a vertical TBL representing the sidewalls of conduits. The  
 408 results constituted a significant finding: The liquid ahead of the solidification front is  
 409 fractionated only if water is present as an initially dissolved phase in the magma  
 410 [Rodríguez and Castro, 2017]. Under identical conditions, runs with dry compositions  
 411 produced no differentiation effect on the liquid phase. Other interesting result is the  
 412 sharp boundary between the crystalline zone (the solidification front) and the liquid.  
 413 The consequence is that a water-bearing magma splits into two systems with a

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414 compositional jump. One system is a differentiated liquid and the other is a crystal-rich  
 415 mush (Fig. 4). The latter is comparable to natural autoliths [*Rodríguez and Castro,*  
 416 2019].

417 In sum, differentiation in a TBL is interpreted as the result of liquid expulsion from the  
 418 solidification front in the course of crystallization and water saturation. The liquid ahead  
 419 of the solidification front is modified by two combined phenomena, namely the  
 420 expulsion of a water-saturated liquid and the arrival of fluids released by boiling and  
 421 vesiculation. Because the system under study is a high-silica andesite (the AGV  
 422 standard), the residual water-saturated liquid has the minimum composition of the  
 423 granite system. This residual melt will be mixed with the pristine liquid ahead of the  
 424 TBL leading to its fractionation. The change in the composition of the system has been  
 425 modeled by using the general equation for *in-situ crystallization* [*Langmuir, 1989*]:

426

$$427 \quad C_M = C_0 F^{(f_A(E-1)/(f_A-1))} \quad (4)$$

428

429 Where  $C_0$  is the initial magma composition (in this case the standard AGV andesite),  $F$   
 430 is the fraction of melt (liquid / liquid + crystals),  $f_A$  is the fraction of liquid returned to  
 431 the magma from the solidification front, and  $E$  is the partitioning coefficient, in this case  
 432 taken as the ratio of the composition of the saturated liquid in the element of reference  
 433 ( $C_{SL}$ ) to the composition of magma in the same element of reference ( $C_M$ ). The  
 434 composition at any distance from the wall, after separation of the cumulate, requires  
 435 integration over discrete increments of magma crystallization in which,  $F$ ,  $E$  and  $f_A$  must  
 436 be recalculated for every increment ( $\Delta$ ) of magma crystallization. We have introduced a  
 437 restriction in the equation to calculate the amount of solid fraction at which the  
 438 intercumulus liquid is expelled from the crystal-rich mush. That solid fraction ( $1-F$ ), or  
 439 cumulate, at every discrete increment ( $\Delta$ ) is determined by the fraction of saturated  
 440 liquid ( $X_{sl}$ ), which is dictated by the ratio of the water content of magma ( $W_0$ ) to the  
 441 water content at saturation (Eq. 1). An iterative calculation at fixed increments of a unit  
 442 volume of magma allows us to know the composition of the modified liquid ahead of  
 443 the solidification front and, by simple mass balance, the composition of the solid  
 444 residue. A plot of silica content of the modified liquid versus the fraction of remaining  
 445 magma is shown in Fig. 5 for three values of initial water content ( $W_0$ ) of 2, 3 and 5  
 446 wt% H<sub>2</sub>O and at pressure of 5 kbar. Because water saturation is key in determining the  
 447 fraction of saturated liquid that is available to modified the pristine liquid, it is clear that

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448 the effect of fractionation is higher for systems containing the higher initial content of  
 449 water ( $W_0$ ). For values of  $W_0 = 5$  the modified liquid reaches minimum silica values  
 450 ( $\text{SiO}_2 > 63$  wt%) of granitic rocks when only 20% of the whole magma is crystalized.  
 451 However, magma with initial water content  $W_0 = 3$  must crystalize about 50% of its  
 452 initial volume to produce a fractionated liquid with  $\text{SiO}_2 > 63$  wt%. Interestingly, the  
 453 composition of cumulates remains almost constant compared with the continuous  
 454 changing compositions of liquids. In nature, cumulates are represented by the fine-  
 455 grained microgranular enclaves interpreted as autoliths, whose composition is very  
 456 uniform within a particular pluton. Also notice that the most fractionated magma ( $W_0 =$   
 457 5) yields the most mafic (less silicic) cumulates.

458 **4 Magma Contamination by Country-Rock Assimilation**

459 Contamination can be seen as an open-system mechanism of magma differentiation in  
 460 the crust; as in the most general case contaminants are richer in evolved components, as  
 461 alkali elements and silica, than the pristine magmas. Contamination may proceed in two  
 462 ways that are not exclusive each other: (1) Assimilation of country rocks and (2)  
 463 contamination by fluids derived from the host. Although contamination can be traced by  
 464 means of isotopic ratios, source contamination can be difficult to distinguish from  
 465 crustal-related processes. Fluids released by prograde metamorphic reactions in the  
 466 thermal aureoles of plutons may expectedly contaminate the intruding magmas.  
 467 However, this process is rarely identified in plutonic intrusions. Assimilation is the most  
 468 documented process in crustal intrusions. Partial or total digestion of country rocks by  
 469 the intruding magma is intrinsic to assimilation as an efficient mechanism that  
 470 contributes to the contamination of magmas in the continental crust.  
 471 Although contamination by country-rock assimilation can be identified in geochemical  
 472 diagrams, the intricacies of the process can be varied. The study of field relations in  
 473 plutons is essential to constrain possible mechanisms. Experimental studies on magma-  
 474 rock interactions are also relevant to test reactions and mineralogical implications and to  
 475 assess the feasibility of assimilation on a regional scale. The two ways, field relations  
 476 and experiments, are explored here on the basis of recently developed studies.

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## 477 4.1 Field Relations Supporting Assimilation and Magma Contamination

478 Contamination of magmas with their host rocks can be identified easily by means of  
479 geochemical and isotopic relations. Paradoxically, although intermediate and silicic  
480 magmas have travelled long distances through the crust until their final storage in  
481 magma chambers or plutons and volcanic eruptions, not all silicic rocks are  
482 contaminated in the same extent and many of them maintain their pristine composition.  
483 There will be scenarios in which magma-host interactions are favored. Assimilation not  
484 only depends on the reactivity between magma and country rocks but on the  
485 dynamics of ascent and emplacement and the rheological behavior of the two systems.  
486 An approach to the mechanical processes leading to assimilation of country rocks, and  
487 the consequent contamination of intruding magmas, can be made on the basis of field  
488 relations in areas of intense assimilation. A summary of these relations, based on  
489 relevant exposures of hybrid granites from the Gredos batholith (Central Spain), is  
490 shown in this section.

491 The Gredos batholith has been revealed during the last decade as one of the most  
492 outstanding and voluminous granitic exposures (more than 300 km in length and 60 km  
493 in width) to explore the interactions between intrusive magmas and a medium- to high-  
494 grade crustal sections. The batholith is mainly composed of  $Bt \pm Crd$  granodiorites and  
495 monzogranites and minor amounts of basic rocks [Scarrow *et al.*, 2009] that depict a K-  
496 rich calc-alkaline suite characteristic of I-type post-collisional batholiths [Castro, 2019],  
497 emplaced during late D2 and D3 Variscan phases (320-290 Ma) [Díaz Alvarado *et al.*,  
498 2013; Díaz-Alvarado *et al.*, 2011]. Detailed studies of the central area of the batholith  
499 have evidenced a laminar structure formed by mostly migmatitic host-rocks and  
500 intrusive layers. The conspicuous magmatic fabrics (foliations, lineations, folds and  
501 shear zones) are continued through the migmatitic structure of the host-rocks, which  
502 involve the synkinematic and sequential emplacement of intrusive magmas assisted by  
503 crustal-scale extensional shear zones, as have been revealed by structural and U–Pb  
504 geochronological studies [Díaz Alvarado *et al.*, 2013; Díaz-Alvarado *et al.*, 2012].  
505 These characteristics of the emplacement process reinforced the long-lasting and close  
506 interaction between magmas and partially melted host-rocks that promoted the intense  
507 mingling and, finally, the chemical hybridization between both systems.

508 Geochemical, field and experimental evidences have shown that the hybridization  
509 between an intrusive magma and its host rock is an effective mechanism of magma



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510 diversification during its ascent and emplacement [*Beard et al.*, 2005; *Díaz–Alvarado et al.*, 2011; *Erdmann et al.*, 2007]. The high diffusivity of some elements (e.g., the alkali  
 511 elements) may favor the crystallization in the magma of particular phases, like Kfs, in  
 512 areas far from the external contacts of the pluton with the host rocks [*Díaz–Alvarado*,  
 513 2017; *London et al.*, 2012]. However, Al-rich phases present in calc-alkaline  
 514 granodiorites, like Crd and Grt, are diagnostic of assimilation processes and only appear  
 515 in reactive domains where metasedimentary xenoliths were consumed or at least they  
 516 lost their integrity (Fig. 6). In those cases, the original mineralogy of the xenoliths has  
 517 been in part consumed by peritectic melting reactions to yield residual and peritectic  
 518 minerals plus a granite melt, and in part is dispersed and camouflaged within the host  
 519 magma. The only mineral species that can be considered exotic, in the sense that they  
 520 are not present in the pristine magma, are the peritectic phases, cordierite or garnet. The  
 521 amount of assimilated material in hybrid granites can be estimated by mass balance  
 522 using the fraction of Crd [*Díaz–Alvarado et al.*, 2011; *Erdmann et al.*, 2007].  
 523 The main conditioning factor for this process is the effectiveness of the heterogeneous  
 524 interaction process between the partially crystallized magma and the partially molten  
 525 metasedimentary host rocks. The rheological characteristics of both systems tend to  
 526 approach during the emplacement process, that is, while the migmatitic system  
 527 increases its melt percentage and the magma continues its crystallization process (e.g.  
 528 [*Vignerresse et al.*, 1996]. The driving forces of the emplacement process, including the  
 529 stress state and the tectonic evolution of the crust, trigger the joint flow and deformation  
 530 of the two systems, which yields a number of heterogeneous structures at all the scales  
 531 [e.g. *Paterson et al.*, 2018; *Paterson et al.*, 1998], that are evidenced by the observed  
 532 field relations (Fig. 7). Those structures can be ascribed to the following mechanisms of  
 533 mechanical interaction between intruding magma and host rock: (1) Viscous folding; (2)  
 534 Host-rock dragging; and (3) Migmatitic tearing apart. These structures can be ascribed  
 535 to the following mechanisms of mechanical interaction between intruding magma and  
 536 host rock: (1) Viscous folding (Fig. 7a–d); (2) Host-rock dragging (Fig. 7e); and (3)  
 537 Migmatitic tearing apart (Fig. 7f).  
 538

539 4.1.1 Viscous Folding and Shearing

540 Complex fold geometries and distinct types of brittle and ductile shear zones are the  
 541 most common structural features in migmatitic terrains [e.g. *Hopgood*, 1999]. In the  
 542 case of a migmatitic crust intruded by a partially crystallized magma whose viscosities

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543 approach during the evolution of both systems, the development of ductile deformation  
544 structures such as folds and shears considerably increases the contact surface between  
545 both systems and the isolation of host rock fragments within the intrusive magma (Fig.  
546 7a, b). This process has been shown as a necessary condition for the complete  
547 hybridization that results in contaminated magma [*Gogoi and Saikia, 2018*].  
548 Assimilation through reaction of host-rock fragments implies the successive injection of  
549 low crystal fraction magma batches, simultaneous development of assorted structures  
550 under a viscous but evolutionary regime, melting reactions and chemical diffusion.  
551 Therefore, deformation of the complex, composite system may be achieved through a  
552 mechanism of viscous folding, which implies a viscosity contrast between magma and  
553 host rock bodies (Fig. 7c, d) [e.g., *Biot, 1961; Chapple, 1968; Johnson and Fletcher,*  
554 *1994; Ramberg, 1961*]. It is expected a viscosity switch along the interaction process,  
555 such that the less viscous unit at the very beginning of the interaction process (i.e., the  
556 intruding magma) becomes the more viscous one as it crystallizes, generating a very  
557 complex and heterogeneous set of folding structures. This is evidenced by the complex  
558 arrangement of Kfs megacryst fabrics that results firstly from the flow and interaction of  
559 crystals in a fluid flow and the subsequent orientation of the same rigid particles  
560 according to the contacts and the stress regime in a highly crystallized magma (Fig. 7a,  
561 b).

## 562 4.2 Experiments on Contamination

563 One of the most outstanding criteria to identify a process of assimilation is the presence  
564 of the peritectic phase Crd and/or Grt, which formed by fluid-absent (dehydration)  
565 partial melting of pelitic metasediments, in a non-anatectic granodiorite or  
566 monzogranite. A proof that Crd is not in equilibrium is such Ca-rich granite magma  
567 composition is supplied by experiments with a Crd-bearing monzogranites, as these  
568 failed to reproduce the Crd-bearing assemblage observed in nature [*García-Moreno et*  
569 *al., 2017*]. These experiments were performed using a synthetic glass with the  
570 composition of a Crd-bearing peraluminous monzogranite of the Iberian Massif. This is  
571 the Cabeza de Araya granite, whose composition is taken as representative of the so-  
572 called “mixed granites” [*Capdevila et al., 1973*], characterized by sharing features of  
573 typical anatectic granites (S-type) and Bt-granodiorites (I-type). Crd-bearing  
574 monzogranites appear in the Variscan belt of Iberia as isolated intrusions or as large  
575 irregular domains inside calc-alkaline granodiorite batholiths. The origin of these

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576 “atypical” granitic series has attracted much attention from petrologists over decades.  
 577 We contend on the basis of geochemical, geological and experimental grounds that they  
 578 are the products of crustal contamination by pelitic and semipelitic host rocks. In the  
 579 case of Cabeza de Araya intrusion [Corretgé, 1971], the Crd-bearing monzogranites are  
 580 located at the margins of the pluton. These represent the less evolved rocks that  
 581 gradually transition into the central parts composed of two-mica granites and aplitic  
 582 leucogranites. Emplacement age of the Cabeza de Araya granites obtained by SHRIMP  
 583 lies between  $308 \pm 1.5$  Ma and  $305 \pm 2$  Ma for the different facies that compose the  
 584 batholith [Rubio–Ordóñez *et al.*, 2016].  
 585 Large prismatic crystals (1 to 4 cm) of cordierite (Crd) are the most distinguishing  
 586 feature of these “mixed” granites. The presence of Crd in this kind of granites has been  
 587 interpreted as result of a peritectic reaction in the local domain of the xenoliths after  
 588 wall-rock assimilation [García–Moreno *et al.*, 2017]. Interestingly, the outer zone of the  
 589 pluton, in contact with the pelitic metasedimentary host, is the richer in Crd. The  
 590 abundance of mafic microgranular enclaves (autoliths) and xenoliths is also greater in  
 591 the margin zones compared to the inner parts. The inferences from field relations were  
 592 tested with varied experimental designs using that and similar compositions.  
 593 Experimental approaches to test contamination processes are rooted in field and  
 594 geochemical relationships in large composite batholiths, which point to physical and  
 595 chemical interactions between the intruding magmas and its host rocks during ascent  
 596 and emplacement [e.g. Beard *et al.*, 2005; Díaz–Alvarado *et al.*, 2011; Erdmann *et al.*,  
 597 2007; London *et al.*, 2012]. Different experimental procedures and strategies have led to  
 598 relevant conclusions about diffusion and reactions between both subsystems.

599 4.2.1 Selective Assimilation Experiments

600 Experimental simulations in granodioritic and monzogranitic systems have shown that  
 601 Crd or Grt do not precipitate during crystallization sequences, even when synthetic  
 602 starting materials representing the whole composition of Crd- or Grt-bearing granitic  
 603 rocks are used [Díaz–Alvarado, 2017; García–Moreno *et al.*, 2017]. The addition of  
 604 aluminous phases to experimental capsules simulates the usual presence of And-, Sill-  
 605 or/and Crd-rich restites in the migmatitic contact zones of intrusive bodies [Acosta–  
 606 Vigil *et al.*, 2002; Díaz–Alvarado *et al.*, 2011]. Local domains are observed around  
 607 xenocrysts in doped experiments, resembling reactive zones of high Al activity in melts  
 608 around crystals or along layered contacts [Acosta–Vigil *et al.*, 2002; Díaz–Alvarado *et*

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609 *al.*, 2011; *García–Moreno et al.*, 2017], but far from the conditions expected for the  
610 massive crystallization of large euhedral diagnostic phases as Kfs and Grt or Crd. The  
611 dissolution of exotic phases is governed by the mineral reaction rates and diffusion  
612 through the melt, besides other conditions as H<sub>2</sub>O content and convection [*Acosta–Vigil*  
613 *et al.*, 2002, 2006]. Nonetheless, the above mentioned experiments show the resilience  
614 of exotic xenocrysts in the intrusive magmas and the presence of local reactive domains.

## 615 4.2.2 Layered Experiments

616 Reaction at the interface between metasedimentary rocks and granitic melt  
617 (granodioritic or haplogranitic depending on the experimental study) indicate that  
618 homogenization took place for particular components as K, Na and H<sub>2</sub>O between partial  
619 melts at both sides of the interface [*Díaz–Alvarado*, 2017; *Erdmann et al.*, 2007;  
620 *London et al.*, 2012]. The melt percentage increases in the pelitic system as it shifts to a  
621 more haplogranitic composition (Erdmann, London op cit.). Two-layer experiments  
622 represent static situations, being the crystallization of diagnostic phases restricted to a  
623 narrow zone close to contact [*Erdmann et al.*, 2007; *London et al.*, 2012]. However, the  
624 application of these results to the dynamic scenario of an ascent conduit, in which the  
625 narrow zone of contaminated magma is continuously removed by flow, contamination  
626 can be effective for large volumes of magma feeding an upper reservoir or pluton.  
627 Contaminated granites can occupy large areas of zoned plutons. The process can demise  
628 with time as the later magma pulses use the core of conduits and are prevented of  
629 contamination. This, combined with increasing fractionation in conduits by  
630 crystallization in a TBL (see above), can be a plausible explanation to many zoned  
631 plutons in which the most contaminated and most mafic granites are disposed at the  
632 outer rims, and the less contaminated and more felsic types are at the core.

## 633 4.2.3 Bulk-Assimilation Experiments

634 Bulk assimilation [*Beard et al.*, 2005] has been reproduced experimentally by  
635 introduction of pelitic fragments into a granodiorite powder (Fig. 8) [*Díaz–Alvarado et*  
636 *al.*, 2011], with significant implications for the linkage of this assimilation  
637 mechanism with geochemical and mineralogical changes observed in large  
638 batholiths [*Díaz–Alvarado et al.*, 2011; *Saito et al.*, 2007]. Partially disintegrated  
639 xenoliths are still recognizable in the experimental runs (Fig. 8). Partial melts inside and

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640 far from the xenolith domain still have important compositional differences (Al, Mg#)  
 641 except for the alkalis, expelled from the xenoliths towards the granodioritic host, with  
 642 the consequent enrichment in K and the early crystallization of Kfs, denoting a sort of  
 643 mixing between the granodioritic and the xenolith-derived melts [e.g. *Díaz-Alvarado*,  
 644 2017; *Massota et al.*, 2018]. As in the layered experiments, euhedral Crd and Kfs are  
 645 crystallized along the reactive xenolith area [*Díaz-Alvarado et al.*, 2011]. A dynamic  
 646 scenario, as it was mentioned above, may contribute to disaggregation  
 647 of xenoliths, leading to total digestion and dissemination of minerals within the  
 648 intruding granite. Xenocrystic Pl, Bt or Qz are camouflaged in the contaminated  
 649 magma, the euhedral Crd (Fig. 8) remaining as the only diagnostic mineral of the bulk  
 650 assimilation, as described in natural examples (Fig. 6). However, a rapid segregation of  
 651 interstitial melts in the crustal xenoliths may inhibit mineral–melt equilibrium and  
 652 prompt the zonation of residual minerals [*Massota et al.*, 2018].  
 653 Although a significant percentage of the assimilated material gained by the  
 654 contaminated magma is unrecognizable, it is possible to assess the extend of  
 655 assimilation by measuring the abundance of Crd in the contaminated granites [*Díaz-*  
 656 *Alvarado et al.*, 2011]. The results show that the assimilated material is approximately  
 657 five times the proportion of Crd. This figure depends on the amount of pelitic  
 658 components (Al, Fe, Mg) of the contaminant; the more pelitic the less fraction of  
 659 contaminant in the final hybrid rock. Mass balance calculations and other  
 660 approximations through Sr–Nd isotopic ratios are in agreement with these results  
 661 [*Clarke et al.*, 2004; *Díaz-Alvarado et al.*, 2011; *Erdmann et al.*, 2007; *Fowler et al.*,  
 662 2001; *Ugidos and Recio*, 1993].  
 663 Batholithic examples, as the Gredos batholith (Iberian Massif), show that hybrid  
 664 magmas may contain between 50% and 10% of assimilated material, depending on the  
 665 proximity to the metasedimentary host, which fits well with the volume of Crd and Kfs  
 666 estimated in the contaminated rocks [*Díaz-Alvarado et al.*, 2011]. The layered structure  
 667 of the batholith and the coherent and tectonically induced viscous deformation of  
 668 intrusive magmas and migmatitic host-rocks favored the increase of the contact surfaces  
 669 between both subsystems, which has been proven essential for the efficacy of bulk  
 670 assimilation. Besides, the sequential character of the emplacement process involves a  
 671 long-lasting high-grade area in the host crust. The similar crystallization ages obtained  
 672 from intrusive magmas and anatectic leucogranites [*Díaz Alvarado et al.*, 2013] formed  
 673 and locally segregated in the migmatitic host-rocks, imply that both the intrusive

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674 magmas and partially melted metasediments sustain a similar, albeit changing,  
675 rheologic state during their heterogeneous and intense interaction, triggering the  
676 geochemical and mineralogical changes that are characteristic of the assimilation  
677 process and similar to the experimentally proved conditions that favor magma mixing  
678 [Laumonier *et al.*, 2014a, b].

**679 5 Concluding remarks**

680 Differentiation and contamination are common processes in continental environments.  
681 On a large extent the fractionated character of the continental crust with respect to the  
682 underlying mantle is in sum the result of a protracted process of combined  
683 differentiation and contamination. The latter is particularly relevant if available  
684 contaminants are terrigenous metasediments, as these represent substantial geochemical  
685 fractionation imposed by surface weathering. In most cases isotopic relations are good  
686 indicators to distinguish between fractionation (closed systems) and contamination  
687 (open systems). However, understanding the mechanisms that lead to magmas to  
688 fractionate and/or to assimilate portions of country rocks, requires a deep knowledge of  
689 complex magma systems. Two approaches, experimental and geological, have been  
690 used in this paper to address the problem.

691 Field evidences from the Gredos batholith (Central Spain) support that assimilation of  
692 pelitic metasediments caused the formation of Crd in local domains of the intrusive  
693 granodiorite (calc-alkaline) magmas. Partial digestion of pelitic migmatites is common  
694 at the contacts, where Crd formed by peritectic melting reactions in the pelites in the  
695 course of xenolith disaggregation. These reactions are confirmed by means of  
696 laboratory experiments using magma–pelite heterogeneous systems at conditions of  
697 granodiorite emplacement of 850 °C and 4 kbar. Experiments reported that Crd is not  
698 reproduced otherwise by crystallization of a glass with composition of a Crd-bearing  
699 monzogranite. These results reinforce the idea that Crd in non-anatectic monzogranites  
700 and granodiorites is in equilibrium within local subsystem created by assimilation of  
701 country rock xenoliths. The existence of a thick (>5 km) sequence of Neoproterozoic  
702 pelites and greywackes, as the regional host of Variscan batholiths, is the reason for the  
703 conspicuous presence of Crd in varied types of granites from tonalites to monzogranites.  
704 For the same reason, anatectic leucogranites with primordial (peritectic) Crd are so  
705 abundant in Iberia. Granites emplaced into older igneous, either volcanic or plutonic,

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706 host rocks are less prone to undergo contamination. This is the general case of the  
 707 Cordilleran granite batholiths. In sum, the reactivity of the host is a fundamental  
 708 factor determining the feasibility of assimilation and contamination. Pelites are the most  
 709 reactive systems and the formation of Crd in non-anatectic granites is diagnostic in such  
 710 cases.

711 In regard of differentiation, our experiments in a thermal gradient or thermal boundary  
 712 layer (TBL) are conclusive about the role of dissolved water in the magma in the  
 713 separation of crystal and liquid, a necessary process to account for the origin of rock  
 714 series that are linked to a parental magma by fractionation. Experiments with a natural  
 715 andesite in horizontal capsules, not affected by gravitational processes, produce an  
 716 interesting phenomenon that may help to understand geological and geochemical  
 717 observations. This is we call *splitting*. Basically, a water-bearing magma crystallizing in  
 718 a TBL is broken in two subsystems with a sharp boundary between them. One  
 719 subsystem is formed by a crystal-rich aggregate, whose composition resembles the fine-  
 720 grained microgranular enclaves that commonly appear in calc-alkaline batholiths, and  
 721 the other subsystem is a fractionated liquid. The latter showing a composition that  
 722 resembles that of the calc-alkaline granodiorites and granites. As enclaves are mostly  
 723 autoliths, they represent magmas fragments with high crystal contents that are dragged  
 724 from walls of ascent conduits. In this sense, we contend that conduits may have a  
 725 primordial role to produce magmatic differentiation in the crust. We found that a  
 726 plausible cause for liquid expulsion from the partially crystallized mush at the TBL is  
 727 boiling and vesiculation, as water saturation is necessarily encountered at any point of  
 728 the solidification front generated in a TBL. Vertical experiments, in which the thermal  
 729 gradient and gravity acceleration vector are parallel, yield that crystal settling can be  
 730 impeded by solid particle interactions, but that gravitational collapse of magma mushes  
 731 from the top of the solidification front is possible.

732

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964 **Captions to figures**

965

966 **Figure 1.** Geochemical variation plots, taken as proxies of phase diagrams, depicting  
 967 possible arrays of fractionation and contamination in calc-alkaline magma systems. **(a)**  
 968 The CaO–MgO diagram showing the curved array of experimental cotectic liquids  
 969 (orange dots) and their corresponding solid assemblages (blue hexagons) formed in  
 970 equilibrium at 3 kbar from an andesitic parental magma [*Castro 2013*]. The orange field  
 971 below the cotectic line represents the area of magmas that carry crystals from the  
 972 cumulate. The blue area represents the field crystal mushes that retain a liquid fraction  
 973 after extraction. Many rocks in batholiths plot in the two areas indicating that  
 974 fractionation is not perfect. Also shown as the lines of contamination with  
 975 metasedimentary rocks in case of open systems. **(b)** Projected space in the diagram  
 976 Opx–An–Or showing cotectic lines from experimental liquids at varied conditions of  
 977 pressure and water contents. Rocks of the Gredos batholith in Central Spain are shown  
 978 as an example. These plot in part in the array of fractionation and in part in that of  
 979 assimilation (Modified from *Castro, 2013*]).

980 **Figure 2.** Electronic compositional images (backscattered electrons) and compositional  
 981 relations of a vertically arranged experimental run simulating magma crystallization in a  
 982 thermal gradient at 5 kbar. **(a)** Capsule section from a horizontally arranged run at the  
 983 same conditions of the vertical run **(b)**. The thermal gradient of the assembly is shown  
 984 in **(b)** with a green curve and green diamonds from double thermocouple measurements.  
 985 The dashed curves  $L_0$  in **(b)** represent the liquidus temperature taken from run CRH5  
 986 (See Fig. 4a). **(c)** Phase map of the bottom part of the vertical capsule **(b)**. **(d)**  
 987 Compositional profiles of glasses (quenched liquid) along the vertical capsule. **(e)**  
 988 Interpretation of gravitational collapse of the upper carapace and liquid expulsion at the  
 989 bottom of the vertical capsule. The dashed curve represent the theoretical position of the  
 990 liquidus in the absence of gravity collapse **(a)**.

991 **Figure 3.** Plot of critical crystallinity ( $X_{cc}=1-X_{sl}$ ) versus initial water content ( $W_0$ ) of a  
 992 granitic liquid using Eq. (3) at variable pressures from 3 to 6 kbar. The critical  
 993 crystallinity represents the crystal fraction at which the remaining liquid reaches water  
 994 saturation and boiling. The three zones of the solidification front (suspension, mush and  
 995 rigid crust) are marked using the boundaries given by [*Marsh, 2002*]. The most

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996 favorable conditions to promote crystal–liquid separation by fluid-assisted filter  
 997 pressing according to experiments [*Pistone et al.*, 2015] are also depicted.

998 **Figure 4. (a)** Mosaics of backscattered electron images from two polished sections of  
 999 runs CRH5 and CRH4 with the AGV andesite (Rodríguez and Castro, 2018) using the  
 1000 thermal gradient imposed by the experimental assemblage. Initial conditions are  $P=5$   
 1001 kbar and  $T=1200\text{ }^{\circ}\text{C}$  at the distance 0–3 mm from the thermocouple. CRH5 was  
 1002 quenched after 315 hours at the initial conditions. The liquidus (Cpx) is set  
 1003 approximately at  $980\text{ }^{\circ}\text{C}$ . In CRH4  $T$  was dropped at the rate of  $0.6\text{ }^{\circ}\text{C}/\text{hour}$  during 308  
 1004 hours (until  $1016\text{ }^{\circ}\text{C}$  at the thermocouple). The composition of glass (quenched liquid)  
 1005 at 2 mm of the thermocouple is more fractionated (richer in  $\text{K}_2\text{O}$  and  $\text{SiO}_2$  and poorer  
 1006 in  $\text{CaO}$  and  $\text{MgO}$ ) in CRH4 compared with CRH5, in which no gradient was applied.  
 1007 **(b)** Field photographs of partially dismembered autoliths from the Gredos batholith in  
 1008 Puente del Congosto (Central Spain).

1009 **Figure 5.** Variation in the silica content of liquids (in a unit magma chamber) that are  
 1010 modified by influx of residual water-saturated liquid coming from the solidification  
 1011 front (thermal boundary layer) at the sidewalls. Liquid curves are calculated with  
 1012 Langmuir's equation [*Langmuir, 1989*] for in-situ crystallization (See text for further  
 1013 explanations).

1014 **Figure 6.** Field relations of partially digested pelitic xenoliths enclosed in calc-alkaline  
 1015 monzogranites and granodiorites of the Gredos batholith (Central Spain). **(a)** Large  
 1016 xenolith of partially molten (migmatite) metasediments showing irregular contacts. **(b)**  
 1017 Detail of another xenolith showing the concentration of large Crd crystals (dark dots)  
 1018 around the contacts. **(c)** Sketch in two stages showing the possible digestion of xenoliths  
 1019 by peritectic melting reaction and the formation of Crd (green dots) and Kfs that appear  
 1020 finally disseminated in the contaminated zones. The arrival of K to the pristine  
 1021 granodiorite shifts the composition of the final contaminated magma to monzogranite.

1022 **Figure 7.** Field examples (Gredos massif, Spain) of heterogeneous structures resulting  
 1023 from the interaction processes between a partially crystallized magma and a partially  
 1024 molten metasedimentary host rock. **(a)** to **(d)** are cases illustrating mechanisms of  
 1025 viscous folding and shearing. **(a)** and **(b)** Field photograph and interpretative sketch of  
 1026 complexly interleaved and folded sheets of migmatites, intrusive Bt granodiorite, and  
 1027 hybrid Kfs–Crd monzogranite. **(c)** and **(d)** Coeval folding ( $f$  is the axial trace) of  
 1028 metatexite and granodiorite intrusive sheets. Shear zones are also seen affecting the



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1029 system. **(e)** Sharp contact between the intrusive granodiorite (lower half of the  
 1030 photograph) and the migmatitic host rock (upper half) showing xenolithic fragments  
 1031 dragged by the intruding magma from its host rock. Inset depicts the final result of this  
 1032 process, with disaggregation of the metasedimentary xenoliths (melt + restitic and  
 1033 peritectic phases) within the granodiorite magma. **(f)** Tearing apart of migmatite  
 1034 mesosome from an intruding wedge of granodiorite magma promoted by the formation  
 1035 of a three-dimensional network of interconnected leucosome veins. Inset shows the  
 1036 individualization and disruption of sheets of mesosome and melanosome into the  
 1037 intruding magma. Grd: intrusive granodiorite. Mig: Migmatite. Leu: Leucosome.

1038 **Figure 8.** Electronic compositional images (backscattered electrons) of experimental  
 1039 run products simulating reaction between a pelitic xenoliths and a granodiorite liquid,  
 1040 after *Díaz-Alvarado et al. [2011]*. **(a)** Section of the whole capsule showing the  
 1041 remnants of partially molten and dismembered xenoliths. **(b)**, **(c)** and **(d)** Details of the  
 1042 same run product showing the formation of euhedral Crd and Kfs as peritectic phases.

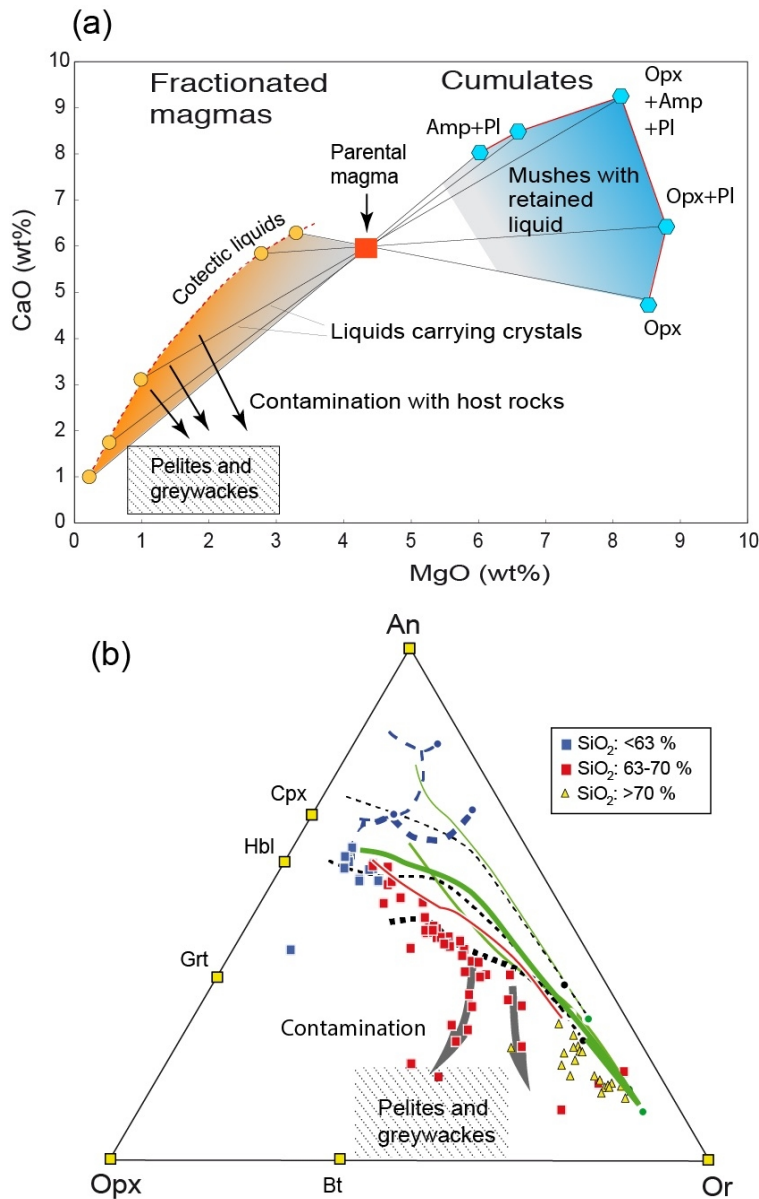


Figure 1 Geochemical variation plots, taken as proxies of phase diagrams, depicting possible arrays of fractionation and contamination in calc-alkaline magma systems. (a) The CaO-MgO diagram showing the curved array of experimental cotectic liquids (orange dots) and their corresponding solid assemblages (blue hexagons) formed in equilibrium at 3 kbar from an andesitic parental magma [Castro 2013]. The orange field below the cotectic line represents the area of magmas that carry crystals from the cumulate. The blue area represents the field crystal mushes that retain a liquid fraction after extraction. Many rocks in batholiths plot in the two areas indicating that fractionation is not perfect. Also shown as the lines of contamination with metasedimentary rocks in case of open systems. (b) Projected space in the diagram Opx-An-Or showing cotectic lines from experimental liquids at varied conditions of pressure and water contents. Rocks of the Gredos batholith in Central Spain are shown as an example. These plot in part in the array of fractionation and in part in that of assimilation (Modified from Castro, 2013)].

142x226mm (150 x 150 DPI)



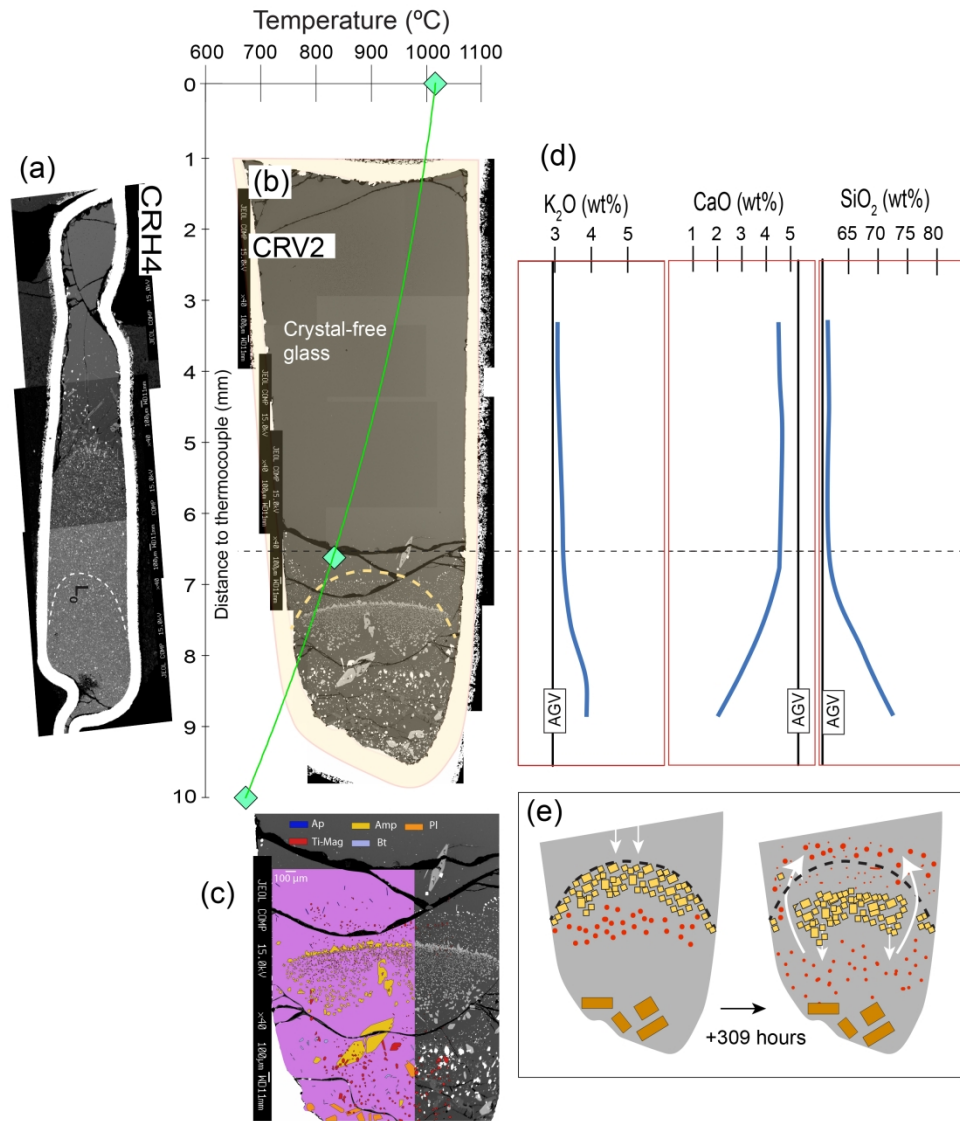


Figure 2 Electronic compositional images (backscattered electrons) and compositional relations of a vertically arranged experimental run simulating magma crystallization in a thermal gradient at 5 kbar [Rodríguez and Castro, 2017]. (a) Capsule section from a horizontally arranged run at the same conditions of the vertical run (b). The thermal gradient of the assembly is shown in (b) with a green curve and green diamonds from double thermocouple measurements. The dashed curves L0 in (b) represent the liquidus temperature taken from run CRH5 (See Fig. 4a). (c) Phase map of the bottom part of the vertical capsule (b). (d) Compositional profiles of glasses (quenched liquid) along the vertical capsule. (e) Interpretation of gravitational collapse of the upper carapace and liquid expulsion at the bottom of the vertical capsule. The dashed curve represent the theoretical position of the liquidus in the absence of gravity collapse (a).

246x277mm (300 x 300 DPI)

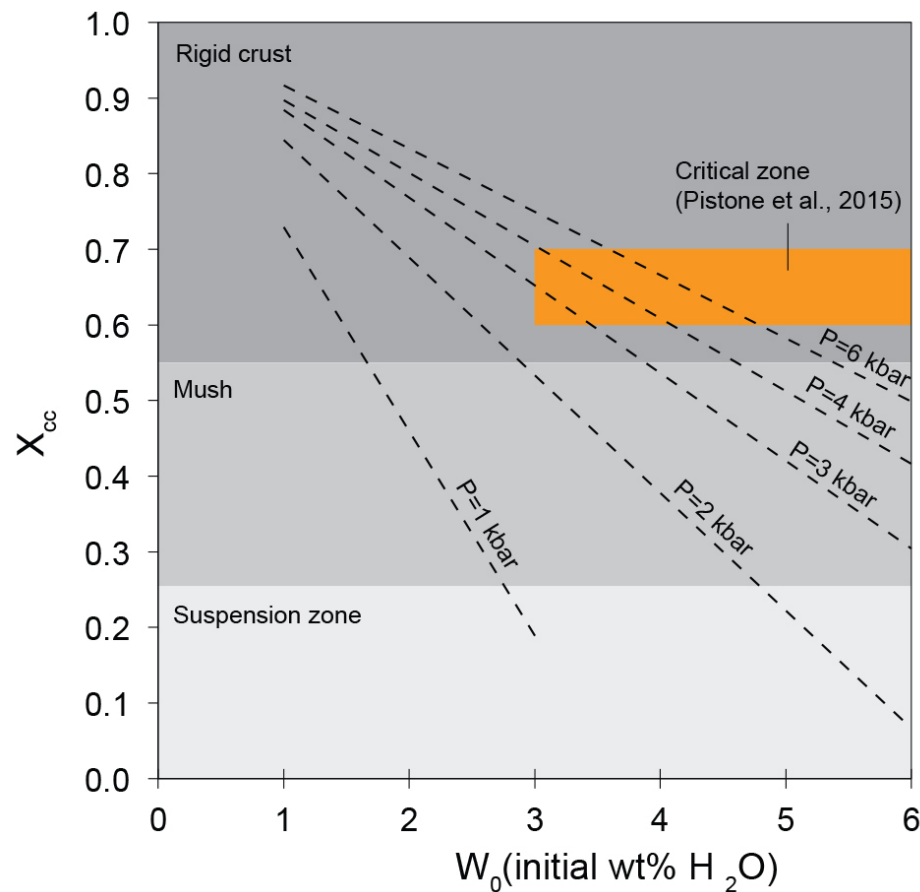


Figure 3 Plot of critical crystallinity ( $X_{cc} = 1 - X_{sl}$ ) versus initial water content ( $W_0$ ) of a granitic liquid using Eq. (3) at variable pressures from 3 to 6 kbar. The critical crystallinity represents the crystal fraction at which the remaining liquid reaches water saturation and boiling. The three zones of the solidification front (suspension, mush and rigid crust) are marked using the boundaries given by [Marsh, 2002]. The most favorable conditions to promote crystal-liquid separation by fluid-assisted filter pressing according to experiments [Pistone et al., 2015] are also depicted.

172x155mm (150 x 150 DPI)

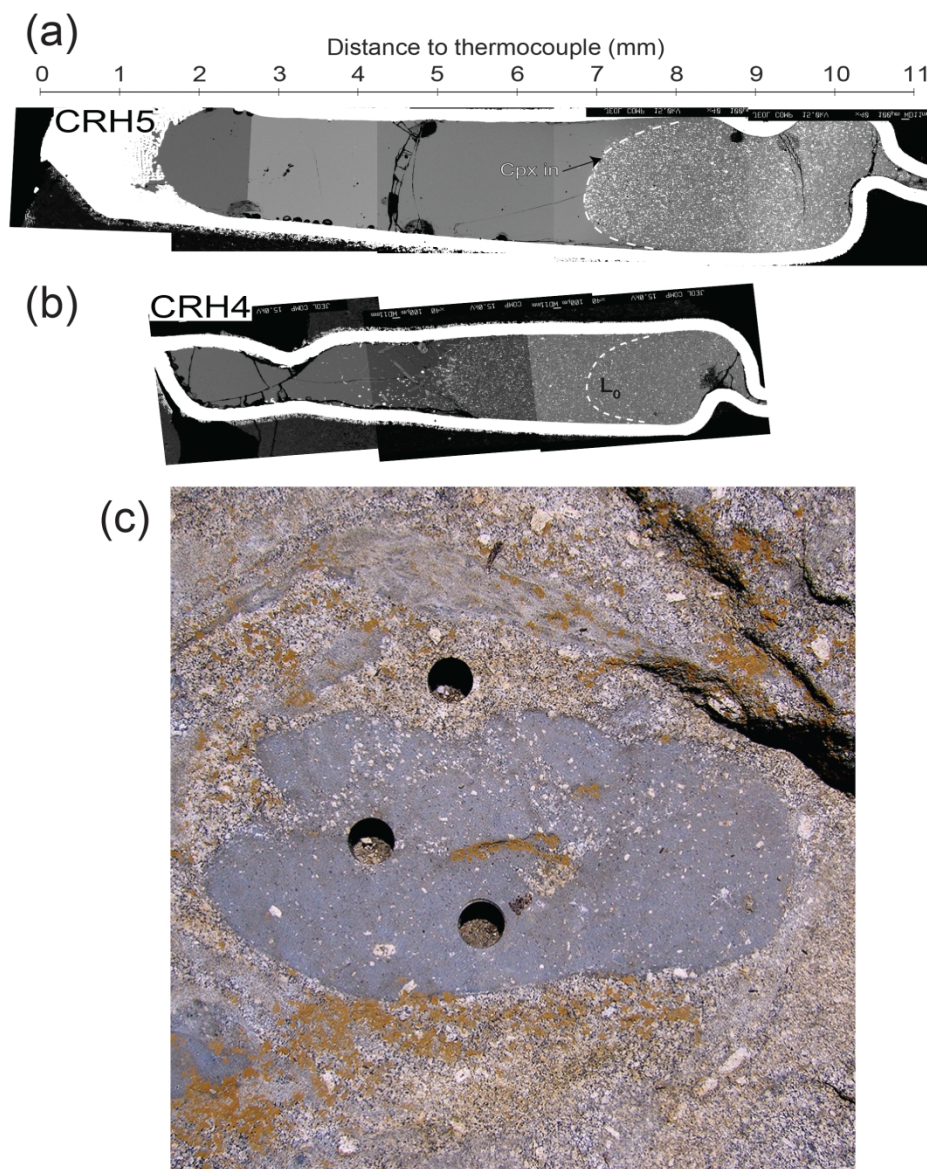


Figure 4 a) Mosaics of backscattered electron images from two polished sections of runs CRH5 and CRH4 with the AGV andesite (Rodríguez and Castro, 2017) using the thermal gradient imposed by the experimental assemblage. Initial conditions are  $P=5$  kbar and  $T=1200$  °C at the distance 0-3 mm from the thermocouple. CRH5 was quenched after 315 hours at the initial conditions. The liquidus (Cpx) is set approximately at 980 °C. In CRH4  $T$  was dropped at the rate of 0.6 °C/hour during 308 hours (until 1016 °C at the thermocouple). The composition of glass (quenched liquid) at 2 mm of the thermocouple is more fractionated (richer in  $K_2O$  and  $SiO_2$  and poorer in  $CaO$  and  $MgO$ ) in CRH4 compared with CRH5, in which no gradient was applied.

b) Field photographs of partially dismembered autoliths from the Gredos batholith in Puente del Congosto (Central Spain).

192x226mm (300 x 300 DPI)

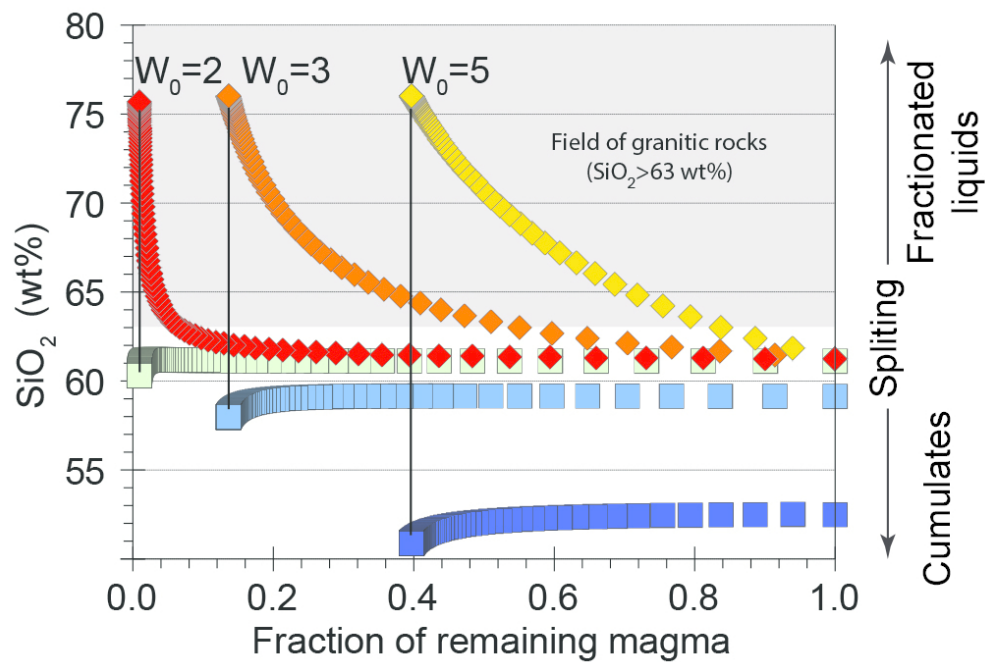


Figure 5 Variation in the silica content of liquids (in a unit magma chamber) that are modified by influx of residual water-saturated liquid coming from the solidification front (thermal boundary layer) at the sidewalls. Liquid curves are calculated with Langmuir's equation [Langmuir 1989] for in-situ crystallization (See text for further explanations).

172x117mm (150 x 150 DPI)

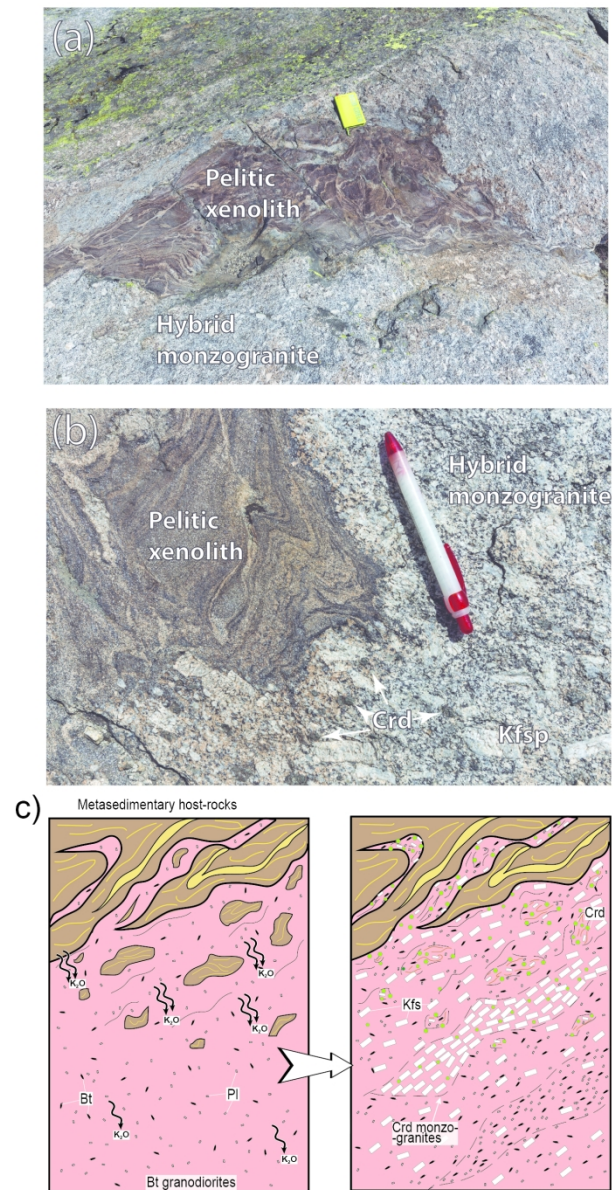


Figure 6 Field relations of partially digested pelitic xenoliths enclosed in calc-alkaline monzogranites and granodiorites of the Gredos batholith (Central Spain). (a) Large xenolith of partially molten (migmatite) metasediments showing irregular contacts. (b) Detail of another xenolith showing the concentration of large Crd crystals (dark dots) around the contacts. (c) Sketch in two stages showing the possible digestion of xenoliths by peritectic melting reaction and the formation of Crd (green dots) and Kfs that appear finally disseminated in the contaminated zones. The arrival of K to the pristine granodiorite shifts the composition of the final contaminated magma to monzogranite.

128x235mm (300 x 300 DPI)



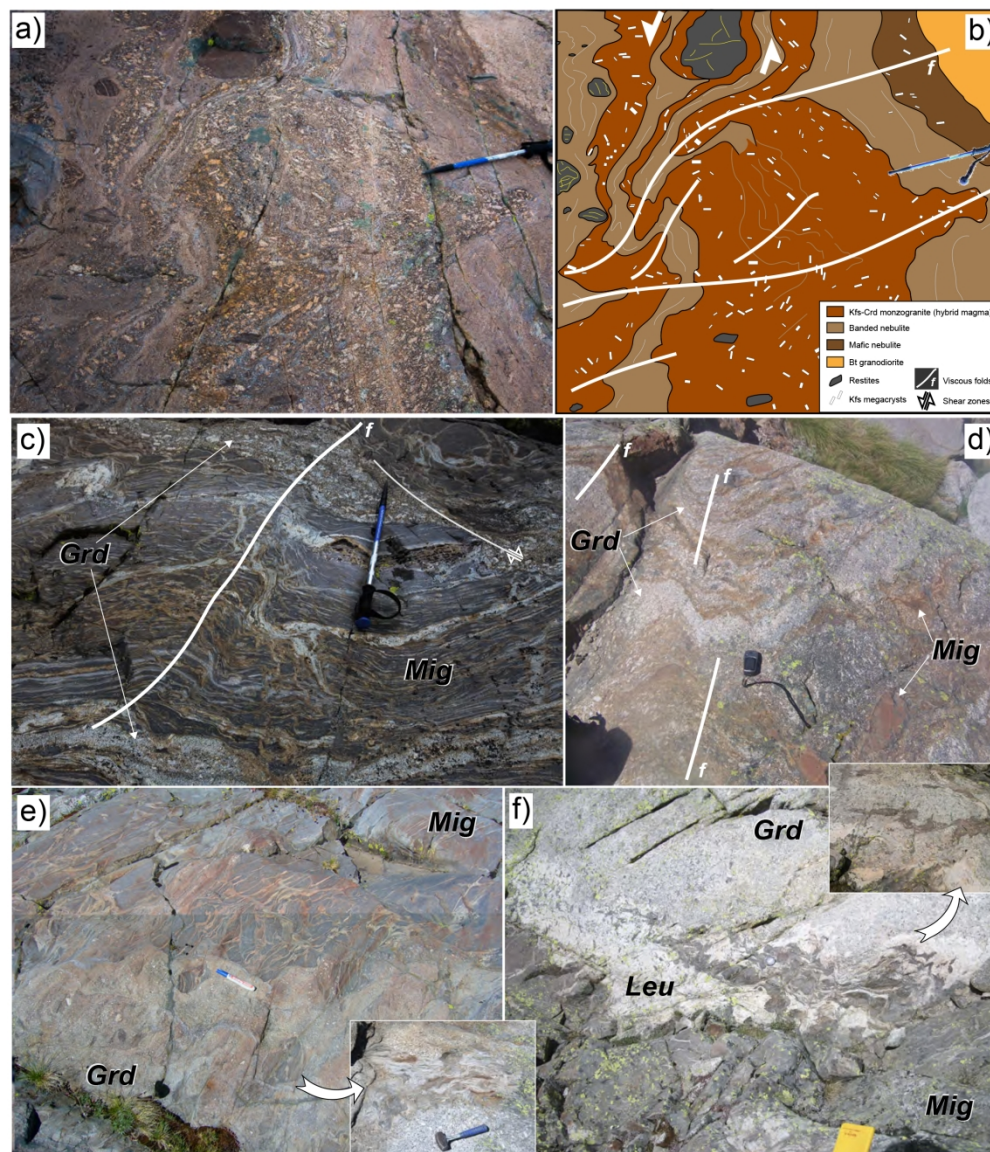


Figure 7 Field examples (Gredos massif, Spain) of heterogeneous structures resulting from the interaction processes between a partially crystallized magma and a partially molten metasedimentary host rock. A) to D) are cases illustrating mechanisms of viscous folding and shearing. A and B) Field photograph and interpretative sketch of complexly interleaved and folded sheets of migmatites, intrusive Bt granodiorite, and hybrid Kfs-Crd monzogranite. C and D) Coeval folding (f is the axial trace) of metatexite and granodiorite intrusive sheets. Shear zones are also seen affecting the system. E) Sharp contact between the intrusive granodiorite (lower half of the photograph) and the migmatitic host rock (upper half) showing xenolithic fragments dragged by the intruding magma from its host rock. Inset depicts the final result of this process, with disaggregation of the metasedimentary xenoliths (melt + restitic and peritectic phases) within the granodiorite magma. F) Tearing apart of migmatite mesosome from an intruding wedge of granodiorite magma promoted by the formation of a three-dimensional network of interconnected leucosome veins. Inset shows the individualization and disruption of sheets of mesosome and melanosome into the intruding magma. Grd: intrusive granodiorite. Mig: Migmatite. Leu: Leucosome.

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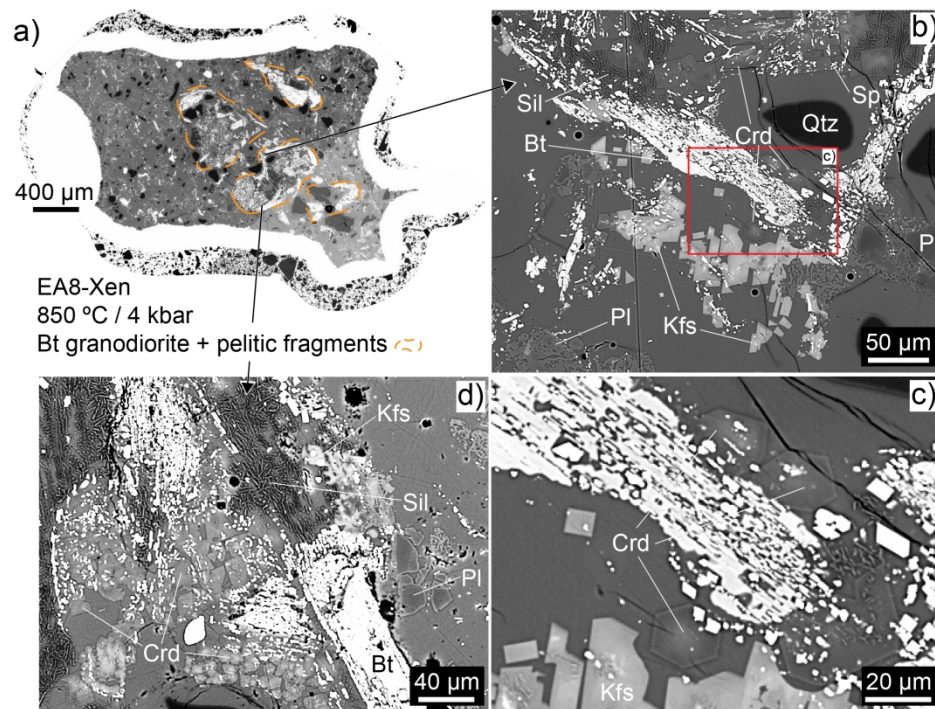


Figure 8 Electronic compositional images (backscattered electrons) of experimental run products simulating reaction between a pelitic xenoliths and a granodiorite liquid, after Díaz-Alvarado et al. [2011]. (a) Section of the whole capsule showing the remnants of partially molten and dismembered xenoliths. (b), (c) and (d) Details of the same run product showing the formation of euhedral Crd and Kfs as peritectic phases.

209x151mm (300 x 300 DPI)