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

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Abstract

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

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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 (SiO₂ > 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 SiO₂, 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 form 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 process differ significantly 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 magmas are characterized by high silica contents. The implication is that viscosity is 80 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,

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 kilometric 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 characteristic of cotectic variations and, hence, of liquid fractionation from one or 139 140 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,

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

186 3.1 Gravitational Collapse and Compaction

187 In spite of the scarce theoretical support to an efficient process of crystal-liquid separation that allows large volumes of silicic magmas to be available in the continental 188 189 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 (rhyolitic) magmas [Lipman, 1988]. The two main approaches to understand the 192 193 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

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

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

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 SiO₂ and K₂O, 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

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
- 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 =crystals/crystals+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 $X_{\rm c}$ 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	$X_{\rm sl} = W_0 / [W_{\rm s}]_{\rm (P)} \tag{1}$
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	[<i>Castro</i> , 2013]:
338	
339	$W_s = -0.27 P^2 + 3.54P + 0.42;$ (for P < 6 kbar) (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)} $ (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}

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 magmas giving rise to zoned intrusions and fractionated cupolas atop of plutons. This 360 361 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 contents and very low pressure (Fig. 3). For a magma to reach saturation at $X_c < 0.5$ at 375 376 the pressure of 3 kbar, the initial water content must be higher than 6 wt% H_2O . 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 ($W_0 < 3 \text{ wt\%}$) are
- 383 required to get X_{cc} within the critical zone of rigid crust of a solidification front. In sum,
- 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

The sidewalls of magma ascent conduits represent large transient interfaces along which
magma looses heat to the surrounding host with the consequent generation of thermal

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
- important role in magma differentiation during ascent; particularly in the case of water-
- 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
- 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
- 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

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

7 $C_M = C_0 F^{(f_A(E-1)/(f_A-1))}$ (4)

428

429 Where C_0 is the initial magma composition (in this case the standard AGV and esite), F is the fraction of melt (liquid / liquid + crystals), f_A is the fraction of liquid returned to 430 431 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 (C_{SL}) to the composition of magma in the same element of reference (C_M) . The 433 434 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 (Δ) 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 expulsed from the crystal-rich mush. That solid fraction (1-F), or 439 cumulate, at every discrete increment (Δ) 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% H₂O 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	$(SiO_2 > 63 \text{ wt\%})$ 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 $SiO_2 > 63$ 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 means of isotopic ratios, source contamination can be difficult to distinguish from 464 465 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 diagrams, the intricacies of the process can be varied. The study of field relations in 472 473 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.

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

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

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±1.5 Ma and 305±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

feature of these "mixed" granites. The presence of Crd in this kind of granites has been interpreted as result of a peritectic reaction in the local domain of the xenoliths after

- 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

abundance of mafic microgranular enclaves (autoliths) and xenoliths is also greater in

the margin zones compared to the inner parts. The inferences from field relations were

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

chemical interactions between the intruding magmas and its host rocks during ascent

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

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

starting materials representing the whole composition of Crd- or Grt-bearing granitic

for rocks are used [*Díaz–Alvarado*, 2017; *García–Moreno et al.*, 2017]. The addition of

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

around crystals or along layered contacts [*Acosta–Vigil et al.*, 2002; *Díaz–Alvarado et*

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al., 2011; *Garcia–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 goberned by the mineral reaction rates and diffusion
- 612 through the melt, besides other conditions as H₂O content and convection [*Acosta–Vigil*
- *et al.*, 2002, 2006]. Nonetheless, the above mentioned experiments show the resilience
- 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 H₂O 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
- outer rims, and the less contaminated and more felsic types are at the core.
- 633 4.2.3 Bulk-Assimilation Experiments
- 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

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

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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) Compositional profiles of glasses (quenched liquid) along the vertical capsule. (e) 987 988 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).

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

993 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

- 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 °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 °C. In CRH4 T was dropped at the rate of 0.6 °C/hour during 308 1004 hours (until 1016 °C at the thermocouple). The composition of glass (quenched liquid) 1005 at 2 mm of the thermocouple is more fractionated (richer in K2O and SiO2 and poorer 1006 in CaO and 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).

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

- 1012 Langmuir's equation [*Langmuir*, 1989] for in-situ crystallization (See text for further
- 1013 explanations).

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)

- 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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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.
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.



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 that of assimilation (Modified from Castro, 2013]).

142x226mm (150 x 150 DPI)

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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 (Xcc = 1–Xsl) versus initial water content (W0) 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 dropen 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 K2O and SiO2 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.

209x243mm (300 x 300 DPI)



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.

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