

Preface

The magmatic to hydrothermal transition and its bearing on ore-forming systems

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Abstract

The exsolution of volatile phases from silicate magmas controls physical and chemical magma properties and influences large-scale geologic phenomena and processes having major societal and economic implications including the release of climate-altering gases to the atmosphere, the explosivity of volcanic eruptions, hydrothermal alteration, and the generation of magmatic–hydrothermal mineralization. These volatile phases exsolve from a wide variety of magmas and cover a very broad spectrum of compositions.

The transition from the orthomagmatic to the hydrothermal stages has important bearing on these fundamentally important geologic phenomena, and this report summarizes the published results of a dozen scientific investigations on the magmatic–hydrothermal transition as applied to volcanic eruption and magmatic–hydrothermal mineralization. These studies involve a variety of analytical and experimental methodologies, and many focus on fluid and melt inclusions from mineralized magmatic systems. A primary goal of each study is to better understand the role of magmatic volatiles and the importance of the magmatic–hydrothermal transition on these geologic processes.

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1. Introduction

The exsolution of magmatic volatile phases from silicate melt causes fundamental changes in the physical and chemical properties of magmas in subsurface environments. These changes result in large-scale geologic phenomena that have major societal and economic implications. Perhaps most significantly,

the globally important consequences of volatile phase exsolution include the release of climate-altering gases to the atmosphere, control on the explosivity of volcanic eruptions, metasomatism/alteration of rock by hydrothermal fluid, and the generation of economically important, magmatic–hydrothermal ore deposits.

The transition interval from the orthomagmatic stage, which is dominated by condensed crystal–melt interactions, to the hydrothermal stage, which is dominated by condensed crystal–melt–magmatic volatile phase interactions (Burnham and Ohmoto, 1980), involves highly mobile and reactive volatile constitu-

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ents that leave abundant signs of their activity, but minimal information otherwise. The interval is poorly understood largely because the textural and chemical evidence in rocks of processes occurring at this stage has been either altered or completely removed. This is true despite extensive investigations that have included: field studies of volcanoes and their associated gases, hot springs, geothermal fields, and acid crater lakes; field investigations of hydrothermally altered and variably mineralized plutonic and volcanic systems; petrologic and geochemical studies of materials from these systems; analyses of thousands of fluid and melt inclusions from such systems; and hundreds of experimental studies attempting to replicate magmatic environments and processes.

Significant progress in understanding the magmatic to hydrothermal transition has been achieved in recent years through the development of microanalytical techniques, such as secondary ion mass spectrometry (SIMS), laser-ablation ICPMS or AES, proton-induced X-ray emission probe (PIXE), and RAMAN microprobe spectroscopy (Hervig et al., 1989; Sobolev, 1996; Webster et al., 1996; Audétat et al., 1998; Thomas, 2000; Halter et al., 2002b, 2004; Kamenetsky et al., 2002; Heinrich et al., 2003 and Reyf, 2004 in this volume). These instruments have provided important new information on the major, trace, and volatile constituents of natural silicate melts before and after saturation of a volatile phase as well as constraints on the composition of magmatic fluids after exsolution. Use of these techniques on appropriate samples makes it possible to bracket the conditions of volatile saturation more closely and, thus, to better constrain the timing of volatile saturation, the conditions responsible for volatile phase separation, and the behavior of individual components during such events.

This special volume represents a selection of papers dealing with the evolution of magmas prior to and during volatile saturation, the composition and role of exsolved volatiles, as well as the potential impact of these processes on volcanism and on the formation of magmatic–hydrothermal ore deposits. The objective is to bring together results from various research domains including igneous petrology/geochemistry, fluid/melt inclusion studies, experimental petrology, ore deposits research, and hydrothermal geochemistry to provide useful, new constraints on the conditions and effects of volatile exsolution in shallowly emplaced magmas.

2. Conditions for volatile saturation and effect of composition

Phase separation occurs when the solubility of the multicomponent volatile phase or phases in the silicate melt is exceeded. Thus, it reflects the fact that the bulk system enters an immiscibility domain where the coexistence of two or more phases (typically a silicate melt and an aqueous fluid) is energetically favored over a single mixed liquid. This miscibility gap can be investigated experimentally (see the contributions by Veksler, 2004 and Schatz et al., 2004) or by observation of natural systems, such as those described by Kamenetsky et al. (2004), Peretyazhko et al. (2004), and Reyf (2004). The size of this miscibility gap and thus the solubility of volatiles in silicate melts depend on pressure, temperature, and bulk composition of the system. The effects of these parameters have been investigated experimentally and the contributions by Veksler (2004) and Webster (2004) discuss major results from such studies and the implications for the understanding of magmatic systems.

The conditions at which systems enter the domain of fluid–melt immiscibility strongly affect the composition of the coexisting fluid and melt phases. Thus, silicate magmas saturate in and exsolve a broad compositional spectrum of volatile phases at highly variable conditions. Even in a single system, compositional evolution along the solvus during volatile exsolution and crystallization of magma can significantly affect the composition of the coexisting melt and volatile phases (Thomas et al., 2000; Audétat and Pettke, 2003).

The exsolved phases include hydrothermal aqueous, aqueous-halide, aqueous-carbonic, and aqueous-sulfate vapors or liquids, and sulfide, carbonate, phosphate, and/or hydrosaline (Cl- and/or F-rich) liquids (Roedder, 1992). Aqueous hydrothermal fluids have the greatest influence on these processes simply because water is the dominant volatile constituent of silicate magmas. Thus, previous studies addressing the magmatic–hydrothermal transition generally focused on the condition of water saturation of silicate magma (Burnham and Ohmoto, 1980). However, related experimental research has shown that even small quantities of CO₂ (Holloway, 1976; Holloway and Blank, 1994) or Cl (Webster, 1997; Webster et al., 1999) lead to volatile phase saturation of felsic and related magmas containing comparatively low amounts of H₂O.

Webster (2004) provides a detailed discussion of the experimental data and compares them to volatile abundances of natural systems to determine the effect of Cl on water solubility in felsic to basaltic magmas.

Changes in the composition of silicate melt as a function of the exsolved volatiles are discussed in several contributions to this volume. The effect of boron has been investigated by Peretyazhko et al. (2004) and Schatz et al. (2004) through melt inclusions and thermodynamic considerations. The effect of Be and F on the behavior of other ore metals is discussed in Reyf (2004). To gain insight into the effect of volatile exsolution on the melt composition in a Li- and F-enriched granitic system, Badanina et al. (2004) investigated melt inclusions and identified the timing of volatile exsolution through changes in melt composition. The effect of an exsolved F-rich fluid on the magma is described by Chang and Meinert (2004). Holloway (2004) discussed the effect of the water content of mafic melts on the redox potential measured in associated rocks.

3. Volatile exsolution and physical processes

The “pneumatolytic” or magmatic–hydrothermal transition begins at the temperature of formation of a separate volatile phase and ends at the solidus (London, 1986). Although hydrothermal processes are typically attributed to late-stage, near-solidus conditions, recent observations on volcanic systems indicate that magmatic volatile phases may exsolve well before the final stages of crystallization. Fluids may exsolve at temperature and pressure conditions at which the degree of crystallinity of the magma is still quite low (Gerlach et al., 1994a,b; Webster and Rebbert, 2001; Wallace et al., 1995 and references cited therein; Webster et al., 2003; Halter et al., 2004), so volatile phase processes may begin much earlier than generally held. Specifically, work on volcanic plumbing systems (Lowenstern, 1994; Gerlach et al., 1994a,b; Wallace et al., 1995; Wallace, 2001; Webster et al., 2003) indicates that early, and perhaps initial, volatile phase exsolution occurs at pressures which are much greater than those that have been attributed, in the past, to hydrothermally altered and mineralized felsic magmas.

At initial volatile saturation, small quantities of multicomponent fluid are generated. Most physical

and chemical changes in the melt occur, however, only as the latter has exsolved a significant amount of volatiles, i.e., when the bulk system has moved well into the immiscibility field between silicate melt and fluid. Significant volatile exsolution is associated with a large volume increase of bulk magma, as the partial molar volume of water in silicate melt is much smaller than in an aqueous fluid (Burnham and Davis, 1974; Burnham, 1997). The lower density of an aqueous fluid relative to melt also reduces the bulk density of the magma, and this provides strong buoyancy forces that drive the ascent of bubble-laden magma plumes to the apical regions of magma chambers and/or cause the ascent of hydrothermal fluid through magma (Candela, 1991).

The mechanisms by which a dispersed fluid phase is focused or concentrated in magma are extremely important because the influence of magmatic–hydrothermal fluids on mineralization and alteration processes varies with the quantity of fluid. As bodies of magma crystallize from their margins inwards and volatile phase saturation occurs, the resultant hydrothermal fluid is transferred through magma, and this is abundantly evident from investigations showing volatile-enriched magma concentrated in the caps of magma chambers (Hildreth, 1981; Candela, 1991; Wallace et al., 1995; many others). It follows that individual bubbles of hydrothermal fluid as well as bubble-laden magma plumes must ascend to the apical regions of magma chambers (Candela, 1991). Mirolitic cavities, such as those described by Peretyazhko et al. (2004) and Kamenetsky et al. (2004), are typical examples of such volatile accumulations in the apical parts of magma reservoirs including those containing pegmatite domains.

Magmatic volatiles also have important fluxing effects, thus lowering the solidus temperature of silicate melt. Fluorine and boron are particularly important in this respect, as evidenced by the contributions by Veksler (2004), Chang and Meinert (2004), and Peretyazhko et al. (2004). Conversely, the loss of large amounts of these volatiles through fluid exsolution in open systems may lead to an increasing degree of melt polymerization. Thus, volatile exsolution and escape from magmatic systems can cause drastic increases in the viscosity and solidus temperatures of residual melt. Consequently, the combination of the strong volume increase of magma with increasing

melt viscosity, resulting from fluid exsolution, is responsible for the highly explosive nature of volcanism that is associated with water-rich, intermediate to acid magmatic systems. Similar processes also generate some of the fine fracture networks required to form disseminated mineralization in porphyry-type ore deposits.

4. Magmatic–hydrothermal fluids

As a volatile phase separates from the silicate magma, many incompatible elements will strongly fractionate into the exsolving phase. Thus, magmatic fluids carry large amounts of sulfur, chlorine, fluorine, alkalis, and metals such as Cu, Pb, Zn, Sn, W, Mo, Au, Li, As, and others (Candela and Piccoli, 1995; Bai and Koster van Groos, 1999). The exsolved volatile phase can vary from a low-salinity, low-density phase containing up to a few tens of percent NaCl_{eq} to a high-salinity, high-density phase containing up to 80 wt.% NaCl_{eq} . The main reason for this variability is that exsolution can occur either in the one-phase or the two-phase field of the H_2O – NaCl system, generating an intermediate salinity fluid or a low-salinity vapor plus a high-salinity hydrosaline liquid/melt (Shinohara, 1994; Webster, 1997). Although absolute metal concentrations are generally higher in high-salinity liquids, new experimental data (Migdisov et al., 1998, 1999; Archibald et al., 2001, 2002; Williams et al., 2002) and application of new analytical techniques (Heinrich et al., 1999) have shown that the vapor phase can transport significant amounts of metals.

There is some confusion in the literature about the nomenclature of exsolved volatile phases: the term “vapor” is agreed upon to designate a low-density, low-salinity aqueous phase, but the high-density, high-salinity phase is typically referred to as either “hydrosaline liquid/melt” or “brine.” Liquids with salinities of 80 wt.% NaCl_{eq} may have little in common with aqueous solutions (i.e., brines), so use of the term hydrosaline liquid, although less common, may be more appropriate. The compositional limit between the two expressions is somewhat arbitrary, as they are not separated by a distinct phase transition, but we suggest that saline aqueous solutions with more than 50 wt.% NaCl_{eq} be referred to as hydrosaline liquids. This distinction between hydrosaline

liquids and brines emphasizes the point that liquids with very high salinities have thermodynamic properties that cannot be approximated by extrapolating properties of aqueous solutions. In particular, activity models developed for aqueous solutions may not apply to hydrosaline liquids. This is indeed true for most solutions with salinities above 10–20 wt.% NaCl_{eq} , but certainly for all above 50 wt.% NaCl_{eq} .

The compositions of the exsolved volatile phase(s), and thus the conditions at which this exsolution occurs, have a major influence on the behavior of major and trace elements and on their distribution among the various phases (Halter et al., 2002a). They will affect the partitioning of metals between silicate melt and volatile phase(s) as well as the metal ratios between the vapor phase and brine or hydrosaline liquid (Bodnar et al., 1985; Heinrich et al., 1999; Kamenetsky et al., 2004; Rusk et al., 2004). As a result, magmatic–hydrothermal fluids carry highly variable concentrations of metals and generate a broad range in styles of alteration and ore deposits. Porphyry systems and associated epithermal deposits are arguably the best-studied examples. The contributions by Beuchat et al. (2004), Chang and Meinert (2004), Reyf (2004), and Rusk et al. (2004) investigate additional examples of such fluids and their role in the formation of magmatic hydrothermal ore deposits.

5. About this volume

Most of the following papers were presented at the Goldschmidt Conference in 2002 in Davos, in a session entitled “Magma Chambers and Ore-Forming Processes at the Magmatic Hydrothermal Interface.” They focus on various aspects of the magmatic to hydrothermal transition and provide important constraints on processes that govern volatile exsolution and compositions of exsolved volatile phase. Abstracts of all the contributions presented at this Goldschmidt session were published in a special issue of *Geochimica et Cosmochimica Acta* (vol. 66, 15A).

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