Revision 1

Granites and rhyolites: Messages from Hong Kong, courtesy of zircon

A recent issue of ***Elements***, edited by Craig Lundstrom and Allen Glazner (2016), is titled "Enigmatic Relationship Between Silicic Volcanic and Plutonic Rocks." This title, and the articles in the issue, reflect the rather remarkable fact that the origins of silicic magmas and the relationship between their erupted and intruded products – rhyolite and granite *sensu lato* – remain a topic of great interest, uncertainty, and heated debate. This, despite the fact that these rocks comprise a large part of Earth's crust, include products of arguably the largest and most impactful eruptions on Earth, and have been puzzled over by investigators for centuries, since before the dawn of Geology as a science. A paper in this issue of *American Mineralogist* by Tang et al. provides new perspectives and insights on these problems that arise from a detailed study of a particularly opportune natural example.

A Brief History. To gain a perspective on views and debates about granite and rhyolite today, it is worth a glance back to where they stood 70 years ago. Both rock types were well known, as were the facts that their chemical and mineralogical compositions were generally similar and that rhyolite was indeed formed from magma. Granite, however – despite its enormous abundance (at least as defined *s.l.*) and importance in the exposed crust – was at the center of bitter dispute that at the time overshadowed disagreements about continental drift (plate tectonics was yet to be proposed). Hutton had suggested in the late 18th century that granite, or at least some granite, was the product of intruding and cooling molten magma, but in the mid-20th century that was far from universally accepted. A memorable day-long session of the 1947 GSA meeting in Ottawa was entitled "The Origin of Granites." The symposium was devoted to debate about whether granite was formed by crystallization of magma, by replacement of pre-existing rock with or without participation of watery or magmatic fluids ("granitization"), or by both of these processes. GSA Memoir 28 (Gilluly, 1948) records this fiery debate, including addresses and discussion by such luminaries as Read, Buddington, Grout, Bowen, and Shand. Interestingly, the word "rhyolite" is not mentioned once (assuming reliability of my recollection from grad school reading of the text, and a recent search). Ten years later, Tuttle and Bowen (1958) published what was essentially a follow-up that very much took rhyolites into consideration: GSA Memoir 74, "The Origin of Granite in Light of Experimental Studies." They noted that the compositions of silicate melts in equilibrium with quartz and feldspar, granites (*sensu stricto* in this case), and rhyolites coincided. This coincidence – of melts produced in the lab, melt-rich rhyolites, and the controversial granites – and the power of the application of phase equilibria effectively ended the debate about whether granites were magmatic. Left open was the question of whether felsic magmas – granites and rhyolites – represented products of partial melting of quartz- and feldspar-bearing rocks (crustal anatexis), fractional crystallization of more mafic magma (potentially mantle-derived), or both, since phase equilibria simply required a melt that was saturated, or nearly saturated, in both feldspar and quartz. And it also left open the question of whether rhyolites and granites have common origins.

Questions Linger and Arise. Sixty years after publication of Tuttle and Bowen's pivotal study, questions linger, and in fact new questions continue to arise, about silicic magmatism, and the relationship – or lack of relationship – between granites and rhyolites remains central. Currently active debate is not as acrimonious as it was 70 years ago, but it sometimes comes close. It includes, but is not limited to, the following questions:

(1) Are silicic magmas mostly generated by partial melting of preexisting crust or fractional crystallization of mafic magma (e.g. Martin and Sigmarsson 2007, Sawyer et al. 2011, Brown 2013, Annen et al 2015, Lipman and Bachmann 2015)? Or by a combination of the two processes (cf. AFC [assimiliation-fractional crystallization; DePaolo 1981], MASH [melting-assimilation-storage-homogenization; Hildreth and Moorbath 1988])? And do the processes by which erupted silicic magmas are generated differ systematically from those by which compositionally similar intrusive magmas are formed?

(2) How commonly – and how – are intrusive silicic magmas physically linked to volcanic counterparts – the "volcanic-plutonic connection?" (e.g. Bachmann et al. 2007; Mills and Coleman 2013; Bachmann and Huber 2016; Lundstrom and Glazner 2016). Do large batholiths contain the residue of super-scale eruptions? Or are batholith construction and supereruptions for the most part mutually exclusive?

(3) What is the nature of the silicic magma bodies that erupt, and those that form batholiths – and are they the same? How much of their volume is *eruptible* (sufficiently mobile to be capable of eruption: melt-rich magma and crystal-richer, more sluggish *mush*) and how much is locked up within melt-poor, uneruptible "*rigid sponge*" or fully solidified magma (Marsh 1981, Hildreth 2004)? Do they contain *cumulate* zones in

which crystals have been concentrated and from which melt was extracted, and are rocks that represent these crystal-rich and complementary melt-rich materials compositionally and texturally distinct within plutons, and in erupted products (e.g. Lipman and Bachmann 2015; Keller et al. 2015)?

(4) How does the distribution of the rheologically distinct zones within these subsurface bodies vary in four dimensions: what is their geometry and scale, and how do they vary through time? These questions have received particular attention recently because they are critical for understanding how batholiths – the dominant volume of Earth's continental crust – are constructed, how eruptions work, and the threats posed by potentially hazardous volcanoes (e.g. Cashman and Giordano 2014, Lundstrom and Glazner 2016). Do the reservoirs from which eruptions emerge and batholiths are constructed contain large, long-lived masses of eruptible magma, sustained by periodic magma recharge, or do discrete melt-rich pockets wax, wane, merge briefly into large bodies, and at times solidify in response to interplay between cooling and rejuvenation by recharge (e.g. Barboni et al. 2016; Klemetti 2016; Miller 2016; Rubin et al. 2017)?

(5) How does magma flux – from mantle into lower crust, from deeper levels into shallower crustal reservoirs (recharging) – vary in space and time, and thereby influence maintenance of melt-bearing magma bodies and eruptibility (e.g. Glazner et al. 2004; Caricchi et al. 2014; Karakas et al. 2017; Tang et al. 2017)?

Evolving Approaches: Time, Pace. Active and ancient silicic magma systems have been probed in recent years using a wide range of field, geochemical, and geophysical approaches. Arguably the most critical issues are connected with time: sequences, absolute ages, and durations of events (see Tang et al. this issue). Rapid advances in assessing ages and durations of magmatic events have led to a proliferation of studies that address questions like those presented above (see Lipman and Bachmann 2015; Wilson and Charlier 2016). $^{40}Ar/^{39}Ar$ analysis of several K-bearing minerals and U-Pb analysis of zircon by isotope dilution thermal ionization mass spectrometry (ID-TIMS) can now achieve previously unattainable precision for age determinations, on the order of ±10 ka. Secondary ion mass spectrometry (SIMS) provides both elemental compositions and ages on spots in zircon crystals ~20 microns in diameter and a micron deep (uncertainties generally >100 ka); laser ablation inductively-coupled plasma mass spectrometry (LA-ICPMS) yields more rapid *in situ* results, but with a larger analytical volume and somewhat lower precision (see also TIMS-TEA, Schoene et al 2010: high precision dating combined with elemental analysis). Even better absolute precision is possible using the U to Pb decay chain for young zircon (<~200 ka): the $^{238}U/^{230}Th$ disequilibria method can yield 1-10 ka uncertainties for ID-TIMS and SIMS analysis. However, several complications lead to less than straightforward interpretation of state-of-the-art results:

(1) Owing to high rates of diffusive loss of daughter Ar in all K-bearing minerals at high temperatures, $^{40}Ar/^{39}Ar$ ages are interpreted to be cooling ages (through sub-magmatic closure temperature, at which point Ar loss becomes minimal). Because volcanic rocks cool instantaneously (within current uncertainty), $^{40}Ar/^{39}Ar$ ages are considered to generally reflect true *eruption* ages. In contrast, zircon U-Pb and U-Th ages, whether by ID-TIMS, SIMS, or LA-ICPMS, should indicate *crystal growth* ages, because zircon is almost immune to diffusive Pb loss as long as its crystal structure remains intact – which it generally, but not invariably, does. Recent work using both ID-TIMS and SIMS

has shown unequivocally that zircon crystals may grow over readily measurable time periods and in volcanic rocks commonly yield ages older than eruption (see Tang et al. 2017 this issue). In other words: zircon ages and ^{40}Ar/^{39}Ar ages are not directly comparable or interchangeable. This discrepancy in meaning of ages can be very useful, because it is critically important to compare ages of crystal growth in magmas with time of eruptions. But: see (2) below. (Zircon also has the potential to reveal eruption age through the rather complicated process of (U-Th)/He dating, because in contrast to Pb or ^{230}Th, He produced by radioactive decay is lost via diffusion from zircon very rapidly [e.g. Schmitt et al. 2010])

(2) A further complication for the seemingly very fruitful comparison of ^{40}Ar/^{39}Ar eruption ages with zircon U-Pb and U-Th ages: difficulties in confident calibration of ^{40}Ar/^{39}Ar standards lead to uncertainties in absolute age determinations that currently exceed the outstanding analytical precision of the analyses.

(3) The obviously lengthy time intervals over which individual zircon crystals can grow in silicic magma systems further clouds optimal interpretation of U-Pb and U-Th dates and elemental analyses. With whole crystals or large fragments as are used in ID-TIMS work, all or much of the growth history is averaged into a single date; even relatively small SIMS analytical volumes can encompass tens of thousands of years, or more, of crystal growth.

(4) Zoning patterns that indicate repeated fluctuations during zircon growth (e.g. Claiborne et al. 2010) suggest, and diffusion chronometry strongly confirms, that processes of importance in magma systems act on timescales far shorter than are currently accessible by absolute age dating (e.g. Cooper et al. 2016). Measured

compositional profiles in crystals can be modeled using known diffusivities as a function of temperature to estimate time vs. temperature histories, and these estimates constrain how long the crystals resided in magma at high T. Such studies commonly imply very rapid fluctuations in temperature and brief immersion of crystals in melt, on the order of years to decades (e.g. Gualda and Sutton 2016; Rubin et al 2017). At present, diffusion chronometry is imprecise in a relative sense (uncertainty/estimated duration) and, without absolute dates with precision necessary for distinguishing very closely spaced events, correlation of events that it identifies is very difficult.

Granite-Rhyolite Relations: Insights from Hong Kong. In this issue, Tang et al. (2017) present an extensive zircon-based study of silicic volcanic and intrusive rocks in Hong Kong that span a 26 million year history. These rocks, the products of caldera-forming eruptions and underlying shallow plutons that in total comprise a substantial composite batholith, represent one of the world's best exposed examples of a large, physically connected intrusive-extrusive system. The authors elected to use the SIMS approach and thereby generated a very large data set that documents and compares growth histories – time and composition – of zircon in the intrusive and extrusive rocks. They demonstrate coherence between the zircon-recorded histories of erupted and intruded magmas. Zircon crystals in granites and rhyolites have the same wide ranges in composition that define similar populations and trends. Ages also reveal closely similar growth patterns throughout most of the 26 million year interval, though it appears that intrusion continued for about two million years after volcanism had largely or entirely ceased. The authors emphasize that "Composite plutons, like those which occur beneath Hong Kong…, grow by increments.

Their overall averaged growth rates are a misleading representation of complex, episodic and dynamic growth histories," and that "…volcanism for the Repulse Bay Volcanic Group and Kau Sai Chau Volcanic Group associated with the High Island caldera complex [the largest Hong Cong volcanic-plutonic complex] represent continuous (*within analytical uncertainties*) magmatic activity over ~5 Myr…."

Limitations on precision of SIMS analyses preclude evaluating in detail ages and related compositional variations on timescales now believed to apply to recharging and thermal fluctuations, and zircon data cannot directly measure timing of eruptions. And, more broadly, investigating magmatism in the uppermost crust doesn't directly relate to the question of whether granites *as a whole* share genetic kinship to rhyolites. But Tang et al.'s evidence seems unequivocally to demonstrate that, for their excellent Hong Kong example, magmas that formed batholith-scale intrusions and those in large silicic eruptions were closely related and experienced remarkably similar histories.

REFERENCES CITED

Annen, C., Blundy, J.D., Leuthold, J., and Sparks, R.S.J. (2015) Construction and evolution of igneous bodies: Towards an integrated perspective of crustal magmatism. Lithos, 230, 206–221.

Bachmann, O,, and Huber, C. (2016) Silicic magma reservoirs in the Earth's crust. American Mineralogist, 101, 2377–2404.

Bachmann, O., Miller, C.F., and de Silva, S.L. (2007) The volcanic-plutonic connection as a stage for understanding crustal magmatism:.Journal of Volcanology and Geothermal Research, 167, 1-23.

Barboni, M, Boehnke, P, Schmitt, A.K., Harrisona, T.M, Shane, P., Bouvier, A.S., and Baumgartner, L. (2016) Warm storage for arc magmas. PNAS, 113, 13959–13964.

Brown, M. (2013) Granite: From genesis to emplacement. GSA Bulletin, 125, 1079–1113.

Caricchi, L., Simpson, G., and Schaltegger, U. (2014) Lifetime and size of shallow magma bodies controlled by crustal-scale magmatism. Nature, 511, 456-461.

Cashman, K.V., and Giordano, G. (2014) Calderas and magma reservoirs. Journal of Volcanology and Geothermal Research, 288, 28-45.

Claiborne, L.L., Miller, C.F., Wooden, J.L., and Mazdab, F.K. (2010) Trace element composition of igneous zircon: A thermal and compositional record of accumulation and evolution of a large silicic batholith, Spirit Mountain, Nevada: Contributions to Mineralogy and Petrology, 160, 511-531.

Cooper, K.M., · Sims, K.W.W., · Eiler, J.M., · Banerjee, N., (2016) Timescales of storage and recycling of crystal mush at Krafla Volcano, Iceland. Contributions to Mineralogy and Petrology, 171:54

DePaolo, D.J. (1981) Trace element and isotopic effects of combined wallrock assimilation and fractional crystallization. Earth and Planetary Science Letters, 53, 189-202.

Gilluly, J (1948) Origin of granite, Conference at meeting of The Geological Society of America held in Ottawa, Canada, Dec. 30, 1947, Geological Society of America Memoir 28, 139 p.

Glazner, A.F., Bartley, J.M., Coleman, D.S., Gray, W., and Taylor, R.Z. (2004) Are plutons assembled over millions of years by amalgamation from small magma chambers? GSA Today, 14, 4–11.

Gualda, G.A.R., and Sutton, S.R. (2016) The Year Leading to a Supereruption. PLOS One, 11(7): e0159200.

Hildreth, W. (2004) Volcanological perspectives on Long Valley, Mammoth Mountain and Mono Craters: several contiguous but discreet systems. Journal of Volcanology and Geothermal Research 136, 169–198.

Hildreth, W., and Moorbath, S. (1988) Crustal contribution to arc magmatism in the Andes of Central Chile. Contributions to Mineralogy and Petrology, 98, 455–489.

Karakas, O., Degruyter, W., Bachmann, O. and Dufek, J. (2017) Lifetime and size of shallow magma bodies controlled by crustal-scale magmatism. Nature Geoscience, 10, 446-450.

Keller, C.B., Schoene, B., Barboni, M., Samperton, and Husson, J.M. (2015) Volcanic–plutonic parity and the differentiation of the continental crust. Nature, 523, 310-307.

Klemetti, E.W. (2016) Melts, mush, and more: Evidence for the state of intermediate-to-silicic arc magmatic systems. American Mineralogist, 101, 2365–2366.

Lipman, P.W., and Bachmann, O. (2015) Ignimbrites to batholiths: Integrating perspectives from geological, geophysical, and geochronological data. Geosphere, 11, 705–743.

Lundstrom, C.C., and Glazner, A.F. (2016) Silicic Magmatism and the volcanic–plutonic connection. Elements, 12, 91-96.

Marsh, B.D. (1981) On the crystallinity, probability of occurrence, and rheology of lava and magma. Contributions to Mineralogy and Petrology, 78, 85–98.

Martin E, Sigmarsson O (2007) Crustal thermal state and origin of silicic magma in Iceland: the case of Torfajokull, Ljosufjoll and Snaefellsjokull volcanoes. Contributions to Mineralogy and Petrology, 153, 593–605.

Miller, C.F., 2016, Commentary: Eruptible magma: PNAS, v. 113, p. 13941-13943.

Mills R.D. and Coleman D.S. (2013) Temporal and chemical connections between plutons and ignimbrites from the Mount Princeton magmatic center. Contributions to Mineralogy and Petrology, 165, 961-980.

Rubin, A.E., Cooper, K.M., Till, C.B., Kent, A.J.R., Costa, F., Bose, M., Gravley, D., Deering, C., Cole, J. (2017) Rapid cooling and cold storage in a silicic magma reservoir recorded in individual crystals. Science, 356, 1154–1156

Sawyer, E.W., Cesare, B., and Brown, M. (2011) When the Continental Crust Melts. Elements, 7, 229–234.

Schoene a,b,⇑, C. Latkoczy c, U. Schaltegger a, D. Günther (2010) A new method integrating high-precision U–Pb geochronology with zircon trace element analysis (U–Pb TIMS-TEA). Geochimica et Cosmochimica Acta, 74, 7144–7159.

Schmitt, A.K., Stockli, D.F., Lindsay, J.M., Robertson, R., Lovera, O.M., and Kislitsyn, R. (2010) Episodic growth and homogenization of plutonic roots in arc volcanoes from combined U–Th and (U–Th)/He zircon dating. arth and Planetary Science Letters, 295, 91–103.

Tang, D.L.K., Wilson, C.J.N., Sewell, R.J., Seward, D., Chan, L.S. Ireland, T.R., and Wooden, J.L. (2017) Tracking the evolution of late Mesozoic arc-related magmatic systems in Hong Kong using in-situ U-Pb dating and trace element analyses in zircons. American Mineralogist (this issue).

Tuttle, O. F., and Bowen, N. L. (1958) Origin of granite in the light of experimental studies in the system NaAlSi3O8-KAlSi3O8-SiO2-H2O. Geological Society of American Memoir 74, 1- 146.

Wilson, C.J.A., and Charlier, B.L.A. (2016) The life and times of silicic volcanic systems. Elements, 12, 103–108.