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2 Implications of equilibrium and disequilibrium among crystal 3 phases in the Bishop Tuff

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11 *ABSTRACT*

12 Eruption of the Bishop Tuff magma preserved equilibrium of exchange components and
13 element concentrations among magnetite, ilmenite, biotite, apatite, zircon and liquid.
14 Orthopyroxene and clinopyroxene were not in exchange equilibrium with the other MgFe-
15 bearing phases, but they appear to have been in equilibrium among themselves. Internally
16 consistent temperatures recorded by the FeTi-oxide, Ti-in-quartz, and $\Delta^{18}\text{O}$ quartz-magnetite
17 thermometers, coupled with evidence for magmatic corrosion of quartz and sanidine, indicate
18 that an initially low- T (≈ 700 °C), near- H_2O -saturated, high- SiO_2 rhyolite magma was heated up
19 to ≥ 800 °C and its crystal cargo partially melted by recharge of hotter melt from below. Oxygen
20 fugacity and compositions of biotite, ilmenite, magnetite, and silicate liquid initially adjusted by
21 internal rearrangement of components and conservation of oxygen. Partial melting of feldspars
22 liberated Sr and Ba back into the melt. Mixing during recharge eventually re-introduced

23 compatible elements (e.g., Mg, Ba, Sr) as well as foreign crystals of euhedral ortho- and
24 clinopyroxene, which evidently never totally re-equilibrated with the rhyolite liquid. Introduction
25 of CO₂ and accompanying reduction in the aH₂O during recharge raised crystallization
26 temperatures of quartz and sanidine in the rhyolite sufficient to allow marginal regrowth of these
27 phases with enhanced contents Ti, Ba and Sr.

28 *Keywords: Bishop Tuff, Supervolcanoes, silicic magma, petrogenetic processes, disequilibrium*

29 INTRODUCTION

30 Over the last decades, many studies focused on large silicic pyroclastic deposits
31 (ignimbrites) have reported the preservation of original thermal gradients of as much as 100-150
32 °C (~700 °C to >800 °C) from early to late-erupted material (over lengthscales of 10s to 100s of
33 meters; e.g., Lipman 1971; Hildreth 1979; Wolff et al. 1990; Mills et al. 1997; Matthews et al.
34 2012). These thermal gradients were revealed by microprobe analysis of the Fe-Ti oxides (co-
35 existing magnetite and ilmenite crystals), which systematically change composition from the
36 base to the top of stratigraphic sections. Assuming thermodynamic equilibrium, temperatures
37 were estimated from analyses of these opaque oxide minerals by use of then current
38 experimental/theoretical calibrations (e.g., Buddington and Lindsley 1964; Andersen et al. 1993;
39 Ghiorso and Evans 2008). For the Bishop Tuff, however, the assumption of quenched
40 Fe²⁺Ti(Fe³⁺)₂ exchange equilibrium between ilmenite and magnetite, and consequently the
41 reliability of extracted temperatures, was questioned (Lindsley et al. 1990, 1991; Ghiorso and
42 Sack 1991; Frost and Lindsley 1992) on the following grounds: (1) The granite-minimum
43 (“eutectic”) assemblage (Qtz, San, Plag, L, V) close to H₂O saturation could not be stable (as it
44 appears to be) over the measured 100 °C temperature range, and (2) the compositions of FeTi-
45 oxide minerals and pyroxenes failed to correlate in a manner appropriate for chemical
46 equilibrium. Barometry based on the oxides + pyroxene + quartz assemblage produce a range of

47 *P* values (from < 0 to 5 kb) that are hard to reconcile with independent constraints. Lindsley et al.
48 (1990, 1991), Ghiorso and Sack (1991) and Frost and Lindsley (1992) concluded that the
49 ilmenite had undergone late- or post-eruption re-equilibration and that oxide temperatures
50 substantially below the 824 ± 15 °C two-pyroxene temperature were unreliable. Therefore, a
51 sizeable thermal gradient may not have existed in the magma reservoir; it was instead an artifact
52 of additional, poorly understood and late- or post-magmatic processes.

53 This paper presents a reassessment of thermodynamic equilibrium among the various
54 Fe/Mg-bearing minerals of the Bishop Tuff (namely, ilmenite, magnetite, biotite, orthopyroxene
55 and clinopyroxene) and the silicic melt using published data (no new data added here). It is
56 customary for similar large silicic deposits to show evidence of local and partial equilibrium only
57 (Pichavant et al. 2007). Here, we use Roozeboom plots, as commonly employed in metamorphic
58 petrology, to assess thermodynamic exchange equilibrium among the coexisting phases. We
59 emphasize that all selected oxide pairs used have passed the Hirschmann and Bacon (Mn/Mg)
60 test (Hildreth and Wilson 2007). Hence, there would appear to be no *a-priori* reasons for
61 rejecting or questioning the reliability of the temperature estimates based on the oxide pairs (Fig.
62 1).

63 The Fe/Mg ratios (expressed as mole fractions X_{Mg}) of magnetite, ilmenite and biotite,
64 using analyses from Hildreth (1979) and Hildreth and Wilson (2007) plot in a manner strongly
65 suggestive that these phases crystallized close to mutual element-exchange equilibrium (Fig. 2).
66 Roozeboom plots ordinarily show curvature, and they are obliged to terminate at 0:0 and 1:1.
67 Although the data are few, the Fe/Mg composition of the melt also appears to be in equilibrium
68 with those minerals (Anderson et al. 2000). In any case, this conclusion follows from the role of
69 melt as the medium for element transfer among the minerals. In contrast, orthopyroxene and

70 clinopyroxene, which are found mostly in the late-erupted, hotter and more crystal-rich parts in
71 the Bishop Tuff deposits, fail to pass this test of exchange equilibrium with the FeTi oxides and
72 biotite. The two pyroxenes show constant values of X_{Mg} while the oxide minerals and biotite
73 exhibit clear positive correlations that extend in the case of the FeTi-oxides virtually to $X_{Mg} = 0$
74 (Fig. 2). Orthopyroxene and clinopyroxene do, however, appear to be in equilibrium with each
75 other. We note that the biotite compositions do not extend to low X_{Mg} values, and so the
76 extrapolation to the origin of the graph is unclear. A correction for Fe^{3+} might raise the X_{Mg}
77 values of biotite in the hotter, more oxidized samples (Fig. 1 and 2). The minor element
78 compositions of apatite and zircon vary consistently with temperature (Hildreth and Wilson
79 2007, p. 974), so we include them among the equilibrium minerals.

80 **IN DEFENSE OF FeTi-OXIDE THERMOBAROMETRY IN THE BISHOP TUFF**

81 In the following, we cite further reasons for considering that the pre-eruptive temperature
82 range measured by Fe-Ti oxides in the Bishop Tuff is robust and reliable:

- 83 1. Only chilled, unoxidized glassy samples with homogeneous oxide pairs were used for T - fO_2
84 determination (Hildreth and Wilson 2007, p. 973).
- 85 2. The lowest estimated temperatures (~ 700 °C) correspond to the expected temperature of the
86 haplogranite eutectic at low P (1-2 kbar) in H_2O -rich systems (Tuttle and Bowen 1958;
87 Whitney 1988; Holtz et al. 1992). It is not necessary to postulate post-depositional re-
88 adjustment of ilmenite to account for these low temperatures (Lindsley et al. 1991). The low
89 X_{Mg} values (low T) of the oxides show a remarkable clustering on the Roozeboom diagram
90 (Fig. 2). It is hard to believe that, with possibly one or two exceptions (Fig. 1), they
91 underwent the more random process of post-depositional re-equilibration. If ilmenite alone
92 (not magnetite) underwent a late-stage re-adjustment of the Ilm component, it is curious that

93 the Mg/Fe (Fig. 2) and Mn/Mg (Hildreth and Wilson 2007, Fig. 15) ratios retained their
94 equilibrium proportions.

95 3. Alternative thermometers applied to the Bishop Tuff (Ti-in-quartz, $\Delta^{18}\text{O}$ quartz-magnetite
96 thermometry, Hildreth and Wilson 2007) provide the same answer; a thermal gradient of \geq
97 100 °C (from ~ 700 °C to ≥ 800 °C). We note the concomitant variations in H_2O
98 concentrations in liquids from $\sim 5\text{-}6$ wt% in early-erupted samples to ~ 4 wt% in late-erupted
99 samples (Anderson et al. 2000, Fig. 2) suggesting that the magma was not fully H_2O -
100 saturated at ~ 2 kbar and high temperature. According to VolatileCalc (Newman and
101 Lowenstern 2002), a rhyolite magma with 6 wt % H_2O and 100 ppm CO_2 at 700 °C (typical
102 of the early-erupted material) yields an $X_{\text{H}_2\text{O}_{\text{vapor}}} \sim 0.95$, whereas one with 4 wt % H_2O and
103 600 ppm CO_2 (typical of the late-erupted material) at 820 °C yields an $X_{\text{H}_2\text{O}_{\text{vapor}}} \sim 0.59$.

104 4. Magnetite is modally more abundant than ilmenite in the Bishop Tuff by one to two orders of
105 magnitude (Hildreth 1979). This means that for closed-system temperature changes, the
106 magnetite composition should follow the Usp isopleth (Frost and Lindsley 1992). With
107 magnetite to ilmenite tielines in the $\text{TiO}_2\text{-FeO-Fe}_2\text{O}_3$ chemographic diagram essentially
108 rotating around magnetite, an increase in temperature of 100 °C leads predictably (Ghiorso
109 and Evans 2008, Fig. 14) to a 6-7 mol.% decrease in the Ilm component of ilmenite, and a
110 one-log unit increase in relative $f\text{O}_2$. This is exactly what is observed in the Bishop Tuff (Fig.
111 1). The concomitant and correlated variations in X_{Mg} of magnetite, ilmenite, biotite and melt
112 can in principle be achieved by closed-system re-equilibration (crystal-liquid fractionation
113 and conservation of oxygen) with increasing temperature. In this case, some partial melting
114 of biotite is involved, and all phases increase in X_{Mg} .

115 5. The fact that pyroxenes appear not to have been in exchange equilibrium with ilmenite,
116 magnetite and biotite (Fig. 1), was previously noticed by Lindsley et al. (1990, 1991) and
117 Ghiorso and Sack (1991). These authors attempted to apply the Opx-Qz-Mag-Ilm
118 equilibrium to an estimate of magma chamber pressure. However, they found an
119 unrealistically large range (from < 0 to 5 kb). According to the P - T phase diagrams (Whitney
120 1988; Johnson and Rutherford 1989), pyroxenes are not expected to be stable in the lower
121 temperature (< 770 °C at 2 kb for orthopyroxene; Evans and Ghiorso 1995, Figure 5a),
122 water-rich, high-SiO₂ rhyolite compositions that form most of the Bishop Tuff. In the general
123 case, their place should be taken by biotite, cummingtonite, or hornblende, depending on
124 magma composition. The pyroxenes cannot therefore be incorporated into any sort of “test”
125 of the accuracy of Fe-Ti oxide thermobarometry; they are clearly not in chemical equilibrium
126 with the oxides (Fig. 2). It is not unusual for the orthopyroxene also to be out of exchange
127 equilibrium with cummingtonite and biotite (e.g., Schmitz and Smith 2004). The euhedral
128 nature of the pyroxenes (Hildreth and Wilson 2007, p. 970) in our opinion, indicates a lack of
129 surface reaction with the present melt in the short time available (e.g., Pichavant et al. 2007;
130 Wark et al. 2007); their crystal faces developed earlier.

131 **EVIDENCE FOR MAGMATIC RECHARGE**

132 The elevated magmatic temperatures and the presence of pyroxenes in the late-erupted
133 parts of the Bishop Tuff are consistent with a late recharge event that brought deep, less evolved
134 magma and heat into contact with the high-SiO₂ rhyolite reservoir (e.g., Wark et al. 2007).
135 Recharge from below is found to be commonplace in large and small upper crustal reservoirs
136 (e.g., Sparks et al. 1977; Hervig and Dunbar 1992; Spera and Bohrsen 2004; Bohrsen and Spera
137 2007; Hildreth and Wilson 2007; Ruprecht and Bachmann 2010; Druitt et al. 2012; Lohmar et al.

138 2012), right up to a few decades prior to eruption (Wark et al. 2007; Gualda et al. 2012). A late
139 partial mixing between the resident high-SiO₂ rhyolite and a hotter, pyroxene-bearing recharge
140 (month to years before eruption) is consistent with evidence from fluid dynamics that the thermal
141 gradient could not be preserved for long periods of time (i.e., more than a few decades or
142 centuries for large bodies) in a crystal-poor magma chamber capable of undergoing convection
143 (Ruprecht et al. 2008; Huber et al. 2012; Lohmar et al. 2012). These thermal gradients must be
144 transient features.

145 A paradoxical observation of the Bishop Tuff system, and many other zoned ignimbrites
146 (e.g., Carpenter Ridge Tuff; Whitney, 1988; Ammonia Tanks Tuff; Mills et al. 1997; Deering et
147 al. 2011), is that the hottest part of the magma is also the most crystal-rich. Whereas part of the
148 crystal assemblage comprises more mafic crystals, stable at higher temperatures, these refractory
149 crystals do not account for all the crystal population; some of the low-temperature phases
150 (feldspars and quartz) are still present. Sanidine and quartz crystals in the Bishop Tuff show
151 obvious resorption textures and later growth of rims that are: (1) high in Ti in quartz (yielding
152 higher temperature using the Ti-in-Quartz thermometer; Wark et al. 2007; Gualda et al. 2012)
153 and (2) high in Ba in the sanidine (Anderson et al. 2000). Such high Ti-rims in quartz and high
154 Ba rims in sanidine crystals are found in several other localities (both plutonic and volcanic;
155 Hervig and Dunbar 1992; Wiebe et al. 2007; Matthews et al. 2012).

156 The presence of the low temperature minerals with reversely-zoned rims in the late-
157 erupted parts of the deposits not only requires that some of them survived the reheating process,
158 but that they were able to recrystallize shortly prior to eruption in a significantly hotter
159 environment. It is reasonable to think that the short-lived heating event registered by the Ti rims
160 in quartz (< 100 years; Wark et al. 2007), for reasons of kinetics alone, might fail to completely

161 dissolve the granitic mineral assemblage. Three minerals participate in the melting reaction (Qz
162 + Sa + Pl \rightarrow L), and the progress of the reaction involves the kinetics of dissolution of all three
163 of them, as well as diffusion in the liquid of Si, Na, K, and Al. In all these processes, the slowest
164 will dictate the pace. In particular, quartz dissolution is expected to be relatively slow (albeit
165 poorly constrained) at low temperature and high viscosity close to its saturation level (Shaw
166 2006).

167 A more challenging observation to explain is the regrowth of quartz and sanidine at
168 temperatures \geq 780-800 °C. Two processes can be envisaged: (1) the total P dropped
169 significantly, or (2) the total P remained roughly constant, but the $a_{\text{H}_2\text{O}}$ in the melt declined
170 significantly (Whitney 1988; Johnson and Rutherford 1989).

171 A drop in total P would require that the deepest parts of magma body ascended towards
172 the surface. Such a process seems unlikely for the inferred geometry of the reservoir and
173 rheological properties of these silicic magmas. Alternatively, the P drop could be related to
174 eruption and evacuation of the top of the reservoir. Such syn-eruptive crystal growth has been
175 called upon in other systems (Lipman et al. 1997) and is an expected consequence of large-scale
176 evacuation from shallow magma reservoirs (Bachmann et al. 2012). However, the Bishop Tuff
177 appears to have been erupted within a few days (Hildreth and Wilson 2007), which would appear
178 too short for such syn-eruptive growth to occur.

179 Assuming that the total P remained constant, the second possibility to trigger growth of
180 sanidine and quartz at higher T is a drop of $a_{\text{H}_2\text{O}}$. This may be brought about by the injection of
181 CO_2 from magma below (Hildreth and Wilson 2007; Wark et al. 2007). The Bishop Tuff is
182 known to have more CO_2 in its late-erupted melt inclusions (Wallace et al. 1999), and this
183 correlates very well with Ba (Anderson et al. 2000, Fig. 6). Also, there is evidence in several

184 natural systems that deep recharge commonly carries abundant CO₂, fluxing the shallow, H₂O-
185 rich upper crustal reservoir (Bachmann et al. 2009; Humphreys et al. 2009; Blundy et al. 2010).

186 The high-Ba rims in the Bishop Tuff sanidine crystals, within a system dominated by
187 crystal fractionation (in which Ba is highly compatible), has been highlighted as a paradox.
188 Anderson et al. (2000) noted that: (p. 465) “*Mixing can explain the sense of the zoning (... of Ba*
189 *in sanidine...), but not the form of the (...trace element...) correlation. Fractional crystallization*
190 *can explain the form of the correlation but not the sense of the zoning!*”. However, the recharge
191 - *partial remelting* model reported by previous contributions (Hildreth and Wilson 2007; Wark et
192 al. 2007), and highlighted in this paper, accounts for the sense of zoning while still keeping a
193 dominant crystal fractionation signature. The sanidine-rich zones in the deep part of the magma
194 reservoir (mush with accumulation of crystals, including sanidine) can be expected to melt
195 significantly upon recharge, releasing much Ba to the melt. A significant amount of this Ba does
196 not come from the recharge, and hence would not produce a strong mixing trend. As sanidine
197 become stable again (due to the same mechanisms that stabilize quartz, as discussed above) the
198 rims incorporate large amounts of Ba, producing the observed reverse zoning. This dissolution-
199 recrystallization could have happened several times during the growth of the Bishop Tuff magma
200 body as evidenced by longer diffusion timescales calculated for Ti in the cores of some quartz
201 crystals (> 1000 ky; Gualda et al. 2012) and for Ba in some sanidine crystals (10-100 ky
202 depending on temperature of diffusion; Morgan and Blake 2006).

203 **MODEL REFINEMENTS**

204 We propose the following refinements of the model for the Bishop Tuff, largely building
205 on previous work (Hildreth and Wilson 2007; Wark et al. 2007) and on par with multiple other

206 systems, large and small, old and young, in the volcanic and plutonic record (Murphy et al. 2000;
207 Bachmann 2010; Sparks et al. 1977; Wiebe et al. 2004):

208 1. A large silicic mush is present in the upper crust (1.5-3 kb), from which high-SiO₂
209 rhyolitic liquid is extracted, pooling as a highly eruptible melt-dominated region in the
210 upper level of the reservoir (Hildreth 2004; Bachmann and Bergantz 2008). This high-
211 SiO₂ rhyolite is kept at low temperature near the H₂O-saturated haplogranite eutectic for
212 most of the time (~ 700-720 °C), thermally buffered by the large amount of crystal mush
213 around it (Huber et al. 2009) and multiply saturated with feldspars and quartz, as well as
214 biotite and Fe-Ti oxides.

215 2. As this mush is periodically replenished by deeper recharge, some of the hotter, less
216 differentiated melt can penetrate up into the base of the high-SiO₂ rhyolite cap. Hence,
217 the upper part of the mush, transformed into a low-temperature cumulate by progressive
218 melt extraction, can partially remelt and become mixed with the recharge and the base of
219 the rhyolitic cap. Hence, higher temperature, and foreign crystals that are not in chemical
220 equilibrium with the rest of the mineral assemblage (e.g., pyroxene in this case) can be
221 imposed on the lower part of the reservoir.

222 3. Assuming that recharge is a potential eruption trigger (e.g., Sparks et al. 1977; Pallister et
223 al. 1992; Nakagawa et al. 2002; Ruprecht and Bachmann 2010; Druitt et al. 2012), and
224 that the timescales of < 100 years obtained from Ti diffusion in the reversely-zoned
225 quartz in the Bishop Tuff are accurate (Wark et al. 2007), mixing did not have enough
226 time to yield perfect homogenization (Huber et al. 2012). Therefore, the deposits record
227 gradients in temperature, trace elements, and mineral modes that were transient; more
228 mixing would have induced more homogeneous deposits, as are evident in others large

229 silicic provinces, particularly in the Taupo Volcanic zone (Dunbar et al. 1989; Matthews
230 et al. 2012).

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234 Bishop Tuff, Fred Anderson for providing comments on an early version of the manuscript, and
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236 own.

237 **FIGURE CAPTIONS**

238 Figure 1: (a) Δ NNO (b) $\text{Fe}^{2+}/(\text{Fe}^{2+}+\text{Mg})$ Ilm and (c) $\text{Fe}^{2+}/(\text{Fe}^{2+}+\text{Mg})$ Mag as a function of
239 temperature (Ghiorso and Evans 2008) for the Bishop Tuff (data from Hildreth 1979 and
240 Hildreth and Wilson 2007). The temperature of the water-saturated haplogranite eutectic at 2 kb
241 (Johannes and Holtz 1996) is shown for reference. Possibly some of the five lowest temperature
242 data points (below 700 °C) suffered from late- or post-depositional re-equilibration.

243 Figure 2: Roozeboom plot (X_{Mg} Ilmenite vs. X_{Mg} for biotite, orthopyroxene, clinopyroxene and
244 magnetite (for the oxide minerals, the iron component is Fe^{2+} calculated from the formula; for
245 biotite and the pyroxene iron is total Fe).

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