Experimental and textural investigation of welding: effects of compaction, sintering, and vapor-phase crystallization in the rhyolitic Rattlesnake Tuff

Anita L. Grunder a,*, Didier Laporte b, Tim H. Druitt b

a Department of Geosciences, Oregon State University, Corvallis, OR 97331, USA
b Laboratoire Magmas et Volcans, Université Blaise Pascal-CNRS, OPGC, 5 rue Kessler, 63038 Clermont-Ferrand, France

Received 14 November 2003; accepted 8 October 2004

Abstract

The abrupt changes in character of variably welded pyroclastic deposits have invited decades of investigation and classification. We conducted two series of experiments using ash from the nonwelded base of the rhyolitic Rattlesnake Tuff of Oregon, USA, to examine conditions of welding. One series of experiments was conducted at atmospheric pressure (1 At) in a muffle furnace with variable run times and temperature and another series was conducted at 5 MPa and 600 °C in a cold seal apparatus with variable run times and water contents. We compared the results to a suite of incipiently to densely welded, natural samples of the Rattlesnake Tuff.

Experiments at 1 At required a temperature above 900 °C to produce welding, which is in excess of the estimated pre-eruptive magmatic temperature of the tuff. The experiments also yielded globular clast textures unlike the natural tuff. During the cold-seal experiments, the gold sample capsules collapsed in response to sample densification. Textures and densities that closely mimic the natural suite were produced at 5 MPa and 600 °C and 0.4 wt.% H₂O, over run durations of hours to 2 days. Clast deformation and development of foliation in 2-week runs were greater than in natural samples. Both more and less water reduced the degree of welding at otherwise constant run conditions.

For 5 MPa experiments, changes in the degree of foliation of shards and of axial ratios of bubble shards and non-bubble (mainly platy) shards, are consistent with early densification related to compaction and partial rotation of shards into a foliation. Subsequent densification was associated with viscous deformation as indicated by more sintered contacts and deformation of shards. Sintering (local fusion of shard–shard contacts) was increasingly important with longer run times, higher temperatures, and greater pressures. During runs with high water concentrations, sintering was rare and adhesion between clasts was dominated by precipitation of sublimates in pore spaces.
A few tenths wt.% H₂O in the rhyolite glass promote the development of welding by sharp reduction of glass viscosity. Large amounts of water inhibit welding by creating surface sublimates that interfere with sintering and may exert fluid pressure counter to lithostatic load if sintering and vapor-phase sublimates seal permeability in the tuff.

© 2004 Elsevier B.V. All rights reserved.

Keywords: welding; ignimbrite; sintering; vapor-phase crystallization

1. Introduction

Welding in ignimbrites is expressed by loss of pore space, flattening of pyroclastic particles and sintering of glassy clasts (Smith, 1960a,b). The appearance of nonwelded pyroclastic rocks differs dramatically from that of welded equivalents, making lateral correlation of units difficult. Transitions between welding facies can be exceedingly abrupt both vertically and laterally. Vertical transitions from nonwelded to densely welded can occur over a few centimeters and lateral changes can occur over a few tens of meters (e.g., Smith, 1960b; Briggs, 1976; Kamata et al., 1993; Streck and Grunder, 1995). The abruptness of transitions between welding facies suggests that the conditions that promote welding combine to overcome a critical threshold.

The main factors considered to control welding are compaction by loading, accompanied by expulsion of interstitial gas, and the deformation and sintering of glassy pyroclasts when they reside above the glass transition temperature (e.g., Smith, 1960a,b; Ross and Smith, 1961; Guest and Rogers, 1967; Riehle et al., 1995; Giordano et al., 2000). Sintering of glassy clasts, in turn, depends on the temperature, composition and water concentration of the glass, inasmuch as these govern viscosity, as well as the shape and size distribution of particles (e.g., White, 1962; Smith, 1960a; Friedman et al., 1963; Guest and Rogers, 1967; Schmincke, 1974; Sparks et al., 1999).

Experimental studies to determine the conditions of welding for silicic pyroclastic deposits indicate that high temperatures, in excess of 900 °C, are required at 1 atmosphere (At) (Yagi, 1962, 1966). With the addition of load, the temperature for dense welding (i.e., virtually no porosity) was reduced to 800 °C for anhydrous silicic ash (Bandelier Tuff and Mt. St. Helens) at ~3 MPa (Bierwirth, 1982) and further reduced to about 750 °C at ~5 MPa (Boyd, 1961), with run times of a few days. Lower temperatures of dense welding, 635 °C and 685 °C, were achieved for rhyolite ash with the addition of water (Bandelier Tuff—Friedman et al., 1963; Yellowstone—Boyd, 1961, respectively).

To further investigate the conditions of welding, we conducted a series of experiments using natural rhyolitic ash from the 7.1-Ma Rattlesnake Tuff of eastern Oregon. One set of experiments was conducted at 1 At from 600 to ~1200 °C and another was conducted at 5 MPa and 600 °C (plus a few experiments at 1 and 20 MPa) in which run duration and water concentration were varied. We here compare the results of the experiments to variations in density, foliation and clast deformation associated with natural welding of the Rattlesnake Tuff.

2. The Rattlesnake Tuff as starting material

A sample of glassy nonwelded ignimbrite from the base of the Rattlesnake Tuff was used as starting material for a set of welding experiments (Sample RT-62). This ignimbrite is typically a few tens of meters thick and is exposed over an area greater than 9000 km² (Streck and Grunder, 1995). The Rattlesnake Tuff is nonwelded to partially welded with pumice in sections distal from the inferred source, but exhibits dense welding and strong welding zonation proximally. Nonwelded Rattlesnake Tuff is nearly entirely white to light gray ash with no adhesion of clasts. The ash has sorting parameter, σₚ, of 1.2, with median and mean grain size of 290 and 280 μm, respectively, and is better sorted than most ignimbrites (typical σₚ in the range of 2 to 4; Walker, 1971). Shard shapes are mainly platy to cuspatc with mean aspect ratio (length over width) of 5.1 to 5.5, with subordinate pumiceous ash as well as sparse, nearly spherical bubble shards.
with mean aspect ratios of 1.3 (Fig. 1E, Table 1A). The surface of the glass shards in the starting material is minutely pocked when viewed by scanning electron microscope (SEM) (Fig. 2E).

The Rattlesnake Tuff rhyolite is metaluminous to slightly peralkaline. Pumice clasts and fiamme collected from the ignimbrite define five compositional clusters that range from 75.4 to 77.7 wt.% SiO$_2$ and from 2 to 0.8 wt.% FeO, respectively (calculated as 100% volatile free; Streck and Grunder, 1997). The color of the ejecta, including all morphological types of ash, varies with increasing FeO from clear to brown in thin section (Fig. 1), or white to gray in hand sample, respectively. The ash and pumice are nearly aphyric, with a maximum of about 1.3 wt.% phenocrysts in the most Fe-rich compositions. Juvenile dacitic pumice clasts and small quenched mafic inclusions are sparse (<1%) constituents in the Rattlesnake Tuff.

Inferred pre-eruptive zircon saturation temperatures range from about 800 to 880 °C for the most evolved and least evolved rhyolite, respectively (Streck and Grunder, 1997). Normative feldspar components of the rhyolites are consistent with differentiation at about 100 MPa and slight water-undersaturation (ibid.).

Streck and Grunder (1995) describe five facies of welding increase; with increasing welding intensity these are nonwelded, incipiently welded, partially welded with pumice, partially welded with fiamme and densely welded. The increase in welding is attended by a change in color from pale to dark gray and increased vitreous luster culminating in dense black glass of the densely welded facies, where it is not devitrified. Like in many ignimbrites, welding zonation is overprinted by devitrification and vapor-phase crystallization textures, particularly in upper portions of the unit.

The density of the ignimbrite and adhesion and deformation of shards increase with welding (Fig. 1A–E). Densities of natural samples examined in this study range from about 1 g cm$^{-3}$ in the loose, nonwelded tuff to 2.3 g cm$^{-3}$ for the densely welded facies (Table 1A) (c.f. 2.34 g cm$^{-3}$ for densely welded facies of Streck and Grunder, 1995). The increase in density is equivalent to a decrease in porosity from 65% to ~0, assuming that the porosity is interconnected, which appears generally true in welded pyroclastic deposits (e.g., Ragan and Sheridan, 1972; Sparks and Wright, 1979; Quane and Russell, 2004). The preferred orientation of white (low-Fe), platy and cuspatte shards increases significantly from random in the nonwelded facies, to moderately foliated in incipiently and partially welded facies, to strongly foliated in the densely welded facies (Table 1 and see below for methods). Aspect ratios (length relative to thickness) of white, platy and cuspatte shards viewed in sections perpendicular to the foliation range little (from 5.1 to 5.8) with the higher values corresponding to the more densely welded facies. In contrast, the aspect ratio of bubble shards, most of which are white, increases incrementally from 1.3 to about 4 with increasing welding and reaches a value of 10 in the densely welded sample.

Sintered contacts between grains, where glassy fragments have locally fused, are present but rare in the “incipiently welded” facies (Figs. 2A and 3A). Adhesion among clasts appears mainly to result from encrustation on the shard surfaces and contacts (Fig. 3B). Sintered contacts are common in the partially welded facies with pumice (Fig. 2B). With increased welding to partially welded with fiamme, virtually all shards are sintered to neighboring shards. Thin seams of pore space and surface sublimes are preserved along contacts. In the densely welded facies, contacts among grains are completely fused with only rare small pockets of porosity. Although sparse, these spaces are partially filled by grains with a sugary texture that are most likely vapor-phase sublimates (Fig. 2D).

3. Experimental and analytical techniques

One set of experiments was conducted by heating Rattlesnake Tuff ash in alumina ceramic crucibles at 1 At in a muffle furnace, with temperatures ranging from 600 to 1200 °C and run times from 1 h to 3 days (60–4405 min; Table 1C). Another set of experiments was run in gold capsules in an externally heated pressure vessel made of nickel-based superalloy (Nimonic 105) and pressurized with nitrogen gas. Most experiments were conducted at 600 °C and 5 MPa with variations in water content from 0.1 to 1.6 wt.% (Table 1B; Fig. 6A). A few experiments were conducted at 600 °C, but 1
and 20 MPa, and at 700 °C and 1 MPa with variable water.

The rationale for using 600 °C as the main experimental temperature is that it is slightly lower than temperatures at which dense welding has been achieved experimentally (i.e., 620 °C, Boyd, 1961; 635 °C, Friedman et al., 1963). Also, 600 °C is at the hot end of the temperature spectrum measured in cooling, unwelded pyroclastic flow deposits (e.g., Ryan et al., 1990).

The starting material for the experiments had an initial density of only 0.74 g cm$^{-3}$ (66% porosity), when completely uncompacted. The density increased to 1.34 g cm$^{-3}$ (43% porosity) after light compaction of the ash by tapping on the container. The sample was sieved to <550 μm and dehydrated at 500 °C at 1 atm for 2 days, after which it contained 0.1 wt.% H$_2$O. The ash was examined in oils to confirm that the particles were vitric both prior to and after dehydration. The water concentration was determined by comparison to the weight of material fully dehydrated at 1100 °C overnight. The ash initially contained 3.68 wt.% water as determined by loss on ignition; a few grams of ash were loaded into an Pt tube that was welded at one end and had been prebaked. The ash was then slowly heated to 1100 °C over a few hours and held at 1100 °C for 2 h. An incremental heating experiment conducted over about 10 h determined that 75% of the water was lost between 300 and 500 °C. Samples were cooled to room temperature for weighing.

For pressurized experiments, about 100 mg of sample were sealed, by welding, into a ~1.5-cm-long gold capsule with a 0.2 mm wall and 4 mm diameter. Water was added to most of the charges as a controlled weight of silicic acid (containing 10.4±0.1 wt.% H$_2$O, as determined by loss on ignition at 250 °C for 2 h); the weight of the starting material and silicic acid were determined using a balance precise to ±0.05 mg. In one instance, liquid water was added by micro-syringe and in others the glass was partially dehydrated to the desired starting water concentration (Table 1B). The sealed capsules were weighed before and after the run to assure that the charges did not leak water. The glass shards should have reached equilibrium with the added water in ~2 h, given their typical short dimensions of ≤50 μm and water diffusivity of $3\times10^{-13}$ at 600 °C (Zhang et al., 1991).

The capsule was loaded in the pressure vessel with its long axis vertical. Pressure was applied and the temperature was then increased to the target temperature in about 50 min. Run times ranged from 1 h to 1 month, after which the pressure vessel was taken out of the furnace and was cooled with compressed air. The pressure was then released, the cooled capsule was weighed, punctured and weighed again, placed in an oven at 150 °C for half an hour and weighed again.

During the course of the experiment, the gold capsules collapsed to variable extents due to densification of the samples. Collapse was not isotropic; the change in volume during the experiment was accommodated by axial shortening perpendicular to a plane defined by the long axis of the capsule and the straight welding seam at the top of the capsule. Shortening of the capsule was accompanied by the development of a flattening foliation in the sample parallel to the length of the capsule. We assumed coaxial strain.

Run products were cut out of the capsule; for those that were coherent, one piece was mounted for scanning electron imaging and another piece was mounted for thin sectioning, both with the fractured or cut surface perpendicular to the flattening foliation. Additional fragments were used for density

Fig. 1. Textures of natural samples (A–E) and experimental products (F–H) of Rattlesnake Tuff in thin section; sections of experiment products were mounted in dyed epoxy. Experimental conditions were 5 MPa, 600 °C, 0.4 wt.% H$_2$O and variable run times. bs indicates bubble shards and ps (E) indicates pumice shard. Note the color difference among shards, which is related to variable composition, mainly FeO concentration. Textural and density data are in Table 1. (A) Sample 96 is incipiently welded tuff (according to textural criteria of Streck and Grunder, 1995). (B) Sample 115.1 is partially welded with pumice. (C) Sample 177T3 is partially welded with fiamme. The lower margin of a fiamme is at the top of the image. (D) Sample 102 is densely welded. Small fiamme are present and a green equant clinopyroxene is marked cpx. The marked bubble shards are empty; filled bubble shards were not used for textural analysis. (E) Starting material sample RT-62. (F) Run product RW-11; the interior of one bubble shard is emphasized with an ellipse. (G) Run product RW-7. (H) Run product RW-8. In this sample, bubble shards are difficult to distinguish from shards that have color zonation from white exterior to brown interior without the microscope. Color zonation within shards also developed in other runs with high water concentration and (or) long run times (RW-20 through 25 and RW-3) and is presumed to reflect oxidation.
Table 1

Textural and density data for Rattlesnake Tuff natural samples and experimental runs

A. Natural samples

<table>
<thead>
<tr>
<th></th>
<th>Density (g cm$^{-3}$)</th>
<th>Porosity %</th>
<th>Sample color</th>
<th>L/W n-b shards n</th>
<th>L/W bubble shards n</th>
<th>sigma n</th>
</tr>
</thead>
<tbody>
<tr>
<td>RT-62 nonwelded (nw)</td>
<td>~1 (VI)</td>
<td>57</td>
<td>white, light gray</td>
<td>5.3</td>
<td>27</td>
<td>1.3</td>
</tr>
<tr>
<td>96 incipiently welded</td>
<td>1.43</td>
<td>38</td>
<td>light gray</td>
<td>5.5</td>
<td>37</td>
<td>1.8</td>
</tr>
<tr>
<td>115.1 partially welded with pumice</td>
<td>1.63</td>
<td>29</td>
<td>light gray</td>
<td>5.1</td>
<td>36</td>
<td>2.5</td>
</tr>
<tr>
<td>177T3 partially welded with fiamme densely welded (bulk)</td>
<td>2.18</td>
<td>6</td>
<td>medium gray</td>
<td>5.8</td>
<td>79</td>
<td>4.4</td>
</tr>
<tr>
<td>102 white shards</td>
<td></td>
<td></td>
<td></td>
<td>5.7</td>
<td>37</td>
<td></td>
</tr>
<tr>
<td>102 light brown shards</td>
<td></td>
<td></td>
<td></td>
<td>30</td>
<td>51</td>
<td></td>
</tr>
<tr>
<td>102 dark brown shards</td>
<td></td>
<td></td>
<td></td>
<td>24</td>
<td>17</td>
<td></td>
</tr>
</tbody>
</table>

B. Gold capsule experiments, at 600 °C (h)

<table>
<thead>
<tr>
<th>Run no.</th>
<th>Time (min)</th>
<th>$P$ (MPa)</th>
<th>H$_2$O (VII) wt. %</th>
<th>Density (g cm$^{-3}$)</th>
<th>Porosity %</th>
<th>Sample color</th>
<th>L/W shards n</th>
<th>L/W bubble shards n</th>
<th>sigma n</th>
</tr>
</thead>
<tbody>
<tr>
<td>RW-4</td>
<td>4305 5</td>
<td>0.1</td>
<td>1.31</td>
<td>43</td>
<td>pinkish gray</td>
<td>5.1</td>
<td>42</td>
<td>1.8</td>
<td>6 35 49</td>
</tr>
<tr>
<td>RW-23</td>
<td>20160 5</td>
<td>0.1</td>
<td>1.77</td>
<td>23</td>
<td>near black</td>
<td>5.8</td>
<td>52</td>
<td>2.0</td>
<td>4 42 54</td>
</tr>
<tr>
<td>RW-1</td>
<td>120 5</td>
<td>0.4</td>
<td>1.56</td>
<td>32</td>
<td>grayish orange</td>
<td>5.0</td>
<td>55</td>
<td>1.4</td>
<td>5 48 31</td>
</tr>
<tr>
<td>RW-12</td>
<td>240 5</td>
<td>0.4</td>
<td>1.61</td>
<td>30</td>
<td>orangish gray</td>
<td>5.3</td>
<td>22</td>
<td>1.3</td>
<td>5 51 28</td>
</tr>
<tr>
<td>RW-11</td>
<td>1440 5</td>
<td>0.4</td>
<td>1.79</td>
<td>23</td>
<td>black</td>
<td>5.5</td>
<td>42</td>
<td>1.6</td>
<td>8 37 56</td>
</tr>
<tr>
<td>RW-7</td>
<td>2895 5</td>
<td>0.4</td>
<td>2.11</td>
<td>9</td>
<td>black</td>
<td>5.7</td>
<td>42</td>
<td>3.8</td>
<td>8 20 41</td>
</tr>
<tr>
<td>RW-8</td>
<td>20160 5</td>
<td>0.4</td>
<td>2.20</td>
<td>5</td>
<td>black</td>
<td>7.9</td>
<td>51</td>
<td>9.0</td>
<td>3 9 28</td>
</tr>
<tr>
<td>RW-14</td>
<td>49040 5</td>
<td>0.4</td>
<td>2.16</td>
<td>6</td>
<td>black</td>
<td>7.9</td>
<td>51</td>
<td>9.0</td>
<td>3 9 28</td>
</tr>
<tr>
<td>RW-13</td>
<td>1440 5</td>
<td>0.8</td>
<td>1.65</td>
<td>29</td>
<td>black</td>
<td>7.9</td>
<td>51</td>
<td>9.0</td>
<td>3 9 28</td>
</tr>
<tr>
<td>RW-22</td>
<td>2880 5</td>
<td>0.8</td>
<td>1.70</td>
<td>26</td>
<td>near black</td>
<td>5.9</td>
<td>32</td>
<td>1.9</td>
<td>5 36 44</td>
</tr>
<tr>
<td>RW-20 (VIII)</td>
<td>20160 5</td>
<td>0.8</td>
<td>1.67</td>
<td>28</td>
<td>orangish gray</td>
<td>7.0</td>
<td>38</td>
<td>2.3</td>
<td>6 25(35) 47(23)</td>
</tr>
<tr>
<td>RW-25</td>
<td>2880 5</td>
<td>1.1</td>
<td>1.45</td>
<td>37</td>
<td>near black</td>
<td>5.7</td>
<td>62</td>
<td>1.5</td>
<td>6 37 37</td>
</tr>
<tr>
<td>RW-24</td>
<td>20160 5</td>
<td>1.1</td>
<td>1.68</td>
<td>27</td>
<td>near black</td>
<td>5.6</td>
<td>44</td>
<td>1.5</td>
<td>5 38 43</td>
</tr>
<tr>
<td>RW-2</td>
<td>120 5</td>
<td>1.2</td>
<td>nw</td>
<td>orangish gray</td>
<td>5.1</td>
<td>38</td>
<td>1.3</td>
<td>2 nw</td>
<td></td>
</tr>
<tr>
<td>RW-3</td>
<td>2880 5</td>
<td>1.6</td>
<td>nw</td>
<td>black</td>
<td>5.3</td>
<td>33</td>
<td>1.3</td>
<td>9 nw</td>
<td></td>
</tr>
<tr>
<td>RW-16</td>
<td>2880 1</td>
<td>0.2</td>
<td>nw</td>
<td>orangish gray</td>
<td></td>
<td></td>
<td></td>
<td></td>
<td></td>
</tr>
<tr>
<td>RW-17 (IX)</td>
<td>2760 1</td>
<td>0.2</td>
<td>1.15</td>
<td>50</td>
<td>near black</td>
<td></td>
<td></td>
<td></td>
<td></td>
</tr>
<tr>
<td>RW-15</td>
<td>4140 1</td>
<td>0.4</td>
<td>nw</td>
<td>gray</td>
<td></td>
<td></td>
<td></td>
<td></td>
<td></td>
</tr>
<tr>
<td>RW-5</td>
<td>120 20</td>
<td>0.1</td>
<td>1.29</td>
<td>44</td>
<td>light orange</td>
<td>5.5</td>
<td>40</td>
<td>1.4</td>
<td>4 51 70</td>
</tr>
<tr>
<td>RW-6</td>
<td>2860 20</td>
<td>0.5</td>
<td>2.31</td>
<td>0</td>
<td>black</td>
<td>5.6</td>
<td>35</td>
<td>(X)</td>
<td>54 53 70</td>
</tr>
</tbody>
</table>

C. 1 Atmosphere experiments

<table>
<thead>
<tr>
<th>Run no.</th>
<th>Time (min)</th>
<th>Temp. (°C)</th>
<th>Density (g cm$^{-3}$)</th>
<th>Sample color (III)</th>
</tr>
</thead>
<tbody>
<tr>
<td>R-0.1</td>
<td>60</td>
<td>700</td>
<td>nw</td>
<td>yellowish, loose</td>
</tr>
<tr>
<td>R-0.2</td>
<td>75</td>
<td>830</td>
<td>nw</td>
<td>light orange, loose</td>
</tr>
<tr>
<td>R-1</td>
<td>120</td>
<td>900</td>
<td>nw</td>
<td>light orange, loose</td>
</tr>
<tr>
<td>R-2</td>
<td>115</td>
<td>1030</td>
<td>1.12</td>
<td>rose-orange, sintered</td>
</tr>
<tr>
<td>R-3</td>
<td>1050</td>
<td>1030</td>
<td>1.09</td>
<td>brick red and sintered</td>
</tr>
<tr>
<td>R-5</td>
<td>4405</td>
<td>1030</td>
<td>1.41</td>
<td>vitric brick red, pulled from walls</td>
</tr>
<tr>
<td>R-6</td>
<td>280</td>
<td>1200</td>
<td>2.15</td>
<td>vitric black, wetted to wall</td>
</tr>
<tr>
<td>R-8</td>
<td>285</td>
<td>1100</td>
<td>1.75</td>
<td>thready brick red pulling from sides of vessel</td>
</tr>
<tr>
<td>R-11</td>
<td>120</td>
<td>1225</td>
<td>2.01</td>
<td>vitric, dark brick red, pulling from sides</td>
</tr>
<tr>
<td>R-14</td>
<td>2550</td>
<td>1200</td>
<td>2.09</td>
<td>vitric, dark gray, wetted to wall</td>
</tr>
<tr>
<td>R-20</td>
<td>840</td>
<td>900</td>
<td>nw</td>
<td>orange, loose</td>
</tr>
<tr>
<td>R-23</td>
<td>60</td>
<td>1030</td>
<td>~1</td>
<td>orange, barely coherent</td>
</tr>
</tbody>
</table>
analysis. Samples in ceramic crucibles were cut in half with a saw and a piece was removed for density determination. The temperature of the pressurized experiments was monitored by a Pt/Pt10%Rh thermocouple inserted into the end of the bomb near the experimental charge. The thermocouple was calibrated to the melting temperature of RbCl salt. Temperature was kept within ±8°C of the desired value during the run. Pressures ≥5 MPa were measured by a transducer precise to 0.1 MPa, and were maintained to ±0.2 MPa during the experiments. In the 1-MPa experiments, pressure was measured using a 4-MPa gauge precise to 0.05 MPa, and was maintained within ±0.1 MPa.

Three main attributes were quantified for run products: density, shard orientation and shard shape (Table 1). Densities were obtained by comparing the weights of the sample in air versus water of monitored temperature. Porous samples were sprayed with silicon water repellent to prevent invasion of water into the pores. The densities of samples sprayed with silicon were slightly affected not only by the addition of the low-density spray (0.71 g cm⁻³), but also by the adherence of tiny bubbles to the sprayed surface once immersed in water. The estimated precision of the density measurement was ±0.1 g cm⁻³ for the small samples produced in experiments and that were sprayed with silicon and ±0.05 for larger (natural) samples or unsprayed samples.

The orientation of shards was measured in thin sections as an angular deviation of the long axes of the shards from an arbitrary plane (here chosen as the plane defined by the foliation). The degree of preferred orientation (foliation) was cast as a 1 sigma standard deviation of shard axis orientations from the mean orientation, analogous to the fabric orientation angle of Dunnet (1969). This two-dimensional view of the flattening is hereafter referred to as the foliation of shards. For the densely welded sample (102), the foliation of shards was compared for shard populations of different color (Table 1A) and thus composition. The white shards reflect the overall foliation pattern; measurements of foliation on other samples and experimental charges focused on white shards.

Deformation of non-bubble shards and bubble shards was determined from the ratio of their long axes to their short axes in thin sections cut perpendicular to foliation. For deformation of non-bubble shards, only white shards were measured. Bubble shards were not deformed significantly enough to measure accurately in this study. For bubble shards, deformation was measured in thin sections cut parallel to foliation as an angular deviation of the short axes of the shards from the foliation plane. The degree of preferred orientation (fooling of bubble shards) was cast as a 1 sigma standard deviation of shard axis orientations from the mean orientation, analogous to the fabric orientation angle of Dunnet (1969). This two-dimensional view of the flattening is hereafter referred to as the foliation of bubble shards.

### Table 1 (continued)

<table>
<thead>
<tr>
<th>Run no.</th>
<th>Time (min)</th>
<th>Temp. (°C)</th>
<th>Density (g cm⁻³)</th>
<th>Sample color (III)</th>
</tr>
</thead>
<tbody>
<tr>
<td>R-24</td>
<td>120</td>
<td>1100</td>
<td>1.20</td>
<td>orange red, sintered</td>
</tr>
<tr>
<td>R-25</td>
<td>420</td>
<td>1100</td>
<td>1.60</td>
<td>brick red, sintered</td>
</tr>
<tr>
<td>R-26</td>
<td>865</td>
<td>600</td>
<td>nw</td>
<td>light yellow gray, loose</td>
</tr>
</tbody>
</table>

nw=nwelded, no density or alignment measurements done on nw experiments.

I. Measurements compensated for silicon spray estimated precision ±0.1. Samples 6, 14, and 102 not sprayed.

II. Porosity calculated as the ratio of (density sample–2.3)/2.3 where 2.3=bulk density.

III. Color is from visual appearance, mainly dictated by changes in color of the dark shards.

IV. L/W (length/width) for non-bubble (n-b) white shards, except as indicated for sample 102. n=number measured.

V. Shard foliation as 1 σ standard deviation of the mean of shard orientations measured relative to a plane (see methods in text).

VI. Density measured by weight of known volume of loose ash in graduated cylinder. Completely uncompacted, density=0.74 g cm⁻³. Lightly compacted density=1.35 g cm⁻³.

VII. H₂O added as silicic acid to runs with >0.1 wt.% H₂O; for RW-2 and 3, H₂O was added by microsyringe.

VIII. Numbers in parentheses are for small, distinctive textural domain in charge with shards ~40° from norm.

IX. Runs all at 600 except RW-17, run at 700°C.

X. No bubble shards recognized.

---

**Fig. 2.** Scanning electron micrographs of natural samples (A–D) and experimental products (E–H) of Rattlesnake Tuff. Experimental conditions were 5 MPa, 600 °C, 0.4 wt.% H₂O and variable run times. Samples (A–D) are labeled according to textural criteria for welding facies of Strack and Grunder (1995). Sample numbers are as in A–D in Fig. 1. The box in A outlines the position of the magnified view in Fig. 3A. (E) Run product RW-12 illustrates the relict pocked texture on the surface of the starting material as well as one of the scarce sintered contacts between grains. There is slight development of encrusting crystallites. (F) Run product RW-11. (G) Run product RW-7. (H) Run product RW-8 (Table 1).

incipiently welded

partially welded with pumice

partially welded with fiamme

densely welded

50 µm G.

5 MPa, 600°C, 0.4% 2 days

50 µm B.

5 MPa, 600°C, 0.4% 4 hrs

50 µm A.

partially welded with pumice

5 MPa, 600°C, 0.4% 2 weeks

50 µm D.

5 MPa, 600°C, 0.4% 4 hrs

50 µm E.

5 MPa, 600°C, 0.4% 2 days

50 µm F.

5 MPa, 600°C, 0.4% 1 day

50 µm H.

5 MPa, 600°C, 0.4% 2 weeks
shards of any color were measured, but only those that were, empty, individual bubble shards, as opposed to glass-filled bubble shards or bubbles in pumiceous shards, which are typically elongated (lineate).

In addition to the quantitative attributes above, several qualitative attributes were taken into consideration, including clast-to-clast adhesion relationships and the abundance of vapor-phase sublimates. These attributes were characterized by examining fracture surfaces of doubly coated (carbon and gold) fragments of the experimental samples using the secondary-electron imaging mode of the scanning electron microscope.

4. Results

4.1. Experiments at 1 atmosphere

At atmospheric pressure, the Rattlesnake Tuff ash samples did not weld in runs at temperatures of 900 °C or less despite run times as long as 14 h. Adhesion

![Image of welded natural samples and run products]

Fig. 3. Scanning electron micrographs of welded natural samples and run products. (A) and (B) are natural incipiently welded tuff, sample 96. (A) A detailed view of the sintered contact illustrated in Fig. 2A. (B) Detailed view of incipiently welded sample 96 with sublimates encrusting the grain surfaces and grain contacts. (C) Run product RW-3, with well-formed surface sublimates (5 MPa, 600 °C, 1.6 wt.% H₂O, and 2-day run time). (D) Run product RW-20, with ubiquitous surface encrustation by crystals. None of the contacts are sintered (5 MPa, 600 °C, 0.8 wt.% H₂O, and 14-day run time).

![Graph of final density vs. temperature]

Fig. 4. The final density of 1 At experiments increases with temperature. The run time, in minutes, is in italics next to each datum. The open point at 900 °C is the final density after the run longest run time at that temperature that resulted in a nonwelded sample.
of clasts, with attendant densification to about 1 g cm$^{-3}$, occurred between 900 and 1030 °C (Fig. 4). The density of welded ash increased with temperature and run duration. At 1030 °C, density varied little between runs of 1 and 17 h, and only increased significantly after 3 days. At 1200 °C, samples became equally dense for run times of a few hours to nearly 2 days (2550 min).

The general textural evolution during densification at 1 At was a smoothing of shard surfaces and a reduction in pore space; angular spaces between angular shards changed to subangular spaces between globular shards and finally near-round bubbles in completely fused glass (Fig. 5). While most of the rounded spaces in the welded material are probably texturally modified relicts of original pore spaces (Rust et al., 2003), some may be vesicles caused by water exsolution, as the starting material was not completely dehydrated (Gottsman and Dingwell, 2001).

The color of the ash changed from near-white to pale orange and then brick red with increasing time up to 1100 °C. The three experiments near 1200 °C produced black, conspicuously glassy samples with densities of ~2.1 g cm$^{-3}$, equivalent to a porosity of 9% (using bulk density of 2.3 g cm$^{-3}$).

With increased welding, the samples contracted, pulling away from the wall of the crucible and leaving vitric threads between the welded ash and the wall. Only the longest run at 1200 °C (2550 min) resulted in a welded product wetting the crucible wall.

4.2. 5 MPa experiments

Experiments conducted at 600 °C and 5 MPa resulted in a sharp initial increase in sample density with time, followed by a reduction in densification rate at a given water concentration. For the suite with 0.4 wt.% H$_2$O, density increased sharply and constantly from 2 h to 2 days and stayed at the same density from 2 days to 1 month (Fig. 6B). Foliation of shards developed rapidly, in concert with the density increase, and continued to develop at a lesser rate even as density became constant (Fig. 7A). Experiments conducted with either more or less than 0.4 wt.% H$_2$O produced less dense samples with lesser

---

Fig. 5. Scanning electron micrographs of 1 At experiments with run temperatures and times indicated. (A) Experiment R-2; (B) Experiment R-3; (C) Experiment R-8; (D) Experiment R-6.
foliation of clasts (Table 1B). Deformation of bubble shards was similarly greatest in the suite run at 0.4 wt.% H2O, recording the greatest strain rate between 1 and 2 days (Fig. 7B). Significant deformation of other shards was achieved only in the longer runs at 0.4 and 0.8 wt.% H2O (Fig. 7C). The SEM images of experimental products run with 0.4 wt.% H2O are similar to those of natural samples in having an increased proportion of sintered contacts with increasing density and the ubiquitous presence of tiny crystals on shard surfaces and contacts (Fig. 2).

The extent of mineral encrustation on shard surfaces was greater in samples run with more water. Samples run with 0.1 wt.% H2O developed only sparse surface sublimates. For samples with 0.4 wt.% H2O, surface sublimates made a discontinuous layer
nearly 1 μm thick (Fig. 2). The samples with ≥0.8 wt.% H₂O developed a thick (≥1 μm) continuous surface coating of sublimates and had virtually no sintered contacts (Fig. 3C and D). To a lesser degree, mineral encrustation on shard surfaces also became better developed with time (Fig. 3C vs. D). Sample RW-20 (2-week run time at 0.8 wt.% H₂O) is particularly noteworthy as it has well-developed surface sublimates (Fig. 3D), little deformation of bubble shards, but has substantial deformation of non-bubble shards (Table 1B).

Qualitative EDS analysis of surface sublimates yielded peaks for Si, Al, O, K, and Na (analyzed in charge RW-3). We take these crystallites to be alkali feldspar plus possibly a silica phase, both typical vapor-phase crystallization products in silicic ignimbrites. In some instances, particularly for longer run times, the crystallites had clearly developed crystal faces (Fig. 3C); in other cases they formed small botryoidal clumps that are likely to be radial clusters of crystallites. We observed no devitrification of the glass shards.

4.3. 1 and 20 MPa experiments

Of the three experiments conducted at 1 MPa, the two at 600 °C yielded loose ash and the one at 700 °C produced a barely coherent sample. An increase from 1 to 5 MPa, at 600 °C, caused nonwelded ash to become densely welded (density increase to 2.1 g cm⁻³) (compare 0.2 and 0.4 wt.% H₂O experiments at 1 MPa and RW-7 with 0.4 wt.% H₂O at 5 MPa; Table 1B, Fig. 6A). At 600 °C, rhyolite is water saturated at 0.46 wt.% H₂O for 1 MPa and at 1.02 wt.% H₂O for 5 MPa (Newman and Lowenstern, 2002). A further
increase to 20 MPa (with 0.5 wt.% H₂O) induced only a slight further increase in density (compare RW-7 and -6, Table 1B; Fig. 6A). This 20 MPa experiment was the only one to produce a sample with no porosity (density of 2.31 g cm⁻³) and no identifiable relict bubble shards. Although intensely welded, as confirmed by the absence of distinct grains in SEM images, the sample did not develop foliation or deformation of the non-bubble shards, as measured by their axial ratios (Table 1B).

5. Discussion

The suite of samples run at 600 °C, 5 MPa and 0.4 wt.% H₂O most closely approximated both the densities and textures observed in natural samples (Figs. 1, 2, 6, 7). We first discuss physical changes within that suite and then compare them with natural samples and other experimental runs.

5.1. The suite at 600 °C, 5 MPa and 0.4 wt.% H₂O

In this suite (600 °C, 5 MPa and 0.4 wt.% H₂O) the densification of ash with time took place in three stages. The first stage (between 0 and 4 h) was compaction by spatial rearrangement of shards under the effect of external gas pressure, but with little or no alignment or deformation of clasts (Figs. 6B and 7). About 70% of full densification (1.6/2.3) was achieved in this stage. During the second stage of densification (from 4 h to 2 days), volume loss by compaction continued to be the main densification mechanism; about 90% of full densification was achieved and was accompanied by substantial sintering (Fig. 2B,C). About two thirds of the shard foliation fabric developed by rotation of shards into the foliation plane, unaccompanied by significant deformation of the non-bubble shards, consistent with volume strain. Bubble shards flattened from near spherical to axial ratios of about 4. Using an initial value of axial ratio of 1.3 and assuming alignment of the long axes in the foliation, then the strain rate was at least on the order of 10⁻⁵ s⁻¹ (Fig. 7B), and most of the strain in this stage was achieved between 1 and 2 days. The third stage of densification (from 2 days to 2 weeks) represented a change to significant shard deformation. Density increased only little, but most of the bubble shard deformation and essentially all of the non-bubble shard deformation occurred. Estimated strain rates were 10 times greater for bubble shards than for non-bubble shards (using assumptions as above, and initial axial ratio of 5.2 for non-bubble shards), because they were accommodating both pore space reduction (volume strain) and viscous deformation resulting in pure shear.

The experimental charge of 2-day duration closely mimicked the natural sample of the facies “partially welded with fiamme” with respect to density, sintering textures and textural attributes (Figs. 1C and G and 2C and G) except that foliation was more strongly developed in the experimental charge (Fig. 3A). The most telling difference between the natural and experimental suites is among dense samples. Deformation of non-bubble shards and shard foliation are more developed in the 14-day experimental charge than in the densely welded natural sample, indicating greater ductile deformation in the experiment. The greater strain recorded in the experiment can be attributed, in part, to the greater pressure of the experiments compared to pressures achieved in the accumulating Rattlesnake Tuff. The ignimbrite is rarely more than 40 m thick (close to 1 MPa lithostatic load).

At 1 MPa and 0.2 wt.% H₂O and virtually the same run time, welding was not achieved at 600 °C and only barely at 700 °C, suggesting that the Rattlesnake Tuff emplacement temperature was higher or that welding time was greater than 2 days. The absence of significant deformation of non-bubble shards, on the other hand, suggests that the ignimbrite was not in a viscous deformation regime for very long.

5.1.1. Temperature

Increased temperature dramatically enhances welding, as expressed by the density increase accompanying sintering at 1 At (Fig. 4). The minimum welding temperature is between 900 and 1000 °C, and is in keeping with minimum welding temperatures for rhyolite of 900 °C and 920 °C obtained by Yagi (1966) and Taneda (1957), respectively. The four experiments conducted at 1030 °C underwent about half the rate of densification compared with the three experiments at 1100 °C. At high temperature (see 1200 °C runs, Fig. 4), time is less important in producing sintering. These temperatures are unreal-
istic for most natural settings as they are at or above the magmatic temperature of many, even most, rhyolites that occur as densely welded ignimbrites. Nevertheless, the globular textures we observed may have analogs among very hot or very low viscosity pyroclastic deposits transitional to agglutinates (e.g., Hay et al., 1979; Soriano et al., 2002).

A minimum welding temperature near 600 °C for natural silicic (not peralkaline) glasses emerges from compilation of experimental data under varying conditions of water undersaturation to water excess at pressures as great as 8 MPa and durations of 1 h to several days (Quane and Russell, 2005—this issue). For example, at ~5 MPa, and 0.45 wt.% H2O, Boyd (1961) found no welding at 590 °C and some welding at 620 °C at run time of ~3.5 days (Fig. 6A); at ~18 days, the minimum welding temperature was between 550 and 590 °C, but no dense welding was achieved.

5.1.2. Water

The presence of water in vitric pyroclasts reduces their viscosity (e.g., Shaw, 1972; Dingwell et al., 1996) and so would enhance welding through viscous deformation. Sparks et al. (1999) argue that resorption of water from trapped interstitial gas in ignimbrites may be an important process for reducing viscosity and causing dense welding near vents and may also account for the occurrence of dense fiamme in less densely welded ashy matrix. Experimental data amply support the notion that addition of some water drastically reduces the temperature at which welding takes place. In this study, dense welding occurred at 0.4 wt.% H2O, for otherwise constant experimental conditions (Fig. 6). At water concentration of 0.1 and ≥0.8 wt.% welding was less (Fig. 6). The sharp increase in welding between 0.1 and 0.4 wt.% H2O coincides with a glass viscosity decreases from 10^{13.5} Pa s to 10^{11.5} Pa s (calculated for 600 °C, using the formulation of Hess and Dingwell, 1996). The glass transition, a kinetic boundary between viscous and elastic response to applied stress, occurs at about 10^{12} Pa s for a wide range of natural volcanic glasses (Giordano et al., 2005—this issue). We attribute the sharp onset of welding, including sintering and deformation of shards, to the rapid reduction in viscosity caused by a modest increase in water, causing the glass to deform viscously.

Interestingly, a few tenths wt.% H2O are typical values for fresh rhyolite glass and suggest that it is a common level to which rhyolite magma is degassed during eruption. Dingwell (1996) points out that rhyolite magma is likely to degas readily to 0.2 to 0.4 wt.% H2O through bubble exsolution during ascent, but that “viscosity quench” would be induced by the dramatic increase in viscosity with water loss, inhibiting further rapid water loss. We suggest that the water necessary to facilitate welding is commonly residual from degassing. Rapid accumulation of pyroclasts, particularly in and near vents may also trap water, that when resorbed, would further facilitate welding (Sparks et al., 1999).

There are two causes for the reversal in degree of welding in experiments with high water concentrations, first, the increased water pressure, and second, the increase in vapor-phase sublimates. In our experiments the gold capsule sealed in the water, so that the effective pressure on shard contacts decreased with increasing water pressure, with the effect of reducing sintering (adhesion by fusion) between glassy clasts. In a natural setting, however, the ignimbrite is likely to be highly permeable during much of its emplacement and welding history. Our experiments may simulate natural conditions where pervasive incipient sintering creates sealed pore space equivalent to the gas-retention regime (versus the compaction regime) of Sparks et al. (1999).

A second effect of increased water concentration in the experiments was the increased development of sublimates on the surfaces of the shards. By armoring the glass, these crystals are likely to inhibit absorption of water and so inhibit reduction of viscosity. Perhaps more importantly, the crystal-armored surfaces reduce glass–glass contacts and so inhibit sintering among glassy fragments (Tuffen et al., 2003). The occurrence of vapor-phase crystallization in the upper parts of ignimbrites provides a natural analog to the water-rich experiments. The presence of such sublimates likely accounts for the more gradual declines in density with stratigraphic height in ignimbrites compared with the sharp increases in density nearer the base (e.g., Riehle et al., 1995, fig. 14). In contrast, more gradual density profiles in upper sections have been calculated with models of cooling by rainfall (Riehle et al., 1995).
5.1.3. Pressure

Pressure, in the form of accumulating load during emplacement, is the main driving force for compaction and expulsion of interstitial gas (e.g., Smith, 1960b). Under otherwise comparable experimental conditions, increasing pressure increases the degree of welding. Under dry conditions, an increase from 1 At to 4.8 MPa decreased the minimum welding temperature from above 900 °C to between 650 and 690 °C for rhyolite ash (Boyd, 1961; Fig. 6A) (albeit neglecting the difference in run times, 16 versus 72 h, respectively). In our experiments, 1 MPa (0.2 wt.% H$_2$O) was insufficient to cause welding at 600 °C, but 5 MPa (0.4 wt.% H$_2$O) induced strong welding and 20 MPa (0.5 wt.% H$_2$O) caused complete welding (Fig. 6A). Increased pressure drives close packing of clasts, and so favors sintering and viscous deformation, provided the glass resides above the glass transition.

6. Conclusions

We conducted experiments on natural ash from the rhyolitic Rattlesnake Tuff and compared the results with naturally welded samples.

At 1 At, welding occurs by sintering, initiates between 900 and 1000 °C, and produces globular clasts. The degree of welding increases with temperature more than with time. The temperatures required for welding at 1 At are unreasonably high compared to emplacement temperature of most rhyolite ignimbrites; textures are not like in natural samples but may have analogs among agglutinates.

With the addition of pressure and water, welding is divided into two main stages: a mechanical compaction regime and a viscous deformation regime. Initial compaction results in densification by loss of pore space and most of the rotation of shards into a foliation. With progressive welding, the viscous regime becomes more important, and results in sintering and deformation of shards. Transition to the viscous regime is abrupt, which accounts for the typically sharp basal contacts between poorly and densely welded facies of the tuff. At 600 °C, the viscosity of metaluminous rhyolite decreases sharply to below ~10$^{12}$ Pa s with a few tenths wt.% H$_2$O, and so crosses the glass transition boundary from elastic to viscous response to applied stress (Giordano et al., 2005—this issue). A few tenths wt.% H$_2$O are likely to be common in pyroclastic rhyolite glass owing to viscosity quenching (Dingwell, 1996); while external sources or resorption of water will promote welding, they are not be required. Large amounts of water inhibit welding by creating surface sublimates that interfere with sintering and may exert fluid pressure counter to lithostatic load if sintering and vapor-phase sublimates seal permeability in the tuff.

Rock densities and textures close to a natural suite of progressive welding in the Rattlesnake Tuff were produced with experimental conditions of 600 °C, 0.4 wt.% H$_2$O, 5 MPa and ranging in time from hours to 2 weeks; dense welding occurred after days.

Acknowledgements

This work was initiated during Anita Grunder’s sabbatical leave at Clermont-Ferrand and was supported by a CNRS grant (PNRN: Programme National des Risques Naturels) to Tim Druitt. Didier Laporte supported the experimental and SEM work. Martin Streck provided the starting material and natural samples. Discussions with John Dilles, Jeff Lee, Andrew Meigs, Kelly Russell, Martin Streck, and Steve Quane were particularly helpful. Hugh Tuffen and Martin Streck provided thoughtful reviews. AG thanks Maurice and Madeleine Jafféux for lessons in sintering glass onto lava at 1 At in the form of “lave emaillé”.

References

学霸图书馆（www.xuebalib.com）是一个“整合众多图书馆数据库资源，提供一站式文献检索和下载服务”的24小时在线不限IP图书馆。

图书馆致力于便利、促进学习与科研，提供最强文献下载服务。