

SOME NOTES ON THE MELTING OF GRANITE

ROY W. GORANSON.

There has been considerable discussion concerning the melting interval of granite,¹ but up to the present time there seems to be no unanimity of opinion as to the actual values of these temperatures. Some of this uncertainty can probably be traced to the fact that there is no one clean-cut answer to the problem. For the term granite is used not for a definite composition but to classify a rather indefinite range of compositions. Variations in one or more of the components of granite, within this composition range may alter appreciably the value of the liquidus temperature.

There are also experimental difficulties with which to contend, and it is perhaps owing to the high viscosity of dry silicate melts with consequent difficulty of attaining equilibrium within the time available for a laboratory measurement that some writers have assumed liquidus temperatures for granite to lie above those of gabbros.

It may also be possible that the high melting temperatures of crystalline silica (quartz, tridymite, and cristobalite), have influenced opinions on melting temperatures, although geologic evidence points to the conclusion that granite represents a late stage in fractionation by crystallization from a cooling magma originally more femic.

There is still another consideration to be introduced in any attempt at evaluating the crystallization temperature interval of a granite magma. For all magmas contain volatile components, of which perhaps only a small part becomes bound up in the crystalline phases (granite), the greater portion of these volatiles playing a complex rôle in what are commonly called post-magmatic processes. Of these volatile constituents water is the most abundant, and in this paper is presented an account of some results obtained by melting Stone Mountain granite in the presence of water.

¹ Granite, like any other mixture of more than one component except a eutectic composition, does not melt or solidify at a point but over a temperature interval. The temperature at which the last crystals disappear or the first ones appear, T_L , is called the liquidus temperature; the temperature at which the first liquid appears or last liquid disappears, T_S , is called the solidus temperature. In the case of granite the temperature at which the last crystals formed is not a true solidus temperature but represents the point at which the residual liquid became separated from the crystalline phases. $T_L - T_S$ is the melting interval for the mixture.

PART I. EXPERIMENTAL RESULTS.

DESCRIPTION OF MATERIALS USED.

The granite used in these melting determinations came from Stone Mountain near Atlanta, Georgia. It is a normal biotite-bearing muscovite granite of light grey color. The granite glass, used in some experiments and listed in the experimental results as such, was obtained by melting this granite in a platinum-rhodium resistance furnace until the glass obtained was essentially homogeneous. The obsidian listed in the experimental results occurs at Cerro Noagua, New Mexico. The composition of the granite and obsidian are given below in columns 1 and 2 respectively. The analyses were made by Dr. E. S. Shepherd of this Laboratory.

	1.	2.
SiO ₂	73.39	76.35
Al ₂ O ₃	14.41	12.34
Fe ₂ O ₃	0.09	0.47
FeO	0.70	0.60
MgO	0.27	0.03
CaO	1.05	0.38
Na ₂ O	3.96	4.29
K ₂ O	5.07	4.39
H ₂ O+	0.39	0.91
H ₂ O-	0.05	0.00
TiO ₂	0.27	0.09
ZrO ₂	tr.	tr.
P ₂ O ₅	0.57	0.02
Cl	0.03	0.06
S	0.02	tr.
BaO	0.06	tr.
Sum	100.34	99.85
Density	2.66 at 20°C	2.33 at 25°C*

* The density of the normal obsidian is 2.3297 at 25° C, of the ignited obsidian is 2.3372 at 25° C as determined by a pycnometer method using a water thermostat.

EXPERIMENTAL DETAIL.

The granite, ground extremely fine (less than 200 mesh), and water were inserted in a platinum capsule which was then sealed. Sufficient hydrostatic pressure had to be put on these capsules to compensate for the water vapor pressure inside them. The method of loading and sealing the capsules, and description of the bomb used for obtaining the necessary pressure and temperature are given in an earlier paper.² When

² Goranson, Roy W., this Journal, 22, 481, 1931.

obsidian or granite glass was used the procedure was identical with that for granite.

The experimental results are presented in Tables A and B, the experiment number being listed at the left for reference. The temperature, pressure, and time of run are given in degrees Centigrade, bars, and hours respectively. Temperature was controlled by a regulator operated on the Wheatstone bridge principle using the furnace winding as one arm of the bridge. The regulator controlled the temperature within $\pm 5^\circ\text{C}$. Pressure was read on a Bourdon-type gauge which was calibrated by a dead-weight gauge of the Amagat type. The dial is calibrated in kilograms per sq. cm. and can be read to ± 10 kilograms per cm.², which is sufficiently accurate for the present purpose.

Runs longer than 10 hours were discontinuous, as it was not safe to leave the bomb unattended.

Results obtained on Stone Mountain granite are given in Table A, those obtained on Stone Mountain granite glass and Cerro Noagua obsidian are given in Table B.

TABLE A.

Experimental Results on Stone Mountain Granite.

T denotes the temperature and *P* the pressure during the run.

- A1-30 *T* = 1000° C; *P* = 950 bars; 1 hour run.
Granite + no water: no glass observed. The biotite is darkened.
Granite + 2.7% water: about 40% glass.
Granite + 15% water: about 80% glass. Crystalline grains are all rounded and corroded. Hematite plates have formed.
- A1-32 *T* = 900° C; *P* = 1500 bars; 3 hour run.
Granite + no water: a few small isotropic patches of index about 1.485. Biotite completely altered.
Granite + 4.8% water (water however leaked out sometime during the run); patches of glass of index about 1.498 enclosing branches and scales of hematite. The residual quartz grains are all rounded.
Granite + 3.8% water: all glass of index about 1.488. The glass is sprinkled abundantly with tiny bubbles.
- 2A-12 *T* = 888° C; *P* = 960 bars; 3 hour run.
Granite + 5.3% water: chiefly glass. A few rounded quartz grains are present. Small hexagonal plates of hematite have developed.
- 2A-8 *T* = 862° C; *P* = 590 bars; 4 hour run.
Granite + 8.8% water: Light grey cylinder of glass interspersed with crystalline grains. Index of glass about 1.488.
- 2A-12 *T* = 821° C; *P* = 960 bars; 3 hour run.
Granite + 5% water: about 30% glass. Quartz grains are all corroded and rounded. Hematite and magnetite were observed.
- 2A-18c *T* = 816° C; *P* = 960 bars; 21 hour run.
Granite + 2% water (lost all the water some time during the run): nearly all glass. Only a few rounded crystalline grains present.

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- 2A-12 $T = 778^{\circ} C$; $P = 960$ bars; 3 hour run.
Granite + 5% water: The crystalline grains are cemented with glass. Hematite has separated out.
- 2A-18a $T = 778^{\circ} C$; $P = 960$ bars; 4 hour run.
Granite + 50% water: about 85% glass. The crystalline grains present are all rounded.
- 2A-20 $T = 723^{\circ} C$; $P = 960$ bars; 33 hour run.
Granite + 17% water (lost all water some time during run): about 90% glass. Some recrystallization seems to have taken place in the glass, perhaps after the water had seeped out. The original crystals present are all badly corroded.
- 2A-20 $T = 723^{\circ} C$; $P = 960$ bars; 55 hour run.
Granite + 20% water: Some portions of the glass seem to be devoid of any crystalline material. Other portions have a few shreds of crystalline material together with development of hematite flakes. Probably less than 0.1% crystalline material.
- 2A-22 $T = 704^{\circ} C$; $P = 960$ bars; 47.5 hour run.
Granite + 9.5% water: 50% or more of glass. The crystalline material present consists of rounded feldspar and quartz together with recrystallized hematite. Tiny bubbles are sprinkled through the glass.
- 2A-23 $T = 704^{\circ} C$; $P = 960$ bars; 102 hour run.
Granite + 4.4% water: about 70% or more of glass. All but the larger quartz grains have disappeared. There is some recrystallization of hematite flakes and small needles of rather high index. There are also tiny bubbles sprinkled through the glass.
- 2A-26 $T = 704^{\circ} C$; $P = 960$ bars; 207.5 hour run.
Granite + 4.2% water: 75% or more of glass. The crystalline grains are chiefly highly corroded quartz. Recrystallization of hematite flakes and small needles of rather high index. Tiny bubbles are sprinkled throughout the glass.
- 2A-26 $T = 704^{\circ} C$; $P = 960$ bars; 262.5 hour run.
Granite + 7% water (lost all water some time after 55 hours): about 70% glass. The crystalline grains are chiefly corroded quartz. There is recrystallization of hematite and some colorless needles which perhaps took place after the water had escaped.
- 1B-24 $T = 600^{\circ} C$; $P = 385$ bars; 460 hour run.
Granite + water: a small amount of glass was observed. The biotite is completely altered to magnetite and chlorite; the oligoclase has been attacked and probably partially dissolved by the water. Needles similar to those described in the 460 hour run of Table B occur in void spaces.

TABLE B.

Experimental Results on Cerro Noagua Obsidian and Stone Mountain Granite Glass.

- 3A-28 $T = 750^{\circ} C$; $P = 980$ bars; $4\frac{3}{4}$ hour run.
Stone Mt. granite glass + 6.2% water: Recrystallization of hematite and of a few small, high index rods has taken place.
- 3A-30 $T = 900^{\circ} C$; $P = 490$ bars; 4 hour run.
Stone Mt. granite glass + 3.7% water: A few scattered rod-like crystals have formed. They are extremely small and probably make up only a very small fraction of a per cent of the whole amount.
- 2A-2 $T = 815^{\circ} C$; $P = 570$ bars; 4 hour run.
Obsidian + no water: sintered to a long rod. Some crystallization has taken place, the glass being liberally spattered with tiny birefracting grains.

- 2A-18b $T = 751^{\circ} \text{C}$; $P = 960$ bars; 6 hour run.
Obsidian + 30% water: a few tiny rods have crystallized out.
- 1B-24 $T = 600^{\circ} \text{C}$; $P = 385$ bars; 460 hour run.
Stone Mt. granite glass + 2.6% water: The powder sintered to a dark grey rod during the run. It appears to be a mass of birefracting grains too small to be clearly resolved and it is impossible to estimate the proportion of glass to crystals. Growing out from the glass are long needles or rods which in places form a radiating fibrous mass. These needles extinguish parallel to the elongation or close to it. Elongation is negative. Index 1.64 ± 0.01 . In places V-shaped ends were observed. These crystals probably grew from the water-rich solution which had leached the components of the crystals from the glass.
- 3A-18 $T = 600^{\circ} \text{C}$; $P = 980$ bars; 4 hour run.
Stone Mt. granite glass + 7% water: The glass is speckled with crystalline material. There are at least three kinds of crystals present: hematite, laths of plagioclase (?) and needles or rods of index higher than the laths and of negative elongation. In some places the laths and rods radiate from a central crystalline grain and resemble spherulitic growths. In places crystalline grains are arranged in parallel and concentric bands; these bands have a microfelsitic texture. This banding may be a kind of flow structure caused by kneading of the capsule due to pressure changes.

DISCUSSION OF RESULTS.

In the presence of water vapor at 960 bars pressure Stone Mountain granite became completely molten in 3 hours at 900°C ; in 55 hours it was more than 99% glass at 723°C with only a few corroded remnants of the larger quartz grains remaining. In 207 hours at 704°C there remained only 20 per cent or less of crystalline material; since the only original material left which could be identified was quartz, this estimate of crystals is probably high. After 4 hours at 600°C Stone Mountain granite glass with 6% or more of water in solution contained numerous tiny crystals. Two distinct species, besides hematite, were observed but could not be identified. One type consisted of plates with the same sign and general appearance as feldspar. The other type was of rods or needles of higher index than the laths and had a negative elongation. In a 460-hour run at 600°C and 385 bars pressure on Stone Mountain granite glass and water, the glass appeared to be essentially crystalline. In this preparation hexagonal outlines were observed which may be quartz, but in none of the other glasses was any material observed which might have been quartz. In order to determine whether quartz would crystallize readily a 3-hour run was made on silica glass plus water at 900°C and 1000 bars. On examination the material was found to have crystallized completely to quartz. Hence, if conditions are

suitable, quartz should undoubtedly crystallize from the granite melt.

A 460-hour run was made on granite plus water at 600° C and 385 bars pressure. After the run a small amount of glass, perhaps less than 1%, was observed cementing some of the grains. The biotite had been completely altered to magnetite and chlorite, and the oligoclase grains appeared to be somewhat leached by the water.

The presence of needle-like rods in the glass even up to 900° was rather puzzling. They have indices of about $1.64 \pm .01$ and negative elongation. Some of these rods had V-shaped ends and in places branched out in a radiating fibrous pattern. One of the runs was of sufficient duration (460 hours), to allow them to grow large enough to permit determination of the foregoing optical data. In this preparation the needles grew out from the glass into the void spaces. (This run differed from the other runs in that a special bomb was built such that the water generated all the pressure and hence allowed voids to remain intact in and around the glass.) The inference is that these needles must have crystallized from the water-rich solution which leached the constituents of this mineral from the glass. There are two non-crystalline phases present in the system; one consisting of water plus material leached from the granite or granite glass, the other consisting of granite glass plus water. The presence of considerable material soluble in the water-rich phase is apparent on evaporating off the excess water after a run, as there was always a white residue left. Whether the rods present in the other glasses had a similar origin is not known, because they were completely enclosed in the glass. They could, however, have grown from the surface of the glass grains and become enclosed on coalescence and flow of the glass.

CONCLUSIONS.

It is concluded that at $700 \pm 50^\circ$ C and under a water vapor pressure of 1000 bars Stone Mountain granite will become, except for hematite, completely liquid. The melt will have 6.5% of water in solution.³

Considerable crystalline material formed in granite glass subjected to a temperature of 600° and a water vapor pressure of about 385 bars for 460 hours. A small amount of

³ Goranson, Roy W., *op. cit.*

glass, probably less than 1%, was present in granite subjected to the same procedure. If equilibrium were attained in this time then at about $575 \pm 25^\circ$ and a pressure of 385 bars 99% or more of the silicate portion of the system granite-water will be crystalline.

In these experiments the iron present seemed to be almost entirely oxidized by the water vapor and, as it is very insoluble in the granite-water melt, much of it crystallized out as hematite at temperatures below 1200° . At 1200° the glass contained no hematite and was of a light straw color.

PART II. GENERAL DISCUSSION.

There are still insufficient data to give a coherent picture, and, to obtain it, hypotheses which are not fully substantiated must be introduced. Therefore the following discussion will be subject to change as more data become available.

Here the hypothesis to be introduced is an expression of the relation between concentration of water and liquidus temperature of granite.

The initial slope of the melting temperature lowering is given by the equation

$$\left(\frac{\partial \theta}{\partial m_1} \right)_P = \frac{1.9864\theta^2}{18\Delta x_1 m_1^2}$$

where θ denotes the absolute thermodynamic temperature, m_1 , the mass fraction of granite, and Δx_1 , the heat of solution of the last crystalline phase of granite to go into solution in gram calories per gram. This equation, if we assume the heat of fusion to be 100 calories per gram, gives us an initial lowering of 10.5° per 1% of water. It is not applicable, however, except for very low water concentrations. The rigorous general equation cannot be used through lack of data.

The results of Morey⁴ on the systems, $\text{H}_2\text{O}-\text{K}_2\text{SiO}_3$ and $\text{H}_2\text{O}-\text{K}_2\text{Si}_2\text{O}_5$, should give a closer approximation at the higher water concentrations. The initial slope for the system, $\text{H}_2\text{O}-\text{K}_2\text{SiO}_3$, is, according to the foregoing equation, about 10°C per 1% of water. Morey's observed temperature lowerings for the system, $\text{H}_2\text{O}-\text{K}_2\text{SiO}_3$, are: 35.8°C for a

⁴ Morey, G. W., *Jour. Am. Chem. Soc.*, **39**, 1207, 1917.

2% water solution, 376° C for a 6.3% water solution. The slope of the liquidus for this system thus steepens considerably with increasing water content. Observed temperature lowerings for the system, $\text{H}_2\text{O}-\text{K}_2\text{Si}_2\text{O}_5$, are: 8° for a 0.6% water solution, 441° for an 8.3% water solution.

From these values we can now calculate a liquidus temperature for the dry granite, i.e. the temperature at which it will be completely molten. The calculated value is $T_L = 1050 \pm 50^\circ \text{C}$.

The calculated liquidus temperatures in degrees Centigrade (and pressures in bars) of Stone Mountain granite are: 1035 (140); 1005 (260); 963 (400); 908 (550); 840 (730); 750 (930); and 600 (1100) for 1, 2, 3, 4, 5, 6 and 7 weight per cent of water respectively.

As will be observed from the above computation, the liquidus does not have a maximum pressure in the crystallization temperature range of a granite magma. Furthermore, the temperature-pressure-water solubility relations are such that an error of 100° in the calculated temperatures will not affect the validity of the foregoing statement.

The end temperature in the crystallization process of a granite will not be a solidus temperature but the temperature at which this normal process becomes interrupted by a separation of the residual solution from the coexisting crystallized phases of granite.

Wright and Larsen⁵ have evolved a set of criteria to determine whether quartz crystallized as the high or low temperature modification, and used it in a study of quartz from granites, dikes, pegmatites, and veins. These criteria have also been used by others for this purpose. The data indicate that quartz in granite, granite porphyry, and aplite dikes crystallized as the high temperature form, and that pegmatite quartz straddled the inversion, i.e. in the initial stages quartz crystallized as the high temperature form and in the final stages as the low temperature form. This would place the initial stage of pegmatite formation as about 700°, the final temperature as perhaps 550° C.

Let us assume we have a granite magma containing 1% of water in solution and at a depth of, say, 10 kilometers. It will begin to crystallize at about 1025° C. Crystallization, with corresponding increase in concentration of water in the residual liquid, will proceed with continued cooling. When the tem-

⁵ Wright, F. E., and Larsen, E. S., this Journal, 27, 421-47, 1909.

perature reaches 700° C about 85% of the original magma will have crystallized, the residual liquid containing about 6.5% of water in solution.

The residual solution, 15% of the original magma, would be available for the formation of aplites, pegmatites, and quartz veins; of this amount $\frac{2}{3}$ would crystallize between 700° and 500°. Of the original magma containing 1% water in solution we would have as final products: 85% of granite (1035°-700°), 10% of aplite and pegmatite dikes (700°-550°), and 5% of quartz veins (500° and lower).

If this granite magma with 1% of water in solution lies at a depth of 4 kilometers crystallization will begin, as before, at about 1025° and continue normally until the temperature drops to 950° when about $\frac{2}{3}$ of the original magma will have crystallized. At this point the pressure necessary to hold the increased concentration of water in solution will become equal to the hydrostatic head at this depth. But we know that the pressure-solubility curve is considerably steeper than the temperature-solubility curve at this pressure,⁶ hence further crystallization will be accompanied by an ebullition of water.

There are then two coexistent fluid phases—one predominating in silicates and the other in water. Water will continue to boil out of the magma as silicates crystallize. In a 460-hour run at 600° and a vapor pressure of 385 bars less than 1% of the granite had melted. If in this time an equilibrium was reached then there would be little opportunity here for the formation of aplite and pegmatite dikes.

If the original granite magma contained 3% of water in solution at 10 kilometers depth crystallization would begin at about 965° and continue quietly until a temperature of about 700° was reached; at this temperature about half of the system would be crystalline, the solution containing about 6.5% water in solution. The pressure necessary to hold this amount of water in solution at 700° will be equivalent to the hydrostatic head at this depth and thus further crystallization would be accompanied by an ebullition of water with formation of two coexistent fluid phases. This would continue until the temperature-pressure curve dropped below the external load pressure.

⁶ Goranson, Roy W., *op. cit.*

SUMMARY.

In any attempt at evaluating the fusion conditions of granite there is a multiplicity of variables to consider. For a granite may vary in composition within rather hazily defined limits. Furthermore, granite magmas also contain other constituents, such as volatile material, which are only partially bound up in the minerals of granite. Of these volatile constituents water is the most abundant. In this paper is presented an account of some results obtained by melting Stone Mountain granite in the presence of water.

Part I. Experimental Results.

The granite, ground fine, and water were sealed up in platinum capsules and brought to the required temperature and pressure in a bomb. Pressure was obtained from the expansion of carbon dioxide, and temperature by means of a platinum resistance furnace enclosed in the bomb.

Equilibrium was difficult to obtain at temperatures below 800°C : for example, 50 hours was required to fuse Stone Mountain granite at a temperature of 723°C and under a water vapor pressure of 960 bars.

It is concluded that at $700 \pm 50^{\circ}\text{C}$ and under a water vapor pressure of 980 bars Stone Mountain granite will become essentially completely liquid, the resulting magma containing 6.5% water in solution; that at $575 \pm 50^{\circ}\text{C}$ and under a water vapor pressure of 385 bars the granite will be 99% + crystalline.

Part II. General Discussion.

After assuming a certain relation between liquidus temperatures of granite and solubility of water the liquidus of dry Stone Mountain granite is calculated to be $1050 \pm 50^{\circ}$. The general course of crystallization of a granite magma containing water in solution is then discussed.