

Swanson (1977) and Fenn (1977, 1986): experimental studies of crystallization in the alkali feldspar and granite systems

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Introduction

Sam Swanson and Phil Fenn were Ph.D. students with William C. Luth at Stanford University when Luth, Dick Jahns, and Frank Tuttle were conducting their investigations of granites and pegmatites. Tuttle retired in 1971, but Jahns and Luth continued through the 1970s and beyond. Tuttle and Luth were engaged primarily in experimental studies of the granite system. Jahns' interests were in granitic pegmatites; his role in experimental investigations pertinent to pegmatites remains a matter of question (see essays #8 and #9 of this series).

Two papers, both landmarks in the field of crystallization in the granite system, were immediate outgrowths of the Ph.D. studies:

- Swanson, S.E. (1977) Relation of nucleation and crystal-growth to the development of granitic textures. *American Mineralogist*, **62**, 966-978.
- Fenn, P.M. (1977) The nucleation and growth of alkali feldspars from hydrous melts. *Canadian Mineralogist*, **15**, 135-161.

Other papers stemming from that same time period appeared later:

- Fenn, P.M. (1986) On the origin of graphic granite. *American Mineralogist*, **71**, 325-330.
- Swanson, S.E. and Fenn, P.M. (1986) Quartz crystallization in igneous rocks. *American Mineralogist*, **71**, 331-342.
- Swanson, S.E. and Fenn, P.M. (1992) The effect of F and Cl on the kinetics of albite crystallization: a model for granitic pegmatites? *Canadian Mineralogist*, **30**, 549-559.

Articles by Swanson (1977), Fenn (1977), and Fenn (1986) are the subject of this essay, and they are provided with this essay in PDF by permission of the Mineralogical Society of America and the Mineralogical Association of Canada. Because the articles are posted, this essay contains a minimum of quotation from the original texts and reproduces none of the figures. To appreciate the importance and novelty of their studies, the following section presents some of the context of petrologic experimentation throughout its history.

Background

Norman L. Bowen established the means and objectives of petrologic experiments that were used ever after. Early in his studies, Bowen (1913) observed that melts spanning the solid solution of the plagioclase feldspars failed to crystallize completely upon cooling. His approach to the determination of the liquidus and solidus temperatures of the plagioclase system, therefore, entailed the melting of wholly crystalline material. In the terms used by petrologists, these were

forward-direction, thermally prograde experiments from room temperature to the final run condition (Figure 1). They simulated the partial melting of crystalline rocks, but only to the extent that the use of finely milled powders melting in air simulate the natural environment. Bowen (1913) was able to partially crystallize some compositions. In so doing, he observed that the compositions of the feldspars that formed evolved from those more calcic than the starting material to or toward the composition of albite, as a succession of compositional zones deposited on the same crystal. The concept of fractional crystallization, for which Bowen is so widely recognized, began with these experiments. However, Bowen (1913) recognized that the liquidus and solidus boundaries that were established through prograde partial melting experiments could not be replicated by thermally retrograde, or reverse-direction, experiments in which an anhydrous feldspar melt was cooled below its solidus. He attributed the sluggishness of crystallization to the high viscosity of the melts, which hindered the ionic diffusion that was necessary to nucleate crystals. One year later, Bowen (1914) published a similar investigation of the liquidus relations in the system of diopside-forsterite-silica. Starting with chemical reagents, he stated that

“The phases found represent equilibrium at the furnace temperatures for inert bodies like the silicates...” (p. 208)

Later in that paper, Bowen (1914) utilized the liquidus diagram to explain the crystallization of mafic melts, such as would be represented by the composition of this system, as a process that occurred at equilibrium, following the liquidus surface as determined by his experiments. In doing so, he inferred that the process of crystallization – cooling of a melt, akin to the thermally retrograde, reverse-direction of experiments – would yield the same results as the prograde melting experiments, even though he knew that this was not true of the plagioclase series. His inference established that melting is a proxy for crystallization. I have suspected, though I have not found in his writings, that Bowen also must have inferred that the crystallization of viscous granitic melts would follow the liquidus surface if given adequate time that was greater than his experimental durations. From this point on, Bowen’s methods and assumptions became the foundation of almost all subsequent liquidus experimentation by petrologists. Throughout petrologic study, the solidus of the granite system (e.g., Tuttle and Bowen, 1958; Luth et al., 1964) has been regarded as the lower pressure-temperature limit at which a granitic melt can exist. In contrast, experiments in the solid state (plus aqueous or carbonic fluids) that have been conducted by metamorphic petrologists have carried the expectation of demonstrated reversibility of results – replications by thermally prograde and retrograde experiments. The same holds for experimental calibrations of trace-elements and isotopic exchange among phases.

The attainment of chemical equilibrium in a rock system of interest became the holy grail of experimental petrology. Such experimental results, if achieved, would define the limits of stability for various mineral assemblages based on the chemical energy that they possessed at different states. The acceptance of such experiments as equilibrium results made them independent of the means by which they were achieved, as if those means were irrelevant to the final result.

The first experiments (of which I am aware) that purposefully explored the crystallization of silicate minerals from undercooled melts (that is, compositions that were completely melted, then

cooled rapidly below their liquidus temperature) were performed by Gary Lofgren (1974). Lofgren (1974) documented the changes in plagioclase crystal habits (the shapes that crystals adopt by virtue of the environment of their crystallization) at temperatures below the liquidus temperature for the compositions chosen. Synthetic mixtures were heated above the liquidus temperature of the system (as determined by forward experiments with crystalline compositions) to melt them completely as a preconditioning step, then cooled quickly at constant pressure to a temperature below the liquidus (Figure 1). The magnitude of that undercooling, which is referred to as the liquidus undercooling and abbreviated as ΔT , determines the driving force to nucleate and grow crystals. The energetic driving force to grow crystals increases with the magnitude of undercooling, but that does not necessarily mean that crystallization occurs more rapidly or extensively with increased undercooling. That is because of the kinetic aspects of the process.

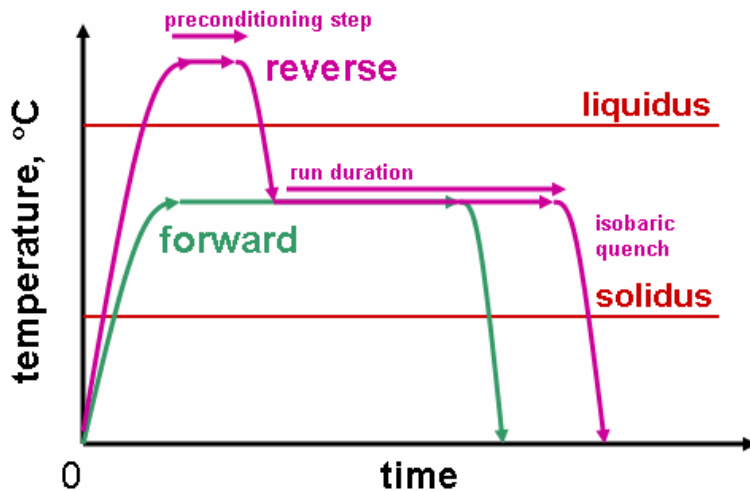


Figure 1. A schematic illustration of experimental methods. Forward: single step to run temperature followed by quench; reverse: pre-conditioning step above the liquidus, followed by cooling to run temperature, and final quench.

Lofgren (1974) showed a progressive evolution of plagioclase crystal habits and compositional zonation that could be correlated with plagioclase crystallization in different geologic environments. These were among the first experiments that sought to correlate the textures of experimentally-grown crystals with the different temperatures of their environment of formation. Using this method, the actual temperatures of crystallization could be estimated from texture alone. Lofgren (1974) used the spherulitic textures of crystals in rhyolite obsidians to illustrate the merits of a textural analysis when crystallization has clearly occurred at non-equilibrium conditions, i.e., well below the liquidus temperature of the system. However, equilibrium liquidus diagrams remain the frame of reference for igneous petrologists, especially in relation to intrusive igneous bodies like granites and pegmatites.

Lofgren's (1974) study was titled "Isothermal Crystallization" because the undercooling step was completed before crystals began to nucleate and grow from the undercooled melts. The crystallization, therefore, took place at one temperature, or isothermal condition. Lofgren (1980) went on to study plagioclase and olivine crystallization with progressive steps of cooling, what is sometimes referred to as programmed cooling, but which Lofgren (1980) branded as dynamic cooling experiments. The use of the term "dynamic" conveyed the true nature of crystallization: a response to change, which arises from cooling in most cases.

In an email to Sam Swanson on March 2, 2015, I asked him whose idea it was to start the series of undercooling experiments that he and Fenn reported. His reply on the same day was:

“The undercooling idea came to us from Bill [Luth] via Gary Lofgren. Bill and Gary were at a meeting where Gary was showing some of his results of undercooling to try and replicate volcanic processes (lunar and otherwise). Bill was taken with the idea -- so much so that he called us (he could not wait a few days to return to Stanford) and said to stop what we were doing and to try undercooling experiments in each of our systems. Some worked, some did not, the rest is history.”

Swanson, S.E. (1977) Relation of nucleation and crystal-growth to the development of granitic textures

Methods. Swanson (1977) employed two synthetic silicate gels as starting materials that consisted of the components of anorthite (An, $\text{CaAl}_2\text{Si}_2\text{O}_8$), albite (Ab, $\text{NaAlSi}_3\text{O}_8$), orthoclase (Or, KAlSi_3O_8), and quartz (Qtz, SiO_2). Though meant to represent the compositions of granite and of granodiorite, the ones synthesized by Swanson (granite: $\text{An}_{7.5}\text{Ab}_{32.0}\text{Or}_{34.0}\text{Qtz}_{26.5}$ and granodiorite: $\text{An}_{19.8}\text{Ab}_{37.3}\text{Or}_{19.8}\text{Qtz}_{23.1}$) were far from the compositions of granites, which plot close to the eutectic or minimum composition of $\text{Ab}_{38}\text{Or}_{28}\text{Qtz}_{34}$ (Tuttle and Bowen, 1958) with the addition of a small anorthite component. Swanson (1977) chose those compositions to make his results comparable to Whitney’s (1975) determination of liquidus relations involving those same bulk compositions. The presence or absence of calcium in the system determines the sequence of feldspar crystallization in undercooled experiments (London and Morgan, 2017), but not necessarily the textures.

Following Lofgren’s (1974) methods, the bulk compositions were sealed in capsules with and without added water, pressurized, and taken to temperature at 8,000 bars for sufficient in time (“96 hours above the liquidus at 1000 °C”) to ensure that the contents were completely melted and homogenized (preconditioning step of the magenta path in Figure 1). Runs were then dropped to the final run temperature for a period of time, and subsequently quenched to room temperature within 20 minutes.

Finely powdered silicate materials adsorb water when in contact with air. This adsorption occurs rapidly, even in the minutes that a vial is open to air during the loading process. In temperate climates, such powders normally adsorb 1-4 wt% of their weight in H_2O through normal handling, and they retain adsorbed water when stored sealed under desiccation (London et al., 2012). To my knowledge, no experimentalists have factored that adsorbed H_2O into the total budget of H_2O in their experiments. Consequently, many experimental products that employ micronized powders are hydrous, and may be near saturation in H_2O at the conditions of experiments, even those that are presented as no water added or H_2O -undersaturated or fluid-absent. Milled silicate gels such as those employed by Swanson (1977) are more reactive than crystalline materials, and they should adsorb even more atmospheric H_2O . So it is curious that Swanson (1977) noted that the granitic composition failed to nucleate crystals for any combination of time and undercooling temperature with more than 3.5 wt% H_2O added to the charge (p. 969); the actual H_2O content might have been significantly higher. It is also notable that London et al. (1989) reported that the milled powder of the anhydrous Macusani obsidian

crystallized readily without added H₂O, but that no crystallization occurred with more than 4.5 wt% H₂O added. That powder, too, was likely hydrated by adsorption to a few percent of H₂O. Comparing the result of Swanson (1977) to London et al. (1989), there is similarity that may indicate a cause and effect between the amount of H₂O and the nucleation of crystals in the melt when powdered starting materials are utilized. In essay #9 of this series, however, I emphasized the importance of experimental methods on results and the vagaries of choices that may introduce what I called “anthropogenic effects.” In that essay, I noted that a powder of the Macusani obsidian crystallized readily at low H₂O but ceased to crystallize as the H₂O content approached saturation (London et al., 1989), much as Swanson (1977) observed. Solid cores of the same obsidian, however, failed to crystallize at low H₂O content, but the degree of crystallinity increased with increasing H₂O (London and Morgan, 2017), all else the same between powder and solid core experiments. So yes, the similarity of behavior described by Swanson (1977) and London et al. (1989) might arise from a replicable property that results from hydrous powdered samples loaded with a lot of interstitial air, but those results are not necessarily relevant to geologic environments, nor are they an intrinsic property of the melts so formed. Such are the pitfalls of what I have called simulation experiments, and it is precisely these process-dependent results that the equilibrium studies sought to avoid. As simulations of a natural process, the experiments ought to conform as closely as possible to a natural state of igneous rock formation. At the face of it, it seems obvious that igneous rocks do not arise from powdered glasses melted with entrapped bubbles of air (see London et al., 2012).

Swanson (1977) explained some of the inherent difficulties in analyzing and representing the data. With regard to the nucleation density – the number of crystals per unit volume of charge – the samples first had to be mounted and polished to produce thin transparent slices. The preparation of thin sections (p. 969) to view the samples necessarily grinds away the surface portion of the charges, and surfaces are where most nucleation begins. Experiments with powdered starting materials always emerge as very thin, flat planar slugs of glass; mounting and grinding those also means that crystals that nucleated on the walls of the charge, which were excluded from the analysis of Swanson’s data, appear to lie entirely within the melt (Figure 2). Swanson (1977) pointed out that in experiments that yielded few crystals, one charge might contain some while an identical charge in the same run contained none. His descriptions of the vagaries of the study elucidate the difficulty of making useful sense of the results. Anyone who had conducted similar experiments has seen and dealt with similar issues. However, as Swanson (1977) noted, when all of the data are considered, patterns emerge that, as he stated, are generally accurate in form but not necessarily so in magnitude in detail.

Swanson (1977) made brief mention of the nucleation lag time (p. 970), which is the interval of time between the start of the undercooled stage of the experiment and the first appearance of crystals in the charge. In fact, the nucleation lag time can only be determined at the end of an experiment as the time from undercooling to final quench. If crystals formed, they did so somewhere within that interval. In order to assess the nucleation lag time, also known as nucleation delay, it is necessary to make time an independent variable of the experiment, meaning to do replicate experiments as a function of time. Swanson (1977) did not do that, but a few others have (e.g., Evensen, 2001; Maneta and Baker, 2014; Sirbescu et al. 2017; London and Morgan, 2017). Those results of nucleation delay, plotted as undercooling versus time, possess a parabolic shape (Figure 3). The parabolic shape of Figure 3 defines two regions where crystals

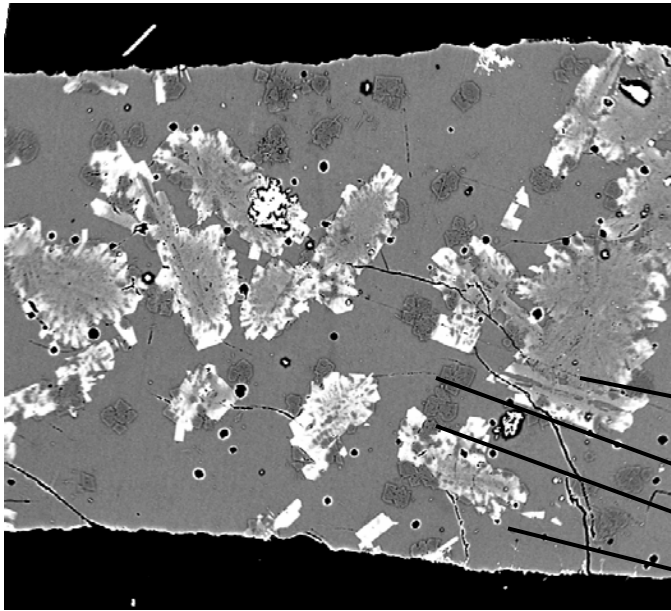


Figure 2. A backscattered electron image of experiment MAC151 with the powdered Macusani obsidian. Some crystals and crystal clusters have nucleated along the margins of the charge. Others appear to lie entirely within glass, but may have been rooted on a surface that was removed or lies below the plane of the image.

plagioclase-quartz graphic intergrowth
 K-feldspar
 skeletal quartz
 glass

might be expected to form but do not. With a low degree of undercooling below the liquidus temperature (the temperature at which crystallization should commence upon cooling if an equilibrium state is attained), no crystallization appeared within $\sim 50^\circ\text{C}$ of the liquidus temperature. This crystal-free domain is not determined so much by kinetic factors but rather by energetic constraints.

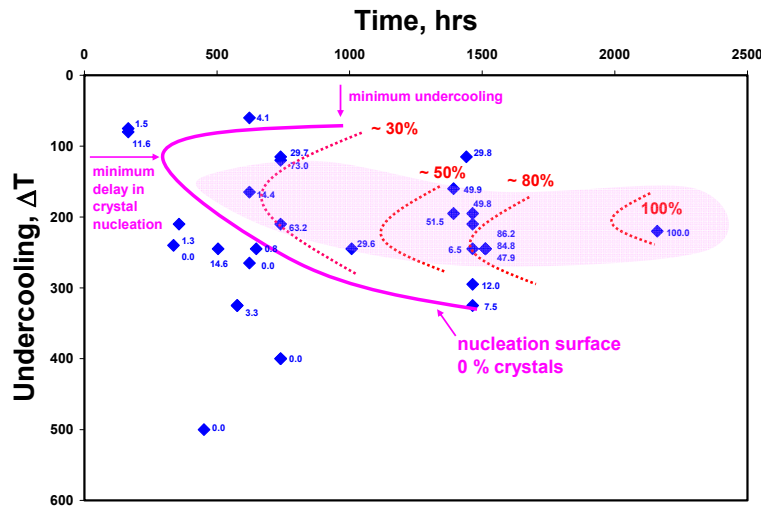


Figure 3. A plot of percentage of crystallization as a function of time and liquidus undercooling for the hydrous Macusani obsidian at 200 MPa. From London and Morgan (2017). The nucleation surface, which marks the first appearance of crystals, is denoted by the solid magenta curve. Red dashed curves give the approximate abundance of crystals in volume percent of the charge.

The equilibrium of the liquidus is defined in terms of the bulk properties of the melt and of crystals that form from it. However, the first-formed crystal nuclei possess a significant component of surface energy (unsatisfied electrical charge), such that the energy of forming these atomic-scale nuclei is greater than the energy of formation for the bulk of a crystal. Therefore, a greater driving force, derived from liquidus undercooling, is necessary to initiate the nucleation of stable crystals (consider that water does not freeze upon cooling to 0°C ; some undercooling of water is necessary to start and sustain the process to make ice in your freezer). In the granite system (Evensen, 2001; also Figure 14-22 of London, 2008), the “minimum undercooling” needed to initiate crystallization is closer to $\sim 100^\circ\text{C}$, but that result was based on

powdered starting materials. Hydrated solid glass cores of the same composition failed to nucleate at any time-temperature conditions.

The crystal-free region below the magenta curve in Figure 3 resulted from a kinetic limitation to the formation of stable crystal nuclei. The driving force to precipitate crystals was high, but the diffusion that is necessary to form and sustain the growth of stable crystal nuclei was too low. Low diffusivity results from a high viscosity of the melt at low temperature, and at the conditions below the magenta curve in Figure 3, the melt may have transformed to glass. The hydrous solid-core experiments of London and Morgan (2017) retained their original shape at least to up temperatures of 550°C (see Figure 4 in essay #9 of this series). At temperatures 100°-150°C higher, the ovoid shapes of the cores indicated that they had melted and flowed. In the same region of 500°-550°C, London and Morgan (2017) observed that thin veils of crystals followed the traces of curvilinear cracks in the glass cores. Both results indicate a high viscosity even for this hydrous and flux-rich melt at temperatures close to those of pegmatite crystallization (e.g., London et al. 2020).

Nucleation Density. Swanson's (1977) Figure 2 (p. 970) presented nucleation density versus the magnitude of undercooling, ΔT , for granite with 3.5 wt% added H₂O and for granodiorite with 6.5 and 12 wt% H₂O. The nucleation density of plagioclase, alkali feldspar, and quartz generally increased with increased undercooling, as would be expected from crystallization theory. In the granite system, plagioclase nucleated first with as little as 50°C of undercooling. The appearance of quartz and K-feldspar required more than 300°C of undercooling. The granodiorite system with 12 wt% H₂O presented similar results. Nucleation of all three minerals occurred at approximately the same undercooling of ~ 150°-200°C in granodiorite with 6.5 wt% H₂O. Nucleation densities reached maxima at $\Delta T \approx 200^\circ\text{-}300^\circ\text{C}$.

Crystal Growth Rates. Swanson (1977) recorded the rates of crystal growth (Figure 3, p. 971) from the maximum linear dimension of the largest crystal that appeared to have grown entirely within the melt, not in contact with its margins. That rate was determined by the time from the attainment of the undercooled temperature to the final quench of the experiment. The rates are given in centimeters/seconds, though the more common units are meters/seconds in contemporary studies. The maxima in growth rates for the granite system correlate with maxima in nucleation density at $\Delta T \approx 200^\circ\text{-}300^\circ\text{C}$. The granodiorite system showed a marked shift in the temperature of maximum growth rate from $\Delta T \approx 200^\circ$ with low water content to $\Delta T \approx 350^\circ\text{-}400^\circ\text{C}$ with higher water content. This can be interpreted to mean that an increased water content of the melt suppressed the nucleation of crystals rather than aiding in their formation closer to the liquidus temperature. Swanson (1977) observed two notable contrasts with the crystal growth data. One, the growth rates decreased sharply past their maxima with increasing undercooling. Two, the growth rates of all minerals in the granodiorite system decreased with increasing water content of the melt.

Textural Attributes. Swanson (1977) described a general trend of textural evolution from radial clusters of coarse or skeletal crystals at lower undercooling to radial fine-grained spherulites at greater undercooling, and eventually minute fibrous single crystals. He related this tendency to form finer, needle-like crystals to the likely decrease in ionic diffusivity in the melt due to its increasing viscosity at low temperature.

Geologic Applications. As this was among the first such studies of crystal growth in granite systems, Swanson's (1977) results had several important implications to the textures of granitic rocks. Foremost among them was Swanson's (1977) remark:

"Observation of growth of reasonably large crystals (several millimeters) in a short period of time geologically (several days) is one of the most significant results of this study." (p. 974).

It is important to remember that the measured growth rates are minimum values. As noted above, the duration in time of crystal growth was unknown from the experiments, but necessarily less than or equal to the duration of the undercooled portion of the experiment. A second reason that these are minima in growth rate stems from the nature of these experiments, in which the crystallization of the melt brought it closer to the liquidus temperature. As a result, liquidus undercooling decreased as crystallization proceeded. This relationship applied to the studies by Fenn (1977) as well, and it is discussed more fully below. Figures 2 and 3 of Swanson (1977), therefore, plot the initial liquidus undercooling prior to the onset of crystallization, not the final state of the system.

Swanson (1977) related crystal dimensions to the crystal nucleation densities of the experiments in the way that geologists have long understood it:

"... crystallization at small undercoolings produces relatively few nucleation sites with large crystals, as is characteristic of plutonic rocks, whereas large undercoolings produce a large number of nucleation sites with small crystals, as found in volcanic rocks." (p. 974-975)

Swanson combined the data for growth rate and nucleation density in Figure 5. Based on the experimental results, he observed that plagioclase ought form large phenocrysts (a population of crystals much larger in size than the remainder of the mineral assemblage) in granites that would cool slowly and crystallize in a region of small liquidus undercooling. However, Swanson (1977) noted that K-feldspar, not plagioclase or quartz, is the phenocrystic phase in granites. Swanson (1977) observed that overall, the growth rate for K-feldspar crystals was higher at its maximum than for plagioclase or quartz.

Plagioclase nucleated closest to the liquidus temperature in all three compositions, but especially in the granite and the most hydrous granodiorite compositions (Figure 2 of Swanson, 1977). London and Morgan (2017) explained why plagioclase would be expected to nucleate first upon cooling. In the undercooled state, the first minerals to crystallize are those that are farthest from their liquidus temperature; they liberate the most heat of crystallization because they are the most supersaturated components of the melt. In the case of the feldspars at a highly undercooled state, anorthite liberates by far the most heat energy upon crystallization compared to pure albite or K-feldspar. Consequently, the response of the undercooled melt is to crystallize the most calcic feldspar that can form, and that driving force is what causes plagioclase to nucleate first. As noted above, in calcium-free systems, K-feldspar is the first feldspar to precipitate for the same reasons of the amount of energy, as the heat of crystallization, that is liberated.

Applications to Pegmatites. Swanson (1977) referred to experiments by Jahns and Burnham:

“Jahns and Burnham (1958) grew crystals from a hydrous silicate liquid formed by melting a sample of natural pegmatite, as pictured in Wyllie (1963).” (p.967)

As described by Swanson (1977), this would appear to have been a forward-direction melting experiment, not a reverse-direction undercooling experiment (Figure 1). In response to my question to him regarding what he knew of the Jahns-Burnham experiments, Swanson replied in an email of March 2, 2015:

“Dick always eluded [sic] to them, but never showed us any results -- we often wondered if the experiments were all ‘leakers’ with all the water they used.”

Swanson (1977) identified Jahns and Burnham with the experiment shown by Wyllie (1963), and he alluded to experiments by Jahns and Burnham in his email, but he never saw any. I add my opinion that this seems strange: if I were having a face-to-face conversation about my experiments with a Ph.D. student who was pursuing similar goals, we would be looking at my experiments on an optical or electron microscope. No description conveys all the facets of such complex results.

Swanson (1977) made one other observation in relation the model of Jahns and Burnham (1969):

“Several generalizations can be made with regard to the growth rates measured in this study. As the amount of undercooling increases, the growth rate for an individual phase increases to a maximum and then decreases (Fig. 3). This is the expected behavior of growth rate measured in a viscous liquid (Tammann, 1925). With a given anhydrous bulk composition, the maximum growth rate decreases as the amount of water in the system increases (Fig. 3). Fenn (1976) also observed a decrease in the maximum growth rate with increasing water content, and this relation is as yet unexplained. Addition of water to a silicate system lowers the viscosity of the silicate liquid (Friedman et al. ,1963; Shaw, 1963) .A greater mobility of components and hence a faster crystal growth rate is expected as a result of lowered viscosity. Decrease in crystal-growth rate with increasing water content is not compatible with the petrogenetic theory equating the growth of large crystals with the evolution of a vapor phase (Jahns and Burnham, 1958).” (p. 972)

Crystal growth from an increasingly hydrous and less viscous melt cannot be equated to crystal growth from an aqueous solution, but the principle in question is the same: decreased viscosity of the growth solution, melt or aqueous fluid, should enhance the rates of ionic diffusion that promote crystal growth. If the opposite trends were observed, then the implication is that the kinetic factors of material transport are not the only ones, or even the principal ones, that determine the rates of crystal growth from the melt. That other factor would be chemical in nature, related to the effect of the dissociation of water in the melt and its consequent effect on the activities (chemical reactivity) of the crystal-forming components of the melt – namely, alkalis, aluminum, and silicon. The fact that the addition of H₂O at pressure greatly reduces the melting temperature of anhydrous granite demonstrates that H₂O destabilizes the crystalline

phases in favor of melt. This is a chemical interaction that might explain the observations of Swanson (1977) and Fenn (1977).

Fenn, P.M. (1977) The nucleation and growth of alkali feldspars from hydrous melts.

Fenn's (1977) article began with a treatise on what should have been familiar to petrologists all along, if they had ever thought about it. For example, Fenn (1977) highlighted some of the limitations that petrologists face in deciphering the history of an igneous rock based only on its final mineralogy and texture. He noted the limitations of the chemical approach through equilibrium relationships and thermodynamic calculations to understand textures:

“In fact, the thermodynamic stability of a mineral is a necessary but not sufficient condition to ensure its presence in the final product. As an example, phase-equilibrium data alone cannot effectively distinguish among a granite, an aplite, a pegmatite, or a rhyolite of similar bulk compositions. Much genetic information is contained in the kinetics and mechanisms of igneous processes, information which cannot be derived from phase-equilibrium studies. As long as textures are used in the interpretation of igneous petrogenesis, fundamental information on the processes involved in textural development is needed. Unfortunately, such information is not currently available for geologically significant systems.” (p. 136)

“Much of the problem lies in the reliance of the experimental petrologist on the attainment and demonstration of thermochemical equilibrium. Such an equilibrium state is, by its very definition, static, whereas the processes and reactions which are being modelled [sic] are dynamic. Phase-equilibrium data are extremely valuable in that they provide for the prediction of the initial and final assemblages of a reaction or process as well as providing a means for bracketing the location of such a reaction or process in pressure-temperature space. The equilibrium data, however, provide little or no information about the mechanism, path, extent, or rate of the reaction or process. The two most basic, and most important, igneous processes are the nucleation of crystals and their subsequent growth from the melt.” (p. 136-137)

Methods. Fenn (1977) synthesized his starting materials in the same fashion as Swanson (1977), *“using a coprecipitated gel technique.”* The initial material is a hydrous nitrate silicate gel with the texture of cottage cheese. That gel is “fired” (not mentioned by Fenn, 1977, or Swanson, 1977) over a burner to drive off water and the nitrate as nitrogen dioxide (a brown, highly toxic gas). The crystalline state of the fired gel is not mentioned, and likely has not been studied by those who make them. When we made such gels in our lab, they were subjected to x-ray powder diffraction to identify any newly synthesized crystals, for example of cristobalite or corundum. None were found, our gel preparations were completely amorphous.

“Two distinct experimental modes were employed. First, what will be referred to as the direct-synthesis approach [forward direction, Figure 1] was used to determine the equilibrium phase relations. In this method a set of capsules is taken from ambient conditions directly to the pressure and temperature conditions of the synthesis experiment, held for a specified period of time (long enough to achieve “equilibrium”) and then quenched. Alkali feldspars formed using this approach crystallized from a finely divided gel and typically have an average grain size of 20 μm .

A second set of experiments using a different technique was performed to study the nucleation and growth phenomena. These experiments consisted of a homogenization period of 96 hours at 1000°C and 2.5 kbar [preconditioning step, Figure 1], followed by a rapid, isobaric quench to a lower temperature at which nucleation and subsequent crystal growth occurred. In this method the alkali feldspars grew directly from an undercooled melt, a process much more akin to natural igneous processes than the synthesis from a gel as described previously.” (p. 140)

The first set of experiments, which were meant to establish the equilibrium relationships between crystals and melt as functions of composition and temperature, became the reference by which the extent of liquidus undercooling, the initial undercooling temperature of the second set of experiments, was established. Once the gel preparation methods became widely used, these were mostly employed in the place of natural minerals because the compositions could be closely constrained. The downside is that the direct synthesis of crystals from amorphous starting materials does not necessarily follow an equilibrium process, at least in part because there are no crystals initially in the starting material. This means that all crystals that form must nucleate, and kinetic barriers to nucleation are among the key subjects of these papers. In my lab, when we ascertained the liquidus relations of the hydrous granite system and of the Macusani obsidian, we subjected powders to a lengthy devitrification step, usually at 500°C at pressure, prior to running them forward. Control experiments established the run time needed to completely crystallize the starting granitic gel or obsidian. The exceedingly fine-grained assemblage of feldspars, mica, and quartz ensured that these minerals would be present up to their liquidus temperature, such that the nucleation of new crystals (and the problems of metastability associated with that process) would not be a factor.

Nucleation Delay. Fenn (1977) devoted considerable attention (p. 142-143) to the time interval between the initial cooling step and the onset of crystallization, which he referred to as the “*nucleation incubation period*”, also the nucleation delay or lag time. Fenn (1977) explained that the nucleation delay is the time interval over which the atomic structure of the melt changes from that of the preconditioning step at high temperature to the quasi-equilibrium state of the lower run temperature. Those who study the properties of melts more than crystals refer to this adjustment as the melt relaxation time. The relaxation time is a function of the viscosity of the melt at the new lower temperature. The more viscous the melt, the longer the melt relaxation time, and consequently the longer the delay of crystal nucleation. Fenn (1977) observed that the nucleation delay was longest for experiments at small and at large undercooling temperatures. Experiments in these regions failed to nucleate crystals in run durations of 72 hours. The same relationship of undercooling to nucleation delay is shown graphically in Figure 3 above.

Nucleation Density and Growth Rates. Figure 9 of Fenn (1977) plotted nucleation density and growth rates together for two compositions, Or₁₀ and Or₅₀ (the remainder is albite component), plus 2.7-2.8 wt% H₂O added, versus the magnitude of liquidus undercooling, ΔT . The two compositions yielded qualitatively similar results. Nucleation density remained exceedingly low from the liquidus temperature to undercooling of $\sim 300^\circ\text{C}$, but rose sharply with greater undercooling. The growth rate data showed well defined maxima at undercooling of 200°C , but fell off sharply with greater or less undercooling. The coincidence of high growth rate and low nucleation density led to a few large crystals of these compositions. The textures of the feldspars evolved from isolated tabular crystals to fine spherulitic clusters with increased undercooling.

In Figures 12 through 14, Fenn (1977) displayed hand-fitted curves for nucleation densities and growth rates for three compositions, Or₁₀, Or₃₀, and Or₅₀, against liquidus undercooling for three different amounts of added water from 1.7 to 9.5 wt% H₂O. For each feldspar composition, at least one set of experiments was saturated or oversaturated in H₂O at the pressure of the experiments (250 mega-pascals, or 2500 bars). The maximum growth rate changed little for the H₂O-undersaturated experiments, but shifted sharply closer to the liquidus temperature by ~ 100-150°C in each of the H₂O-saturated runs. Fenn (1977) observed that the maxima in growth rate in the H₂O-oversaturated samples were consistently lower than in the H₂O-undersaturated ones. In the same three figures, Fenn (1977) noted that the nucleation density changed linearly and dramatically to lower numbers of crystals as the H₂O content increased. The nucleation density curves defined sharp maxima that tailed off to near zero at greater or lesser undercoolings, and the maxima in crystal nucleation shifted from $\Delta T \approx 400^\circ\text{C}$ to $\Delta T \approx 100^\circ\text{-}150^\circ\text{C}$ as the H₂O content increased. For the H₂O-saturated experiments, the maxima in growth rate and in nucleation density coincided in the same range of undercooling. Compared to the H₂O-undersaturated compositions, those results reflected more and finer-grained crystals in the H₂O-saturated experiments in the undercooled regions of maximum growth rates.

Feldspar Compositions. The exceeding thinness of the feldspar crystals, many with included domains of glass, rendered microprobe analyses unreliable. Fenn (1977), therefore, relied on x-ray diffraction patterns to calculate the compositions of the feldspars formed in his experiments. He observed that the compositions of the feldspars that crystallized in forward-direction synthesis experiments were uniformly homogeneous and consistent with the compositions expected at an equilibrium state between crystals and melt. Crystals grown from undercooled reverse-direction experiments, however, were zoned, with domains that reflected what would be construed to be a history of decreasing temperature of growth. Fenn (1977) stated:

“Thus it appears that an alkali feldspar liquid which is quenched rapidly into the subsolidus region will initially crystallize a metastable feldspar-liquid pair, and only upon complete crystallization and homogenization will the equilibrium assemblage be established. A similar effect has been noted in the subsolidus crystallization of alkali feldspars from glasses and gels (Parsons 1969).” (p. 154)

Fenn (1977) later noted:

“...it is interesting to note that even after 240 hours the crystal-growth samples have not reached their "equilibrium" positions; this is especially true for the potassic limb.”(p. 154)

Parsons (1969) was the first to observe that the feldspars tend to record what appears to be a record of crystallization with falling temperature along the liquidus surface of the system, even though the feldspars were actually crystallized at a single subsolidus and subsolvus temperature. This behavior has been thoroughly documented in experiments (London and Morgan, 2017). The explanation offered by London and Morgan (2017) has been mentioned above: the first-formed feldspars are those that release the most heat energy by their crystallization. These would be the feldspar compositions that are farthest from their equilibrium on the liquidus, and thus the highest-temperature feldspars that the composition can produce. Those are the compositional

trends observed by Parsons (1969), Fenn (1977), and London and Morgan (2017). The principal applies to all minerals, which is why the border zones of pegmatites contain a diverse assemblage of the mafic minerals, all of which would have high liquidus temperatures.

In the undercooled experiments of London and Morgan (2017), the feldspar solvus pairs initially recorded a fictive temperature that was significantly higher than that of the experiment; successive pairs approached the actual run temperature, but rarely attained it (Figure 4). The record of high fictive temperatures in the first-formed feldspars was unexpected, because that means that those feldspar pairs regressed to a common temperature among all components with low uncertainty of error, even though it was not close to the actual temperature of their growth. That relationship gives the appearance of an equilibrium state when it is actually far from the equilibrium state. The last-formed pairs came close to the temperature of the experiment, but they were always higher by 10°-25°C. This was true even for experiments conducted as forward-direction syntheses: the calculated temperatures of the feldspars were higher than the actual run temperature. For the feldspars, a close approach to equilibrium is close enough.

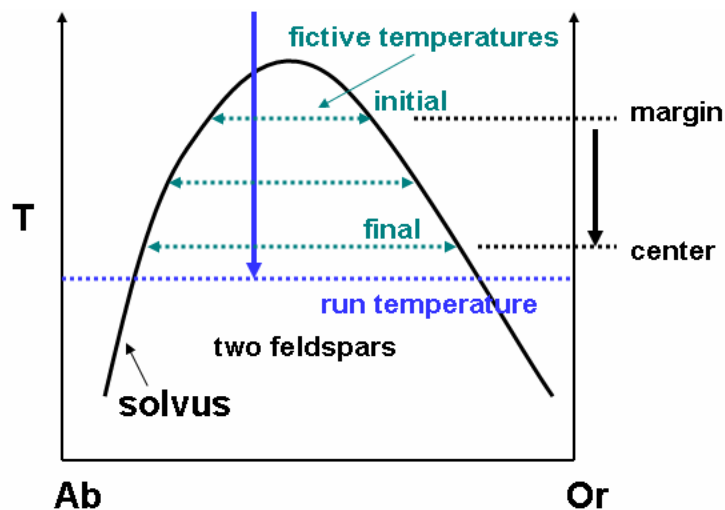


Figure 4. A schematic depiction of the temperatures recorded by feldspar pairs in the experiments of London and Morgan (2017). The blue arrow represents the composition and undercooling step of the bulk composition. Dashed green arrows reflect the compositions of feldspar pairs and the solvus temperatures they record from initial to final crystallization. The highest fictive temperatures were recorded at the margins; Temperatures fell toward the center of experiments but generally did not attain the actual run temperature.

Textural Applications to Igneous Rocks. Fenn (1977) began this section with an honest disclaimer:

“The results of the present study have little direct application to textural development in common igneous rocks. The experiments were designed to extract quantitative nucleation and growth data from a simple system over a wide range of undercoolings. The experimental cooling rates were far higher than can be reasonably assumed for natural systems, and the isothermal, isobaric nucleation and growth steps are not as satisfactory as controlled cooling-rate experiments in the modelling [sic] of natural processes.”(p. 157)

Among his conclusions, however, Fenn (1977) observed that for the H₂O-undersaturated compositions, the relations of growth rate to nucleation density led to a few large crystals at small undercooling temperatures, and increasingly more and finer-grained crystals as the magnitude of undercooling increased. To this extent, the results conformed to the prevalent

interpretations of texture in plutonic (coarse-grained, slowly cooled) and volcanic (fine-grained, rapidly cooled) geologic environments, as Swanson (1977) also noted.

Fenn (1977) also acknowledged that by the nature of the experiments, the magnitude of liquidus undercooling fell as crystallization proceeded. This consequence has two sources: one, the crystallization of the feldspars in the undercooled experiments entailed the progressive fractionation of the melt, driving the liquid composition toward that of the equilibrium on the liquidus, and in effect bringing the liquidus temperature down. For the H₂O-undersaturated compositions, the increase in the H₂O content of the melt with crystallization of feldspars led to a further reduction of the liquidus temperature. Both results stemmed from the non-eutectic compositions of the starting materials, and Fenn (1977) noted that these are poor replications of natural igneous systems that undergo continuous non-linear cooling until they reach the temperature of their environment.

In our work on the simple haplogranite system, I chose as a composition that of the H₂O-saturated granite minimum at 200 MPa (2000 bars). In theory, that composition should have crystallized quartz and a single alkali feldspar in their proportion at the minimum without change in liquid composition. With undercooling, the solvus of the feldspar system would have been crossed at a temperature not much below the liquidus, and quartz and two feldspars would crystallize, but all in the proportion of the minimum composition. Rather than the simultaneous crystallization of feldspars and quartz in their eutectic proportions as might have been expected from the invariant composition of the melt, the liquid crystallized sequentially: first with large skeletal crystals of K-feldspar, followed by radial or graphic intergrowths of sodic feldspar and quartz (Evensen, 2001; London et al., 2006). Subsequently, London and Morgan (2017) used data published by Burnham and Nekvasil (1986) to show that in a highly undercooled melt of minimum composition (and invariant at the run temperatures of the experiments), the energy released by the crystallization of K-feldspar was greater than that of albite or of quartz *if* the system lacked calcium, as did the model system of these experiments. This calculation would account for the appearance of K-feldspar first.

Applications to Pegmatites. Fenn (1977) did not extend his results to the textural evolution of pegmatites, or to the model of Jahns and Burnham (1969), except for this comment:

“This decrease in growth rate as the H₂O saturation level is breached is contrary to the beliefs expressed by Jahns & Burnham (1958) and others who felt that the presence of an aqueous vapor should enhance the crystal growth rate. Similar observations on the effect of excess H₂O on nucleation and growth were reported by Swanson (1976) for alkali feldspars, plagioclases, and quartz crystallized from a synthetic granodiorite liquid at 8 kbar. Lofgren (1973) also reported a reduction in the apparent growth rates of plagioclases grown from hydrous liquids as the H₂O content was increased.”(p. 151-152)

This statement and the similar comment in Swanson (1977) cited above are not correct in detail. Jahns advocated that crystals that precipitated from the aqueous phase should be larger than those that crystallized from the melt (Jahns and Tuttle, 1963, Jahns and Burnham, 1969). In the experiments of Swanson (1977) and Fenn (1977), all crystallization appears to have ensued from the melt. However, Jahns never explained how all of the constituents of the coarse pegmatitic

zones were transported to the growing crystal surfaces through an aqueous solution. Jahns (1955; Jahns and Burnham, 1969) commented on the transport of alkalis and of silica, but omitted the most important component of feldspars: their alumina content. The transport of alumina in an aqueous solution, or rather the lack of it, represents the principal obstacle to a hydrothermal origin of the feldspars in igneous rocks (London, 2008, 2018).

Fenn, P.M. (1986) On the origin of graphic granite

Graphic granite refers to an intergrowth of skeletal quartz in K-feldspar. It is the texture for which the term “pegmatite” was coined by Brongniart (1813): from Homeric Greek *πηγνυμι* meaning “to make stout or rigid by binding together” (Autenrieth 1958), in allusion to the intimate, bound together, intergrowth of the two minerals. Graphic intergrowths of quartz with tourmaline or albite, of albite with phosphates, and probably others are common, but the graphic K-feldspar-quartz intergrowth is so widespread as to be a fundamental texture of pegmatites. It is surprising that graphic granite was not among the essential attributes of pegmatites that were itemized in Jahns and Burnham (1969) and Jahns (1982). Jahns barely mentioned graphic granite in passing in his various publications.

Fenn’s (1986) results derived from liquidus-undercooled experiments with the bulk compositions of the Spruce Pine and Harding composites. His work confirmed that the intergrowth of the two minerals is simultaneous, and that the feldspar is host to one or more skeletal quartz crystals. Graphic intergrowths formed only at relatively large liquidus undercooling of $\sim 160^\circ\text{C}$ for the feldspar phase. The magnitude of undercooling with respect to quartz was $\sim 15^\circ\text{C}$, and Fenn (1986) noted that run products that contained graphic intergrowths had no freely grown quartz. This observation became part of Fenn’s (1986) explanation for the intergrowth as a result of rapid crystallization of the undercooled feldspar, with local saturation of quartz along its surface, even if the bulk melt is not sufficiently (super)saturated with respect to quartz. Fenn (1986) also noted that the graphic feldspar-quartz intergrowths formed in his experiments were significantly larger than the same phases the nucleated as individual crystals in the same experiment (which pertains to quartz, but it is not clear that Fenn (1986) grew monocrystals of feldspar in the same experiments).

Fenn (1986) mentioned that H_2O may form bubbles on the crystal surfaces even though the bulk melt is undersaturated with respect to H_2O (from Fenn and Luth, 1973); London and Morgan (2017) made the same observation of their experiments. All of the experiments from Fenn’s (1986) study produced melts that were initially undersaturated in H_2O . Fenn (1986) speculated that an increasing concentration of H_2O in the melt might promote the formation of graphic granite because the maxima in the growth rates of the minerals shift progressively closer to the liquidus temperature. In that way, a pegmatite-forming melt could maintain a high rate of crystal growth even as the magnitude of liquidus undercooling decreased. Nonetheless, none of the examples of graphic granite intergrowths illustrated in Fenn (1986) is decorated with vapor bubbles, and the presence of an aqueous phase plays no role in his model:

“The origin of the intergrowth of quartz and alkali feldspar known as graphic granite has been shown to be the result of simultaneous growth of the two phases under conditions that favor the planar growth of the feldspar host. An imbalance between the growth rate of the feldspar and the

diffusivity of silica in the bulk melt creates a silica enriched boundary layer that in turn causes the interface to degrade from planar to cellular. Upon further growth of this cellular interface, the grooves between adjoining cells are greatly enriched in SiO₂, and this enrichment causes the nucleation and growth of quartz along with the feldspar. This simultaneous growth was not caused by classical eutectic crystallization but by the kinetic phenomena in the boundary layer adjacent to the interface of the growing crystal.”(p. 329-330)

Fenn’s (1986) explanation of the kinetic origin of graphic granite has not been questioned or challenged. The results have been reproduced in numerous experimental programs that entailed the substantial liquidus undercooling of viscous, H₂O-undersaturated granitic-pegmatitic melts. A few additional studies, however, have shed additional light on the nature of the intergrowth.

Ikeda et al. (2000) employed a 3D x-ray imaging technique to conclude that 90% of the quartz rods in their sample of graphic granite were interconnected. This result gives an accurate picture of a moving feldspar surface, shaped like a maze, wherein quartz precipitates continuously in the shifting troughs. A similar study by Xu et al. (2015) using x-ray and optical techniques found that many of the individual quartz rods are portions of much larger single crystals. The connectivity of rods that comprise large crystals is evident when the quartz rods of a graphic intergrowth have projected into a miarolitic cavity, and the quartz rods develop faceted terminations on the host feldspar (Figure 5). All parts of the same crystal possess the same orientation at the surface. Sharp angular relations of such crystal clusters to one another arise from the fact that the quartz crystals of the intergrowths commonly exhibit Dauphiné or Japan-Law twin relations (Xu et al., 2015). London (2008) noted that when quartz rods project into miarolitic cavities and develop terminations on the host feldspar, the crystallographic growth direction of the rods is evident, and it is almost never the normal direction of elongation along the *c*-axis direction. The rods are elongated along directions that correspond to directions that produce extremely rare rhombohedral or scalenohedral faces. London (2008) cited the Law of Bravais that relates the development of crystal faces to the atomic density of their planes, and suggested that the growth directions of the rods, which almost never develop faces, are planes of low atomic density. London (2008) further speculated that quartz crystals that can adopt this orientation with respect to the feldspar surface can advance at a higher rate because it takes less material to growth the crystal surface, owing to the low atomic density of the planes in the growth direction. London (2008) observed that micas and beryl in pegmatites commonly grow elongate on one of their equivalent *a*-axis directions, not along the *c*-axis that is the normal direction of elongation and one for which a prominent pinacoid face is commonly expressed.



Figure 5. Parallel growth of quartz terminations that emanated from a graphic intergrowth within the host microcline crystal. The red line indicates the direction of the skeletal quartz crystals within the feldspar.

Fenn and Luth (1973) and London and Morgan (2017) observed in experiments that bubbles of aqueous solution nucleate on the advancing crystallization surface (see Figure 2b of essay #9 of this series). There is no indication of a continuous aqueous film that wets the crystallization front, as Jahns envisioned. However, the prerequisite of Fenn's (1986) model is not only that the crystallization front, led by the feldspars, advances at a high growth rate, but that the melt adjacent to that crystal front is sufficiently viscous that the excluded components, principally silica in this case, cannot diffuse away from the boundary layer quickly enough to prevent the precipitation of quartz. That prerequisite would be incompatible with Jahns' core hypothesis that an aqueous solution is the principal fluid medium from which the coarse pegmatitic crystals grow because ionic diffusion through that medium would be rapid. London (2009) suggested that the formation of graphic granite and its sustained growth would not be possible from an aqueous solution because the solute content is so low that crystal growth would be slow, and ionic diffusion is so fast that there would be no build-up of silica adjacent to the feldspar surfaces. To test that hypothesis, experiments by London (2018) utilized the Macusani obsidian with a large excess of H₂O added. The runs were preconditioned to completely melt and homogenize the samples and to equilibrate the melt with the aqueous solution. Rapid quench from the preconditioning step precipitated a very large amount of glass beads that represented the melt components that were dissolved in the aqueous solution at the run condition (Figure 6a). London et al. (1988) calculated that the aqueous solution contained ~ 15 wt% of solutes at this condition, compared to 1-2 wt% for a simple granitic liquid (Burnham and Jahns, 1961). With this result as a control, two experiments replicated the preconditioning step, followed by rapid cooling to 550°C and 450°C, which is 100°C to 200°C below the liquidus temperature of the melt. The hypothesis being tested was this: if graphic feldspar-quartz intergrowths could precipitate from an aqueous solution as a result of supersaturation, then one could not make an aqueous solution that was more supersaturated in quartz-feldspar forming components than this aqueous solution at the undercooled state of the experiments. The experiment at 550°C yielded clusters of euhedral feldspar and quartz, dotted with micas, similar to textures in a miarolitic cavity of a pegmatite (Figure 6b). Graphic intergrowths were absent. The experiment at 450°C produced an abundance of a zeolite-like crystalline product (Figure 6c), but again, no indication of a graphic intergrowth of any phases. To the extent that these experiments are definitive, the answer to the hypothesis is *no, it is not possible to produce graphic quartz-feldspar intergrowths by their precipitation from an aqueous solution.*

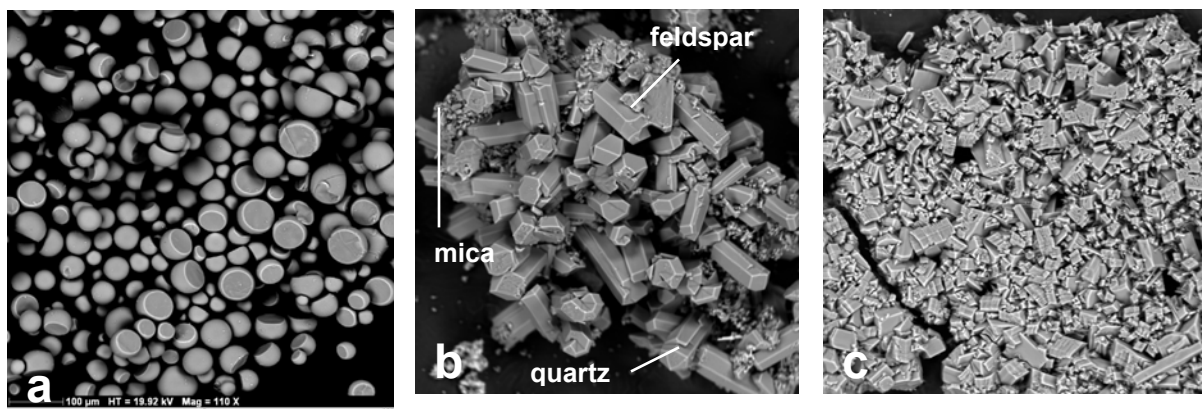


Figure 6. (a) silicate glass beads quenched from vapor; (b) feldspar, quartz, mica at 550°C; (c) a zeolite at 450°C.

The Nature of Crystallization from a Melt

Petrologists have long treated the chemical compositions of igneous rocks as their most fundamental property. That doesn't account for why their other fundamental properties of texture and degree of crystallinity were ignored so completely for so long. Among the few individuals who have pursued experimental studies, the impact of Bowen's approach, and later of C. Wayne Burnham, made the pursuit of equilibrium boundaries among phases the sole objective of the experimentation. They embraced the equilibrium approach through the methods of chemical thermodynamics that were developed by J. Willard Gibbs, but Bowen, for one, might also have realized that how one does the experiments imparts much of the "anthropogenic effect" into the results. However, the lack of experimentation that assessed the crystallization response of silicate liquids became evident with the experimental work of Lofgren, Swanson, and Fenn in the 1970s, and that explains why their studies had such a large impact on the field. What is surprising in hindsight is that their work did not set off a flurry of similar investigations. It would be more than a decade before the next study of the kinetics and textures of crystallization of an undercooled melt appeared in the literature (London et al., 1989).

Swanson (1977) and Fenn (1977) demonstrated that there is no crystallization at the liquidus temperature. Liquidus undercooling, generally by more than 50°-100°C, is necessary to initiate crystallization regardless of the time at temperature. Pronounced maxima in the rates of crystal growth occur at liquidus undercoolings of 100°-250°C. The number of crystals that nucleate as a function of volume or time, however, varies widely with the bulk composition and especially with the magnitude of liquidus undercooling in the system. The relative positions of the maxima in growth rate and nucleation rate or nucleation density determine the crystal sizes. Depending on their coincidence as a function of liquidus undercooling and melt composition, both of which vary over the course of crystallization of an igneous body, a magma could go through a wide range of crystal size distributions as a continuous function of its cooling history. Crystal habits vary with undercooling, but predictably.

In a previous essay, I cited an oft-quoted maxim from Bowen (1928):

"To many petrologists a volatile is exactly like a Maxwell demon; it does just what one may wish it to do." (p. 282)

Bowen's (1928) lament has proven truer in modern times than he could have imagined. Petrologists, like Jahns, have regarded the complex textures of pegmatites and related rocks as necessarily formed by the action of an aqueous solution because their understanding of what a melt can and cannot do was almost nil. Experiments aimed at crystallizing melts, which might have influenced petrologic thinking about the importance of the kinetic aspect of crystallization, were simply not done for the reasons mentioned above. More important than any of the details of their results, Swanson and Fenn illustrated the complexity and variety of crystallization responses of silicate liquids to the effect of cooling alone. The textural complexity far exceeds the common expectation among petrologists that the cooling of silicic melt at subsurface conditions produces ordinary granite, period.

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