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Letter to the President

More thoughts on webcasting

Dear Marty,

I read with interest your letter in the last Geochemical News raising the notion of web-casting portions of Goldschmidt Conferences. Your presentation is well balanced, and I think compelling.

The July 2002 Geochemical News published a letter from me to the Society ("Geochemistry in a sustainable world" pdf) concerning both the actual financial costs of Goldschmidt (and other Society business), but also the issues raised by such matters in terms of international development. I think the then-current officers took it seriously; it was discussed at the next Executive Committee meeting (in Davos, I recall). Since then, the Society has staged Goldschmidts at some more modest venues, and moved them around the world, both steps that I think help equilibrate the burden a bit. Also, Elsevier published the Treatise on Geochemistry in paper editions on a volume-by-volume basis, which makes that work much more accessible. Progress, and much appreciated in places where I do much of my work (e.g., Indonesia, Papua New Guinea, Laos, Brazil, South Africa).

Your leadership in raising the web-casting option is great. I strongly support the notion of making web-casts of selected sessions available to geochemists (including students!) in economically disadvantaged countries. I wonder whether there would not be a case to be made for both the general membership (in our annual registration) and the registrants for the meeting being asked to check a box that would add a nominal sum (US$5?) to the fee to be used specifically for international-outreach activities such as this. Voluntary. If half the membership and half the registrants contributed the cost of a couple cups of coffee, you would have the funding you estimate and leave the meeting revenue-neutral with respect to this.

One additional thought on your articulation of the the costs for a meeting such as Goldschmidt. I have greatly enjoyed, and I think profited intellectually, from the Goldschmidts that I have attended. But we might also consider the opportunity costs. I haven't done the detailed costs analysis, but I would guess that, all in, a week in Davos would cost me about $5,000, and somewhat more than that again in foregone revenue - all out-of-pocket for some of us who do not have grants with travel budgets. That's a lot of money, and the impact of that much (or even a portion of it) applied in other ways might have many environmental and social benefits. It would pay for the web-cast, for a full set of the Treatise for the Library at University of Papua New Guinea, or provide enough treated mosquito-netting or antidiarrheal solution to save a thousand children...

Keep up the good work.

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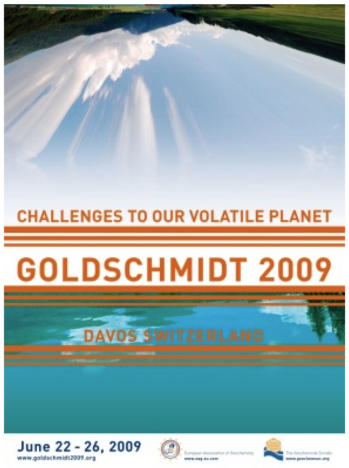
Goldschmidt bloggers needed

[Nine bloggers have been chosen. Follow along at http://geochemicalnews.wordpress.com/]

The Geochemical Society is looking for attendees of the 2009 Goldschmidt meeting in Davos, Switzerland to join GS President Martin Goldhaber in blogging about the meeting. All levels of experience--from undergraduates to emeriti--are encouraged to participate. No previous blogging experience required! The blog will appear on the GS homepage and will be an interactive tool for those on-site as well as those unable to attend. Please consider this unique opportunity to contribute to the society!

Requirements

- Must be attending the 2009 Goldschmidt conference
- Must have access to the internet while at Goldschmidt
- Must have command of English grammar
- Must have ability to write interesting and comprehensible blog entries
- Ability to attend the 'Earth's Future' panel discussion on Wednesday, June 24 is a plus
- Ability to attend the morning plenary talks is a plus



Expectations

- Post at least one blog entry per day while at Goldschmidt
- If necessary, address questions related to posts

Compensation

- 2009 (or 2010 if already a current member) Geochemical Society Membership
- Geochemical Society T-shirt
- Choice of one book (GS Special Publications or RiMG volumes)
- Ticket to the Conference dinner on Wednesday June 24

The deadline for applications is May 27, 2009.

All interested contributors should email the following information to Seth Davis (seth.davis@geochemsoc.org), please write "Goldschmidt Blogger" in the subject line.

1. Your name
2. Your email address
3. Position/experience level
4. If student, degree you are seeking and expected completion date along with your advisor's name and email address
5. Your field(s) of interest
6. Dates you will attend the Goldschmidt conference
7. Sessions you plan to attend
8. Do you plan to attend the Earth's Future panel discussion?
9. Do you plan to attend the the plenary talks?
10. Your T-shirt size (women-please indicate the equivalent men's size)
11. Book Preference - list either a Special Publication or RIMG title

All submissions will be acknowledge by email within 24 hours. If you do not receive an email then your submission was not received. Another email will be sent when the final selections are made.

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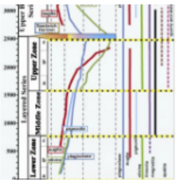
Controversies of Consequence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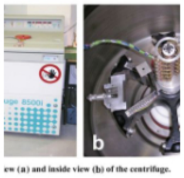
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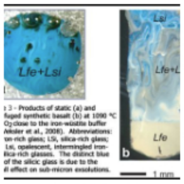
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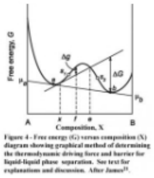
Figure 4

Discussion of Liquid Immiscibility in the Skaergaard Intrusion

I.V. Veksler[1,2], S.A. Morse[3] and A.R. Philpotts[4,5]

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Introduction

(by the *Geochemical News* staff)

The Skaergaard Intrusion of East Greenland is a cornerstone of igneous petrology. The intrusion was discovered by L.R. Wager in 1931 and documented by L.R. Wager and W.A. Deer in 1939[1]. Most of us know the Skaergaard from the magnificent book by L.R. Wager and G.M. Brown[2], which influences nearly all petrologists, including those who have never read it.

Certain compositional trends in the Skaergaard Intrusion still confound petrologists. One of these is the extreme iron enrichment in the layered rocks. The iron enrichment trend is dependant, in part, on when and where magnetite precipitated from the Skaergaard magma. Magnetite fractionation is enhanced by larger oxygen fugacities (fO_2) in the magma. Early magnetite subtraction sends the residual magma on a path toward silica enrichment (the Bowen trend). Smaller fO_2 values delay magnetite precipitation and residual magma becomes enriched in iron (the Fenner trend). These issues play into central questions about the Skaergaard magma: When, where and why did the magma start evolving toward iron- or silica-rich compositions?

In October 2007, Dr. I. Veksler and colleagues published the results of experiments on synthetic basalt whose composition is analogous to the parental magma of the Skaergaard Intrusion.[3, 4]. Their experimental innovation was to centrifuge the hot liquid basalt before it was quenched. Upon quenching, the experimentalists found a thin layer of SiO_2-rich and FeO-poor glass atop quenched basalt. These new data, the authors say, suggest that the Skaergaard Intrusion's compositional trends reflect unmixing of SiO_2-richer, FeO-poorer liquid from its conjugate. Spatial separation of the two liquids because of their different buoyancies, and perhaps other factors, would set a new evolutionary course for the freezing magma. These results might help answer lingering questions about silica and iron enrichment of the Skaergaard magma.

Dr. Veksler's paper elicited rapid responses from scientists with distinguished careers of research and publication on cumulate rocks. Among the commentators are Drs. S.A. Morse[5] and A.R. Philpotts[6], both of whom question the relevance of the new experiments and reassert that fractional crystallization, albeit complicated by post-cumulus processes, produced the compositional trends in the Skaergaard Intrusion. An important disagreement concerns rocks at the top of the intrusion that formed from unmixed liquids. Are these frozen, unmixed liquids compositionally and physically representative of the main magma body? Some people believe the rocks are prima facie evidence of large-scale liquid immiscibility in the magma cavity. Others agree that unmixing occurred, but only in isolated, terminal interstices and small spaces within minerals; it was of little importance in the magma chamber. Please read a judicious sample of the references to discover the origins and evidence for these divergent ideas.

We recently asked Drs. Veksler, Morse and Philpotts about liquid immiscibility in the Skaergaard magma and, more generally, cumulate crystallization in magma chambers. We hope readers recognize that the scientists cannot fully explain their views in this short interchange. Certain passages here are disputed by one or another participant. We all are well-trained in the strict conventions of peer-reviewed publication, but not as well in the unstructured format of this presentation.

An interesting historical aside is that N.L. Bowen curtly dismissed liquid immiscibility in The Evolution of the Igneous Rocks (1928). Chapter II of Bowen's book ends with this one-sentence paragraph: "On the basis of immiscibility of any kind it is impossible to build up an adequate explanation of the associated members of rock series which is the fundamental problem of petrology." One may speculate on the influence of this statement on modern ideas of petrogenesis.

Questions for Dr. Veksler

GN. What guided your choice of initial basalt composition? Can you describe your centrifuge? What were results of your experiments?

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First, I wish to emphasize that these experiments were conducted with great care and attention to detail. Every effort was made to ensure the validity of the experimental protocols . Our careful laboratory procedures give us confidence in the results.

Centrifugation is a simple and very effective method of phase separation, popular among wet chemists, biologists and medics, but seldom used by experimental petrologists. Our centrifuge is a powerful Cryofuge 8500i (Fig. 2) adapted to carry a small 1-atm electric furnace or an Ar- pressurized internally heated pressure vessel. The centrifuge is equipped with a built-in refrigerator that can keep the inner chamber at a constant, low temperature. This option is very useful in our experiments because it allows to get faster quenching and to keep cold seals of the thermocouples at room temperature despite significant heat coming from the rotating furnace. Small samples (50-200 mg) of geological melts and fluids in sealed containers can be heated to temperatures up to 1300°C at the atmospheric pressure or, in case the pressure vessel is used, to 1100°C at pressures up to 100 MPa. The centrifugal pool can go up to 1000g (g = 9.8 m/s^2). We used the centrifuge in the past for falling-sphere viscosimetry of water-bearing granitic melts[7] and in numerous studies of element partitioning between immiscible silicate, borosilicate, carbonate, phosphate, sulphate, fluoride, chloride melts and various brines.[8,9]

Our starting mixtures were based on published electron microprobe analyses of natural glasses from volcanic rocks and melt inclusions in plagioclase phenocrysts that had been interpreted as pairs of quenched immiscible liquids. We tried to collect and test all the reported cases of immiscibility in natural basaltic and andesitic lavas paying special attention to examples of unmixing that had been claimed to take place at or above 1040-1050°C. We calculated and synthesized from reagent-grade chemicals a number of intermediate compositions between each pair of conjugate immiscible liquids. The proportions of liquids were variable and arbitrary. Usually we chose mixtures with equal amounts of both liquids or a higher proportion of low-viscosity Fe-rich immiscible melt. In addition, we prepared two Skaergaard model liquid compositions at about 60% crystallization of the Skaergaard magma. The compositions were based on (1) experimental glasses from a study of crystallization in the Skaergaard parental magma[10] and (2) mass balance model based on subtraction of successive cumulate layers from the bulk Skaergaard liquid.[11]

Centrifugation experiments were carried out on carefully fused and homogenized melts preheated to temperatures above the miscibility gap. We also did a few static experiments in 1-atm furnaces under controlled oxygen fugacity. Centrifugation was crucial for the study of silicate liquid immiscibility because it enabled us to clarify the nature and the origin of sub-micron amorphous exsolutions in quenched glasses. We realized early during the study that unmixing of some silicate liquids, including many ferrobasaltic compositions, developed slowly. When the nucleation density of newly formed droplets is high and their growth rate is slow, unmixing may lead to formation of sub-micron emulsion that would not coarsen for many hours or even days. Such emulsions quench to turbid, milky glasses. On the other hand, very similar opalescent glasses are also known to form when a homogenous liquid passes during quenching through a region of metastable, sub-liquidus immiscibility. Static quench experiments often produce turbid, opalescent glasses but they tell little about the origin of heterogeneity, whether it is due to slow unmixing above liquidus or fast exsolution during quenching. In situ centrifugation at 1000g dramatically enhances phase separation at high temperature and helps to distinguish between the two cases (Fig. 3).

Within a few hours time of our centrifugation runs some immiscible emulsions partly or completely separated in two condensed layers of conjugate liquids that quenched to clear glasses. In those cases high-temperature immiscibility was obvious. In the second group of compositions, phase separation at high temperature was not so clear. Droplets remained very small, less than a micron in diameter, but nevertheless they clearly moved along the direction of the centrifugal pool. This movement macroscopically revealed itself by compositional gradients along the vertical axis of centrifuged samples. Finally, there were compositions that produced turbid glasses without significant compositional gradients, and those were interpreted as products of quench exsolution. One of the model compositions (based on the experimental data) showed morphological signs of unmixing of the second type, with a small amount of sub-micron silica-rich droplets rising to the top of the charge. This composition at 1100-1120°C apparently was very near the border of the miscibility gap, on its Fe-rich side. Immiscibility has never been reported in static experiments on Skaergaard compositions, and the vast majority of other tholeiitic liquids unmixed in static experiments at much lower temperatures (below 1040°C).

GN. How do the laboratory results help explain the problematic modal and compositional trends in the rocks? Do you believe that liquid immiscibility did occur or that it may have occurred? Can you think of way to test for liquid immiscibility in the Skaergaard magma?

Immiscibility almost certainly occurred in the Skaergaard intrusion. Many experimentalists and field geologists agree that the uppermost parts of the Layered Series formed fr om immiscible Fe-rich and silica-rich liquids. In fact, Dr. McBirney was among those who experimentally demonstrated immiscibility between extremely Fe-rich (28-30 wt. % FeO) and silicic (melanogranophyric) liquids that, according to his model, formed at the very end of fractional crystallization at Skaergaard.[12, 13] If there had been any doubts that the Skaergaard magma actually reached the level of Fe concentrations required for silicate liquid immiscibility, those doubts were crushed by direct evidence from contrasting groups of Fe-rich and silica-rich melt inclusions trapped in cumulus apatite from the two uppermost sub-zones of the Skaergaard Layered Series.[14] Furthermore, it has been repeatedly shown that most common basaltic magmas unmix at the last stages of crystallization. This was proven experimentally and petrographically in many classical works.[15, 16]

The question now is when did immiscibility actually start and how important was the impact of liquid-liquid separation on magma evolution at Skaergaard? Our centrifugation experiments put the onset of liquid immiscibility roughly at the transition from the Lower to the Upper Zone of the Layered Series or approximately at 60% crystallization of the magma. This is a crucial stage of magma evolution when Fe-Ti oxides start to crystallize, olivine is about to react out, and liquid lines of descent crystallize, olivine is about to react out, and liquid lines of descent, proposed in numerous alternative models, fan out in different directions towards silica or Fe enrichment. We do not claim that our experiments decisively proved early liquid immiscibility in the Lower and Middle Zones. However, we believe that our study showed that such immiscibility is possible. Our centrifugation study also added serious doubts with regards to conventional experimental methods, whether they really can adequately reproduce immiscibility, and work with ferrodacitic-rhyolitic liquids at temperatures below 1100°C when the kinetics becomes a serious issue.

In our view, further experiments on ferrobasaltic compositions are not likely to bring decisive evidence for or against early liquid immiscibility at Skaergaard. Even if kinetic problems are sorted out, and phase equilibria at low temperatures better constrained, the real impact of liquid immiscibility on Skaergaard magma can be assessed only from the rocks. That is why we closely collaborate with Dr. Marian Holness (University of Cambridge, UK), Dr. Troels Nielsen (GEUS, Copenhagen), Dr. Christian Tegner (University of Aarhus, Denmark), and their students. These people are running very interesting geochemical and petrographic studies of the Skaergaard intrusion. The amount of new information that they extract from Skaergaard rocks using innovative methods such as detailed studies of rock textures[17] or melt inclusions[14] is astonishing. In the near future, important evidence with regards to immiscibility is expected to come from new geochemical studies of the Upper Border Series that are carried out at the University of Aarhus, and inter-cumulus mineral assemblages in the rocks of Layered and Contact Border Series investigated in Cambridge.

Although the exact timing of immiscibility at Skaergaard remains uncertain there is, however, no mystery about the Bowen and Fenner trends any more. The chemistry and phase equilibria principles behind the trends were fully explained by experimental studies of the 1980's and 1990's (see our recent review and discussion[18]). In the introduction, you correctly emphasized the importance of magnetite crystallization and redox conditions. The oxidation state of magma is very important for magnetite stability but so is the melt composition. Regardless of redox conditions, basaltic magma will not evolve towards silica enrichment and rhyolitic residual liquids by fractional crystallization unless there is a sufficient initial concentration and constant increase of alkalis in crystallizing melt. This chemical requirement for the Bowen trend is clear and simple because normative albite and orthoclase are key constituents of rhyolitic melts, and alkali oxide components, being strong Lewis bases, greatly decrease the activity coefficient of SiO_2. Alkali-free basaltic liquids have been shown, on the other hand, to evolve in reducing conditions along the Fenner trend, that is, towards extreme Fe enrichment (up to 31 wt.% FeO) and SiO_2 concentrations around 46-47 wt.%[19]. Common tholeiitic basalts similar to the Skaergaard parental magma are certainly not alkali-free, and that is why crystallization experiments on Skaergaard compositions[10, 20] never showed FeO enrichment above 22 wt. %. However, Fe enrichment in natural tholeiitic magmas at Skaergaard and in other places has been shown to result in liquid immiscibility that produces a mixture of Bowen- and Fenner-type liquids, both in peaceful coexistence. Thus, we conclude from the wealth of experimental evidence that ($FeO+Fe_2O_3$) concentrations significantly above 22 wt. % cannot be reached in Skaergaard-type magmas by fractional crystallization alone but only in combination with liquid immiscibility.

We urge interested experimental petrologists to shift their attention from redox equilibria that have been already studied very well to immiscibility phemomena that have been almost neglected. Silicate liquid immiscibility is rooted in fundamental properties of silicate melts, and it is not an unimportant curiosity. It is very likely that large bodies of gabbros and granites, as well as their volcanic analogues, crystallized from immiscible silicate emulsions. The spell of Bowen's pronouncement that you cite in the introduction is probably the only explanation why generations of igneous petrologists stubbornly ignore silicate liquid immiscibility.

GN. You state "...that unmixing of complex aluminosilicate liquids may be seriously kinetically hampered (presumably by a nucleation barrier)." What sort of barrier do you mean? Wouldn't the magma contain low-energy nucleation sites such as suspended crystallites or gas bubbles? From a thermodynamic standpoint, unmixing must decrease the overall free energy of the magma. Have you thought about why unmixed magma is more stable than homogeneous magma?

Obvious kinetic limitations arise from material transport processes, for instance, from low diffusion rates or high viscosity. If the kinetics of material transport is poor, access to thermodynamic equilibrium may be so slow that in practice the system may look stagnant and show no significant changes during experiments of a reasonable duration. A formidable thermodynamic barrier, on the other hand, may completely block a path to equilibrium, and indefinitely keep a system in a metastable state. According to theoretical models developed mostly for technological glasses (see, for example, a review by James[21]), thermodynamic barriers for melt unmixing may be of different nature.

First, there is a universal energy barrier for hatching of a new phase, which is related to formation of interfaces. Interfaces are sites of excess surface energy, and unmixing, like crystallization, must overcome the interface energy hurdle. Interface energies between immiscible silicate liquids are at the moment unknown. These energies are a key factor of emulsion stability and we are trying to measure them in our on-going experimental project. Heterogeneous nucleation on crystals or gas bubbles that you mention is a way to decrease the interface energy barrier.

In addition, there is a thermodynamic barrier that is specific for unmixing by nucleation and growth. It can be graphically explained in a plot of the Gibbs free energy variations (*G*) with composition *x* at constant pressure and temperature (Fig. 4). In a simple binary system A-B where *G* changes smoothly with composition, the *G*-*x* curve is characterised in the region of unmixing by a local maximum. Tangent points a and b of the common tangent line define the equilibrium chemical potentials of the components μA and μB, and the compositions of the equilibrium immiscible liquids a and b. In the compositional interval between a and b a single homogenous liquid has a higher free energy than a mixture of the two immiscible liquids. Other important elements of the plot are inflexion points s_1 and s_2 where the second derivative $(\delta 2G/\delta x2)T,P = 0$. For the bulk composition *x* lying between points a and s_1 the overall free energy decrease due to unmixing to equilibrium liquids a and b (excluding interfacial effects) graphically corresponds to the vertical segment ΔG between the free energy curve at composition b and the tangent drawn to the curve at the composition x. This energy drop is the driving force for separation of the equilibrium phases. It should be noted, however, that when phase separation develops in a nucleation and growth regime, small sub-critical compositional fluctuations (represented for example, by composition f) will produce an increase in free energy (the segment Δg) The increase represents a thermodynamic barrier for unmixing. Clearly a fluctuation must develop beyond the composition e before the free energy starts to decrease, and the barrier is overcome. Thus, in nucleation region, the system is metastable with respect to infinitesimal compositional fluctuations. In contrast, there is no thermodynamic barrier of this kind for phase separation within the co-called spinodal region between the inflection points s_1 and s_2 as the free energy change is negative for an infinitesimal fluctuation.

GN. If liquid immiscibility affected the rock types and compositions, the densities of the high- and low-SiO_2 liquids and the parental magma must all have differed. Only then could the high- and low-SiO_2 liquids take independent paths through the magma chamber. Do you agree with this reasoning? If so, have you tried to calculate the densities of the two unmixed liquids and the parental basalt?

Yes, this reasoning is correct. Without spatial separation (e.g., by gravity) immiscibility will not produce any noticeable effects on the modal composition and geochemistry of fully crystallized rocks. Textural effects (if any) would depend on how the size of liquid droplets compares to the size of crystallizing mineral grains. There are many empirical models for calculation of melt density from partial molar volumes of major oxide components. According to the latest and most universal one[22], calculated melt densities of the silica-rich and Fe-rich Skaergaard liquids from the centrifugation experiment at 1100°C (sample C-111 in our original paper) are 2470 and 2774 kg/m3. The density difference of 11% is significant. It is similar to or even greater than the volume effect of crystallization of common rock-forming silicates at the atmospheric pressure.

GN. Why do you think that your paper generated so much controversy? Have your thoughts about liquid immiscibility at the Skaergaard Intrusion changed because of the comment and reply process?

As you pointed out in the introduction, the Skaergaard intrusion has been for half a century the most important natural laboratory where many basic concepts of igneous petrology were formulated. At first, there was a simple and beautiful Wager model that explained, let us say, 80% of observations at Skaergaard. Fractional crystallization and crystal settling by gravity certainly are very important processes that shaped the intrusion. However, the remaining 20% of observations remained unaccounted for, and the number of facts that would not fit into the classical model has been growing. It is probably fair to say that during the last 30 years there has been a growing feeling among Skaergaard experts and a broader geoscience community that some important elements are missing from the existing models of layered gabbroic intrusions. The recent review by McBirney[23] summarized those misgivings. If there are 10 or 20 viable explanations for the origin of rhythmic modal layering, we must conclude that we do not understand how the layering formed. Scientists have to use Ockham's razor but the instrument is double-edged. It discourages us from paying attention to details that defy a simple explanation, and make us too wary about radical amendments. The hypothesis of early liquid immiscibility is a rather radical one. We would never have started to consider it seriously if we could find some other way to reconcile key problems of magma evolution at Skaergaard that have been repeatedly debated during the last two decades. Another crucial encouragement came at the moment when we saw melt inclusions in Skaergaard apatite. They were so obvious that we could not believe our eyes! We realized that, amazingly, despite decades of extensive research, some tell-tale signs in Skaergaard rocks may have been overlooked.

When we wrote the paper and prepared ourselves for the review process we were mostly concerned with the apparent contradiction between our centrifugation results and numerous products of static experiments on a broad spectrum of basaltic composition that had not showed immiscibility. Surprisingly, and to our relief, the harshest criticism that we received in the peer reviews, two of which were very positive, and the other two very negative, was directed mostly against the geologic implications rather than the experimental results. That was fine. Many experimental petrologists, with the exception of Dr. Philpotts, met the results of our centrifugation experiments very positively. Field-oriented petrologists have been more sceptical. It took us at least three years to think through and get used to the idea of early immiscibility in the Skaergaard intrusion, and we cannot expect other people to embrace it immediately. We greatly appreciate the interest to our work from Professors McBirney, Morse and Philpotts who are the leading experts on layered intrusions and liquid immiscibility, and value their critical comments. However, peer reviews and the lively discussion that followed have not challenged us so far with an unbeatable argument against the early magma unmixing. No matter what the final verdict, we are happy to see the Skaergaard discussion revived. It is about time for us igneous petrologists to update our models of layered gabbroic intrusions, and agree on fundamental issues that remained unresolved since the previous round of the Skaergaard debate 20 years ago.

Questions for Drs. S.A. Morse and A.R. Philpotts

GN. Aside from their relevance to the Skaergaard complex, what do you think of the experiments by Dr. Veksler and his group? Can they help us understand the petrology and geochemistry of layered intrusions?

Dr.Morse - No. I don't see that they have added much new to the game. The technology is interesting in that they used Don Dingwell's centrifuge to look at the effect of accelerations in melts. And in some of the unpublished later images that I saw with Ilya [Veksler] at a poster session, I was pleased to see that the highly polymerized felsic melts trapped the more fluid, denser melts, and I thought that was a nice illustration of the effect of polymerization. That also made me think of Tony Philpotts's (ARP hereafter) feldspar networks in the Holyoke basalt and in the lab, a study that I think has opened up a welcome new understanding of magmatic differentiation.

Dr. Philpotts - Although the experimental results of Veksler et al.[3] using the centrifuge are interesting, I believe the immiscibility they obtain at ~1100°C is a metastable product formed during the slow quench in the centrifuge is a metastable product formed during the slow quench in the centrifuge. Previous studies have shown that immiscibility in these natural basalt compositions occurs at late stages of crystallization in an extremely iron-enriched liquid residue at temperatures between 1040 -1010°C (see references in Philpotts[6]). Previous studies also have had little difficulty in nucleating and growing large enough droplets (>10 µm) to analyze with the electron microprobe without having to resort to the complexity inherent in the centrifuge technique. In previous studies, the unmixing of liquids was reversed, leaving no doubt that the top of the two-liquid field for these compositions is ~1040°C. By contrast, the centrifuge experiments produced immiscibility only by cooling homogeneous liquid and never heating up a two-liquid mixture until it homogenized; that is, their experiments did not demonstrate reversibility.

Because the Veksler et al.[3] experiments did not demonstrate equilibrium, I don't believe they help us understand processes taking place in slowly cooled layered intrusions.

GN: Please describe one field area where you conclude that liquid immiscibility did occur. What specific field, petrologic and geochemical features led you to this conclusion? What other explanations did you consider for this area and why were they inadequate?

Dr.Morse - I have no field knowledge of this phenomenon so I'm going to defer to Dr. Philpotts on this. I strongly recommend his discussions of liquid immiscibility at Section 14.7 and feldspar networks at p. 329 in his wonderful new book with Jay Ague[25].

Dr. Philpotts - The clearest field evidence for liquid immiscibility is the presence of immiscible glassy droplets in the mesostasis of tholeiitic flood basalts. One of the best examples of this is found in the Holyoke basalt of the Mesozoic Hartford Basin in Connecticut[24] where the mesostasis, which constitutes approximately one-third of this basalt, consists of glassy immiscible droplets of Fe-rich and Si-rich glass. Almost all tholeiitic basalts show some evidence of glassy immiscible droplets in their liquid residues[15] as long as they are pahoehoe flows; aa flows do not show such evidence, probably as a result of their higher oxidation state and earlier crystallization of magnetite, which prevents the iron enrichment necessary to encounter the two-liquid field. In the case of the Holyoke basalt, its residual liquids were very similar in composition to late stage liquids in the Skaergaard intrusion, and it is not surprising that McBirney[27] was able to experimentally produce immiscible liquids in such compositions. The immiscibility he obtained, however, was at much lower temperature (~1010 °C) and in a more iron-rich composition than that the reported in the Veksler et al. experiments.

GN: What are some of the differences between the site where liquid immiscibility did occur and the Skaergaard Intrusion, where you discount the influence of liquid immiscibility?

Dr. Morse - Here I defer to [A.R.] McBirney's 1996 discussion in Cawthorn's book[26] and to my text in the comment[5]. I find it abundantly clear that McBirney had it right and that the immiscibility at Skaergaard happened within the crystal mush. There is only one experiment in the Veksler paper[3] that was really relevant, and that one shows, again, that the mafic conjugate liquid is so close to the bulk starting material, and the felsic member of the pair so far away toward silica, that there could be little if any effect on the course of liquid differentiation.

Dr. Philpotts - I believe immiscibility did occur in the Skaergaard but not at the early stage claimed by Veksler et al[3]. As indicated by McBirney's experiments[27], immiscibility probably played a role in generating the melanogranophyre from the ferrodiorite. This separation, however, occurred at such a late stage in the crystallization that it probably took place in a crystal mush. This would have allowed for only local segregation of the immiscible liquids. Identical thin sheets (~ 1 cm) of melanograophyre form within ferrodiorite segregation sheets in the central part of the thick Holyoke flood basalt, which has the clear evidence of immiscible glasses in its more rapidly cooled entablature[28]. The segregation of the ferrodiorite and melanogranophyre in this flood basalt has been shown to have resulted from compaction of the crystal mush in the lower part of the flow.[29, 30] This is very different from the immiscibility being invoked by Veksler et al.[3], where separation is thought to have occurred at such an early stage of crystallization that large scale segregation of different liquids could have occurred in the magma chamber.

GN: Your published comments reassert the primacy of fractional crystallization at the Skaergaard Intrusion. Is it true, however, that you are discontented with extant explanations for crystallization of the Skaergaard Intrusion? If so, what specific aspects of the rocks do not fit in a holistic model?

Dr. Morse - What do you mean by extant? My view of Skaergaard is that Wager and Deer and Brown all got it right, and everything we do with the real samples only strengthens that conviction. What is a holistic model? Isn't that the standard model?

My own direct involvement with Skaergaard is limited to the thin sections in the case that sat by my elbow one winter in Cambridge and as seen through Stuart Agrell's microscope, with Alex Deer my office-mate.

But my indirect involvement has had to do with phase equilibria, reaction constants, and oxygen and silica activities that resulted in the 1980 paper with Don Lindsley and Richard Williams[31]. This was followed by my own study of Kiglapait Fe-Ti oxides, again with reference to conditions at Skaergaard, in 1980[32]. I discussed these parameters again in 1990[33]. I also discussed the cumulate paradigm invented by Wager and others at Skaergaard[34] and had the great pleasure to collaborate in a small way with Marian Holness and others[17] on a remarkable new direction in the study of cumulate rocks, in which we see liquidus events through the maturing of the dihedral angles of augite among plagioclase tablets, and via the influence of the fractional latent heat. That's a big aspect of wisdom to come.

Dr. Philpotts - As pointed out above, I do believe that immiscibility played a role in the late stages of crystallization of the Skaergaard to produce the melanogranophyre

melanogranophyre

GN: How can we best advance our understanding of the Skaergaard Intrusion and other layered intrusions? What sorts of new research directions have you taken in your own work on layered intrusions?

Dr. Morse - The Skaergaard intrusion is unique, but it is not unusual. We profit by studying other examples of strong differentiation. Other differentiated bodies of troctolitic lineage abound. Kiglapait is one, but in Labrador it is accompanied by two dozen or more bodies with troctolitic affinities. These include Jon Berg's Hettasch Intrusion (referenced in Morse[35]) and Bob Wiebe's troctolitic pillows chilled in molten granite at Newark Island. Examples in the Duluth Complex of Minnesota include at least one (Sonju Lake[36]) specifically found similar to Skaergaard in its paragenesis. As for distant comparisons, remarkably, the Fo-An patterns of Kiglapait and Bushveld are essentially identical in form but offset in Mg.[37]

Skaergaard appears to be a strange attractor of funny ideas, perhaps directly because people think it is unusual. In one such approach, Hunter and Sparks proposed the escape of a late rhyolitic component from the intrusion, thereby in their thinking giving it a Bowen flavor. The ensuing discussions included a strong one by McBirney and Naslund[13] in which they presented the results of a difficult experimental melting study (more later on that). My own discussion,[33] which after revisiting it I must humbly recommend, was devoted to a comparison of Kiglapait and Skaergaard as well as Kilauea and Nain, with particular emphasis on silica and oxygen activities and iron enrichment, phase equilibria, and residual liquids. The point is, the two intrusions, of greatly different size and setting, are so alike that their differences are simply those of degree (measurable at that) rather than kind. This conclusion and the other examples mentioned above strengthen the notion that a common petrologic process operates to yield very similar, and not unusual, results in the rocks.

But who would have guessed that the composition of plagioclase in the liquid, at saturation with augite, would be the same in both Kiglapait and Skaergaard? Taking advantage of this surprising result, I worked from our experimental studies to generate a model for the Skaergaard liquidus temperature with crystallization progress.[38] Most interestingly, this model and that of McBirney and Naslund are practically indistinguishable; they might both be wrong, but at least they agree from very different approaches.

What we still need is a good anchor for the liquidus temperature of the Skaergaard Upper Zone. This is a conceptually simple experiment made very complicated by the fact that the experimental crystals of pyroxene and olivine will be unlike the natural ones slowly cooled, because the natural pyroxenes have lost Al and the olivines lost Ca to their coexisting feldspars during cooling.

I admire the initiative taken by Marian Holness and colleagues, in which petrographic studies are being applied in a very new way to tease liquidus and subliquidus histories out of the Skaergaard rocks. Abundant and welcome new data will be forthcoming from Christian Tegner's analyses of mineral compositions in new Skaergaard sampling.

The variation of mineral compositions with fractionation progress has to my mind always been the meat and potatoes of the study of layered intrusions. I look for appropriate Rayleigh equations that will run through the compositional data and ask what they mean. If they fit at all, they tell us something. I have used plagioclase and XMg data for Kiglapait and Skaergaard as study cases, but also data from the Duluth Complex and other intrusions such as the great Windimurra Intrusion in Australia.[39] The bottom line so far is that the data suggest the existence of an internal magma reservoir with varying probability like $P \sim 0.65$ for Skaergaard and $P = 1.0$ for Kiglapait, but with some variation depending upon whether plagioclase or olivine is being examined. This game seems fruitful.[40]

More recently, I have again taken up the study of residual porosity by the proxy of the An range in plagioclase as determined in grain mounts, so that the sample volume approximates to the equivalent of ~200 g of rock. The results are astounding, even showing spikes in the olivine Fo range that one would not have expected. Stay tuned (but for a peek, check out my Spring AGU abstract for 2009).

Dr. Philpotts - I believe the view that magma chambers are large bodies of liquid that slowly solidify in from the roof and walls while dense minerals accumulate on the floor is probably not accurate. Instead, I think most magma chambers soon become congested with crystals, which are dispersed away from the roof and walls by convection. As soon as these crystals become sufficiently abundant, the magma chamber is filled with crystal mush, and most of the differentiation that takes place in these bodies does so as a result of crystal-mush compaction. The flow of liquid through this porous medium becomes an important factor, as does the recrystallization that takes place in the solid fraction. The result is that many of the textures developed in the so-called cumulate layers are not due to sedimentation as originally proposed, but are more akin to metamorphic textures. The modeling of the flow of residual liquid through piles of compacting crystal mush by Boudreau[41] and Boudreau and Philpotts[30] and the demonstration by Holness et al.[42] that cumulate textures owe much to recrystallization are examples of new approaches to studying layered intrusions.

GN: Dr. Veksler's paper seemed to be rather polarizing. Why was the paper so provocative? Are your ideas about liquid immiscibility or the Skaergaard Intrusion different today than before you read Dr. Veksler's paper?

Dr. Morse - Polarizing for somewhat the same reasons as discussed above, in particular the perspective that the Skaergaard is unusual (and needs to be fixed!). No, my ideas about Skaergaard have been influenced much more by the Holness study and my own work. As to liquid immiscibility, the devil sitting on my shoulder to make me think about that matter has always been, for many happy years, Tony Philpotts. And of course, the incomparable Ed Roedder.

Dr. Philpotts - The paper was controversial because it ignored the findings of previous studies, which were clearly in conflict with the results they claimed from their centrifuge experiments. In the past, experimentalists have devoted considerable effort to demonstrate that their results represent a close approach to equilibrium. This commonly required obtaining reversals of phase transitions. In the case of the Veksler experiments this would have required not only causing a homogeneous liquid to split into two liquids on cooling, but to homogenize those two liquids on heating. This was not done. Previous experimental studies where reversals had been obtained[43] indicated that immiscibility occurred at a much lower temperature than obtained in the Veksler study. Previous studies also demonstrated the ease with which metastable immiscibilty occurred during quenching.[44] Indeed, this metastable separation is made use of commercially by Corning in creating its PyroCeram® products. In summary, although the introduction of the centrifuge to the experiments was novel, the experiments were not carried out with the same rigor as in previous studies.

My ideas about the Skaergaard, or the role of liquid immiscibility in the differentiation of igneous rocks, were not changed by the Veksler[3] paper.

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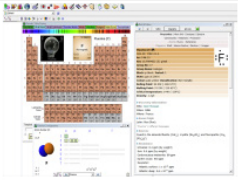
Geochemical freeware

We trolled the internet in search of freeware geochemistry programs. All but a few of the programs listed below are free; others are shareware, payware or partial-ware (also called cripple-ware, but we don't like that word). We stayed away from such superstars as PHREEQC and MELTS.

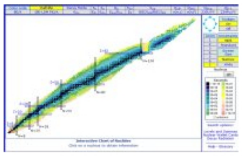
If we had a nickel for every electronic periodic table, we could start our own geochemistry journal and our papers would never be rejected. Similarly common are more or less useful plotting routines, countless unit converters and hundreds of GIS mapping packages. Surprisingly few geochemistry programs are flexible enough to serve more than a small subset of our science. Programs that are adjustable enough to serve multiple specialties are extravagantly overpriced. Priced Aquachem® lately?

We hope that some of the programs are useful to some of you. If you know of useful but perhaps obscure freeware that will help more than one segment of our Society, please send us the URLs. We will publish another software collection when the suggestions reach a critical mass.

Periodic Table Explorer (PTE)
The most complete, informative and elegant periodic table we've seen. Aside from the usual nuclear information, PTE includes: numerical, graphical and animated electron orbitals; brief YouTube movies about elements; electronegativities; isotopic abundances and half-lives; important compounds and reactions; electromagnetic spectra; melting points; densities; elastic properties; production methods; biographies of discoverers; and on and on. No ads, no broken links, no WWW hassles.

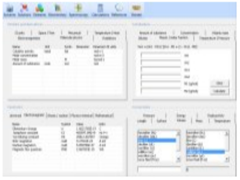


Interactive Chart of the Nuclides
The General Electric Company published the First Edition of the Chart of the Nuclides in 1948. The 16th Edition (2002) is available at www.ChartOfTheNuclides.com ($25.00). It is 1.3 meters by 0.9 meters and comes with a 48-page booklet that explains decay mode. Only the wall chart conveys the sheer majesty nuclide array. On the other hand, it is hardly portable. The National Nuclear Data Center's online version is convenient and, at this writing, some constants were updated on December 2, 2008. Turn off the Tooltips to speed up the table response. Try the fast Java version here.

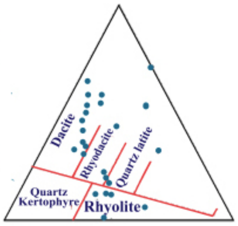


Earth's Core
This program aspires to be an electronic version of The Rock-Forming Minerals (Deer, Howie and Zussman, 1992). It compiles mineral photographs, crystal systems and habits, and locations of major deposits minerals, rocks and gems. The mineral database, glossary of geochemical terminology, and selected biographies are satisfyingly informative. (Quick, what is schmitterite? Uranyl tellurite, of course. Did you know that "oil of vitriol" is a synonym for sulfuric acid?). The program is a beta version. But geochemists will like it even in its diamond-in-the rough form.

Chemtoolbox
This program has two parts: Perhaps 80% of the program is akin to a highly condescended version of the (Chemical Rubber Company) CRC hand book. This part has numerous entries of interest to geochemists. However, it is no match for the CRC, and the reference list does not contain page numbers for the data from each source. I wouldn't feel comfortable referencing Chemtoolbox in a journal article.



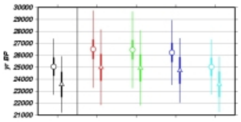
Geochemical Data Toolkit (GCDkit)
A program for analyzing and plotting whole-rock compositions. The text that describes this program is #1 in the Journal of Petrology's download list. Puzzleing. The program calculates several types of normative compositions (e.g. Niggli, CIPW, Barth's catanorm); saturation indices for apatite and zircon; and REE transformations. Here too are statistical tools and common petrologic graphs: AFM diagrams, graphs for determining tectonic setting from trace element compositions, etc. The plots are difficult to edit and not of publication quality. Use GCDkit for a first-pass data overview and to calculate normative compositions. Import the results into a different program for plotting. Excel macros are available for many of the GCDkit plots routines.



Chemical Equation Expert
Balance an equation the old-fashioned way?
1. Balance elements other than O and H. Add coefficients as necessary. 2. Add water to balance oxygen. 3. Balance hydrogen. 4. If the equation doesn't balance or make physical sense, start over.

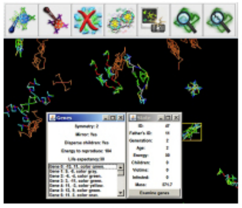
Balance an equation with Chemical Equation Expert?
1. Input products and reactants into boxes. 2. It returns a balanced reaction before you can say Avogadro. It also contains a library of more than 300 canned reactions. If you only know the reactants, you can choose a reaction that yields the desired products.

CRONUS (Cosmic-Ray Produced Nuclide Systematics on Earth Project)
Use these programs to calculate rock exposure ages and surface erosion ages from ^{10}Be and ^{26}Al isotope concentrations in quartz. Concentrations are from measurements of ^{10}Be/^{9}Be and ^{26}Al/^{27}Al by accelerator mass spectrometry. If your work involves these data, you already know about the programs. For the rest of us, it is instructive to go through the documentation and calculate exposure and erosion ages from test data. The plot shows exposure ages for a glacial erratic on Martha's Vineyard. Circles are from ^{10}Be; triangles from ^{26}Al; line lengths are uncertainties; and colors are results from different

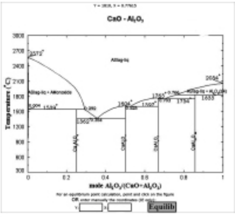


GN139 Content

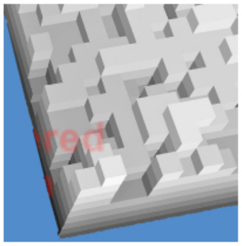
lengths are uncertainties, and colors are results from different assumptions about ^{10}Be and ^{26}Al spallation rates.



Biogenesis
Simulates the evolution of unicellular organisms. It models mutation, genetics, photosynthesis carbon and oxygen cycles, and growth-limiting factors in ecosystems. Organisms are color keyed. Their offspring have the same color but may mutate to better- or maladapted forms. The better-adapted cells reproduce whereas maladapted forms die out. Metabolism and available nutrients are incorpated into the similations. Even better-adapted mutated cells may die before they reproduce if they starve. All in all, the simulation is a great learning tool, and dare we say it, fun.



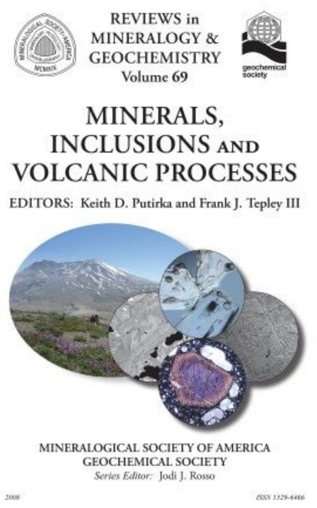
PhaseDiagram Web
The Fact-Web applications are free examples of the huge FactSage thermochemical database and program collection. A complete "Thermodymacs in Geology" course be designed around the Fact-Web. At right is the "PhaseDiagram-Web". First, take in the overall aspect of the diagrams with its cascading peritectics. To a certain type of geek, who shall remain nameless, its beautiful. Now write the reactants, products and reactions at each peritectic and the eutectic. Check your answers by typing a temperature in the lower left box; the program returns he equilibrium phase assemblage. Lucky you; there are 19 more binary phase diagrams to decipher.



LeoCrystal
Simulates crystal growth from first-order thermodynamic principles and Monte Carlo random numbers. Thermal activation reactions define the probabilities that liquids or gases will condense at crystal faces. Comparison of these probabilities to random numbers determines whether new molecules precipitate or dissolve at the topmost crystal layer. User inputs include: the current liquid temperature, the crystal melting temperature, the liquidus temperature, and the fraction of liquid in the crystal-liquid mixture. Graphs and simulated images show changes in the crystal growth rate, roughness of the topmost crystal layer, and changes in total crystal thickness. The program continuously updates and displays enthalpies, frequencies of molecular transitions from non-condensed phases into the crystal, activation energies and other variables. Leocrystal is instructive about high-temperature crystal-liquid systems at the molecular level. In geochemical terms, it helps clarify crystal-liquid interactions at temperatures appropriate for rhyolites through komatiites.

Book Review: RiMG v. 69

Reviews in Mineralogy and Geochemistry, Volume 69, Minerals, Inclusions, and Volcanic Processes edited by Keith Putirka and Frank Tepley III, The Mineralogical Society of America, Washington D.C., 2008, 674 pp. US $40 (ISBN 978-0939950-83-6).



REVIEWS in
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Volume 69

MINERALS,
INCLUSIONS AND
VOLCANIC PROCESSES

EDITORS: Keith D. Putirka and Frank J. Tepley III

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Crystals in volcanic rocks provide valuable records of the history of a magmatic system prior to eruption. Phenocrysts and their inclusions record pre-eruptive conditions, the history of which may be lost when relying upon traditional whole rock analysis. Whereas geochemists were previously forced to deduce the pre-eruptive evolution of a magmatic system knowing only the end result, recent advances in microanalytical techniques reveal detailed information regarding magma storage, evolution, and eruption that was previously hidden from view. The thermodynamic conditions, dissolved volatile contents, and compositional evolution of a magma chamber is written in the crystals as they form; however, the interpretation of this information is complicated by the remarkable heterogeneity that has been observed at small scales (e.g. between zones in a crystal, or between melt inclusions trapped within a single crystal). The Reviews in Mineralogy and Geochemistry volume Minerals, Inclusions, and Volcanic Processes, edited by Keith Putirka and Frank Tepley III brings together a broad spectrum of researchers who are developing theoretical, experimental, analytical, and numerical techniques for extracting the information recorded in these crystals. This collection is more than simply a compilation of current research in the field; it provides detailed "how-to" information for those setting out in this rapidly growing field, where technique is absolutely critical to obtaining useful information. The wealth of knowledge presented in this volume and its corresponding supplementary materials will make it an indispensable resource for anyone working with, or interested in working with, the small-scale clues to magmatic conditions written in crystals and their inclusions.

The volume opens with an experimental view of magma crystallization kinetics by Julia Hammer. This chapter introduces the fundamentals of crystal nucleation and growth as revealed by dynamic experiments, in which pressure and temperature conditions are changed during the course of the run. This is followed by a series of chapters on thermobarometry applications in various systems. A chapter by Keith Putirka covers magmatic thermobarametry using liquid thermometers, crystal-melt thermobarometers, and crystal-crystal thermobarometers, employing primary rock-forming phases such as olivine, pyroxenes, and feldspars. J. Lawford Anderson et al. extend the application of thermobarometry to include minor and accessory phases found in granitic rocks, including zircon, rutile, titanite, and hornblende. Thor Hansteen and Andreas Kluegel discuss high-precision thermobaromtry of fluid inclusions, particularly useful for interpreting crystallization in shallow magmatic systems. A chapter by Jon Blundy and Kathy Cashman describes a variety of methods and approaches for extracting information regarding magmatic conditions from the end-product rocks, extending the discussion from thermobarometric techniques into assessment of crystallization during magma ascent, which leads very nicely into a discussion of magma ascent rates by Malcolm Rutherford in the following chapter. The discussion of magma ascent, crystallization, and degassing presented in these two chapters is particularly relevant to understanding the explosive nature of volcanoes, and is applied in both cases to Mt. St. Helens, among other recent significant volcanic events.

The study of melt inclusions trapped during crystallization of magmatic systems is handled over the next three chapters by Adam Kent, Gordon Moore, and the team of Nicole Métrich and Paul Wallace. The study of melt inclusions has enjoyed much attention in recent years, thanks to the development of new techniques for analyzing and interpreting the pre-eruptive compositional data they encode. In this volume, Adam Kent provides a thorough overview of melt inclusion selection, preparation, and analytical techniques, as well as some of the major pitfalls demanding vigilance on the part of the melt inclusion researcher. He concludes by discussing some features of compositional variation observed in basaltic melt inclusions. Gordon Moore gives an experimental and model-based assessment of volatile (H_2O and CO_2) solubility in melts, providing a basis for interpreting volatile contents and ratios in melt inclusions. Finally, Métrich and Wallace give a thorough overview of volatile analysis in melt inclusions, how volatile contents can be used to interpret degassing histories of basaltic systems, and the relationship between volatile compositions of melt inclusions and the relative explosivities of the eruptions that form them.

The next five chapters deal with compositional variation within and between crystals in igneous systems. Recent technological advances are currently enabling us to examine compositional variability in rocks and minerals on a spatial scale previously unattainable. In the chapter by Frank Ramos and Frank Tepley, the application of microsampling techniques including laser ablation, secondary ionization mass spectrometry, and micro-drilling followed by thermal ionization mass spectrometry are explored in detail, as well as their usefulness for assessing isotopic disequilibrium on the crystal-scale. Oxygen isotope variation among single crystals as well as within a single crystal is discussed by Ilya Bindeman, and uranium series disequilibrium among minerals is expertly evaluated by Kari Cooper and Mary Reid. Both isotope systems provide information on the timescales of magma genesis and residence time, and reveal the presence of xeno- and ante- crysts, in addition to true phenocrysts, in a variety of magma types. A detailed chapter by Fidel Costa, Ralf Dohmen, and Sumit Chakraborty also delves into the timing of magmatic processes, with step-by-step instructions for converting small-scale spatial variability in crystal composition into a timeline of compositional variation in magmatic systems using diffusion modeling. This chapter comes complete with an appendix explaining how to use finite difference methods for numerical modeling of diffusion processes.

The discussion turns to mineral textures in two chapters by Martin Streck and Pietro Armienti. As with many of the previously mentioned methods, quantitative textural analysis of crystalline volcanic rocks provides detailed, time-resolved information regarding pre-eruptive magma chamber conditions, but this information is not easily won. Technique, sample selection, and methods of interpretation are all

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critical to the outcome of textural analysis studies. The Streck chapter describes typical textures observed in common igneous minerals (plagioclase, pyroxene, olivine, amphibole, and accessory minerals) as they pertain to open-system behavior in magma chambers. Armienti discusses the intricacies of generating and interpreting crystal size distributions, including "unfolding" 3D crystal shapes from 2D images, with examples from Stromboli and Mt. Etna. The volume closes with a discussion of magma chamber dynamics as demonstrated by modeling compositional gradients in ignimbrites by Olivier Bachmann and George Bergantz.

The usefulness of this volume extends well beyond what is printed in its pages. In addition to the chapters themselves, extensive supplementary materials are available online at http://www.minsocam.org/MSA/RIM/Rim69.html. These materials include Powerpoint files of the presentations corresponding to each chapter (as they were given at the Mineralogical Society of America Short Course in December 2008) and several downloadable Excel spreadsheets to facilitate the application of the techniques described in the chapters. Also following from the Short Course and Reviews in Mineralogy and Geochemistry Volume is a compilation of teaching materials made available through the On the Cutting Edge program "Teaching Petrology in the 21st Century". These include the on-line tutorials "Teaching Phase Equilibria" and "Geochemical Instrumentation and Analysis", as well as a series of teaching activities on topics including Thermobarometry, Geochemistry of Fluid and Melt Inclusions, Kinetics of Mineral Growth and the Genesis of Mineral Textures, and the Physics of Mineral-Melt Segregation. The teaching materials are available online at http://serc.carleton.edu/NAGTWorkshops/petrology/index.html. Look for Minerals, Inclusions, and Volcanic Processes in the Table of Contents. This Reviews in Mineralogy and Geochemistry volume is exceptional in its timeliness and instructional resources, and is highly recommended for anyone interested in learning about, teaching, or launching a research program pertaining to the detailed magma chamber history that is written into a volcano's crystalline eruptive products.

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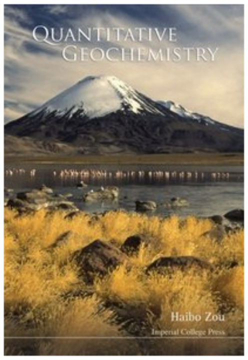
Book Review: Quantitative Geochemistry

Quantitative Geochemistry by Haibou Zhou. Imperial College Press. 2007, 304 pp. ISBN 1860946461



Just as every one of us always has an opinion about the meaning of life, death or politics, it is quite likely that each of us has his or her own unique view of what geochemistry is. This is confirmed by the ever diversifying member-base of the Geochemical Society. So when I was asked to review Haibou Zhou's "Quantitative Geochemistry", I was very intrigued. Zhou's "Quantitative Geochemistry" represents just one specific facet of geochemistry, that is, the use of trace elements or isotopes as tracers in understanding igneous processes involving melting, crystallization or mixing. This is not a book about environmental geochemistry, petrology, kinetics, thermodynamics or physical geochemistry. It is instead an exhaustive compendium of tracer box modeling in the context of magmatic processes. High temperature geochemists are all keenly aware of the basics of tracer box modeling, which are based simply on writing out conservation of mass equations. Of course, given all the possible physical scenarios by which melting or crystallization can occur, analytical representations of mass balance can become quite complicated. Almost every geochemist has spent countless hours writing out and deriving such equations for their specific petrogenetic case study. What "Quantitative Geochemistry" offers is the most complete compendium of analytical solutions to mass balance and transport models relevant to igneous petrology. "Quantitative Geochemistry" is essentially a book of recipes.

Some detailed comments regarding content are in order. In Chapters 1 and 2, Zhou brings together many of the classical mass balance equations (laid out decades ago by such luminaries as Gast, Shaw, and many others) used to model trace element fractionation between melts and their residual solids. Zhou provides the principles of batch and fractional melting, which are concepts that every student of geochemistry is or should be familiar with. Zhou builds on these simple formulations by considering non-modal (eutectic) congruent melting, variable partition coefficients (though there is no discussion about the physical chemistry of element partitioning), and finally incongruent melting, concepts that are often not fully treated in most introductory textbooks. In Chapter 3, Zhou builds on the work of McKenzie on dynamic melting, wherein the rate of melting is limited by porosity. In Chapter 4, Zhou outlines equations for open system melting. Zhou's view of open system melting here is somewhat simplistic, focusing primarily on the case in which the system is simultaneously undergoing melting and being modified by the addition of batch or continuous increments of melts or new solids. There are no discussions of the more realistic, and admittedly more complicated chromatographic and reactive processes. Chapter 5, jumps directly to the use of Uranium series disequilibria in constraining melting rates. Zhou starts off with a brief review of the principles of Uranium series decay and then couples the radioactive decay equations with equations describing melting rates. Chapter 6 discusses trace elements and isotopes in the context of magma mixing and crystallization, laying out the classic assimilation-fractional crystallization equations of DePaolo and more. Chapter 7 is entitled "Inverse geochemical modeling", but what Zhou means here by "inverse" is not the concept of least squares minimization of residuals but rather using trace element concentrations in magmas to extract infer information about melting degree (or melt fraction) or partition coefficients (as opposed to Chapters 1-4 wherein elemental concentrations in magmas are forward-modeled based on assumptions of melting degree and partition coefficients). In my opinion, the first seven chapters are the forte of "Quantitative Geochemistry". I am happy to have this book just for Chapters 1-7 and I think many readers might agree.

I am less excited by the remaining chapters. The remaining chapters seem to be like a mix of good 'ol Texas barbecue and California tofu burgers. Each of these chapters are useful in their own right, but don't expect any rhyme or reason in the choice of themes. For example, chapters 8-9 are primarily technical chapters about error propagation and linear least squares fitting. Although there are clearly many other books that lay out these concepts better, these chapters are still welcome as they provide the reader a primer into these concepts. Chapters 10 and 11 are probably only of interest to mass spectrometrists, and in particular, isotope geochemists. It's not clear to me why these chapters are even in this book. They may, however, still be of some use as they summarize some of the empirical fractionation "laws" used in mass spectrometry. Chapter 11 discusses the concept of "spiking" or isotope dilution; while this is welcome, it would seem that such a chapter would be better relegated to an appendix. Chapter 12 is about the various ways that the decay of U isotopes to Pb can be mathematically presented. This chapter is laid out in the form of FAQs (Frequently Asked Questions by students), i.e., "Section 12.1 Why is the Tera-Wasserburg Concordia diagram concave upward" or "Section 12.2, Why is the conventional Concordia plot concave down?". Once again, Zhou is commended for highlighting basic, but perhaps often overlooked, concepts and features of the U-Pb isotope system, but what is missing is any discussion of the U-Pb system in the context of data or geologic and cosmochemical problems. The discussion of the U-Pb equations comes across more like a fun math game. Chapter 13 is entitled "Geochemical kinetics and dynamics" but is too short and scattered to be of much use. There is a very brief discussion of diffusion, an almost non-existent discussion of advection (despite an individual section allotted to this topic), a brief discussion of bubble growth, and finally, an irrelevant discussion of the projectile motion of a volcanic bomb with a little drag thrown into it. I do, however, like the idea of ending the book with something violent like a volcanic bomb flying through the air.

In summary, this is a book that is well worth your money if what you want is a compilation of mass balance and box model equations for mantle melting. Believe me, it will save you a lot of time in re-deriving all these equations or searching through the literature for published solutions. In this regard, this is a valuable and highly recommended book (Chapters 1-7). The book, however, is not designed to be a textbook for students. It is written very tersely. There is little to no discussion about petrology (phase equilibria or thermodynamics), the physical chemistry of element partitioning, geologic processes, or the physics of melting and melt transport. Although radioactive decay equations are presented in this book, one would do well to have a big picture understanding of isotope geochemistry before examining Zhou's work. There is little discussion about the principles of radioactive decay or their applications to geological problems. Some topics that are missing or minimally discussed and that I thought should have been in a book focused primarily on tracer box modeling include: chromatographic processes, melt-rock reaction, reaction rates and kinetics, and reservoir box modeling. It would also have been good to present equations in more intuitive ways, such as by presenting dimensionless numbers to describe competing processes. These criticisms are certainly not meant to take away from my overall very positive view of this book. At the end of the day, it is hard to put everything into one book.

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book.

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Five questions with Louis Derry

Louis Derry is an Associate Professor in the Department of Earth and Atmospheric Sciences at Cornell University. His research includes biogeochemistry and coupling among climate, tectonics and surface processes. He received a BA in geology from Colorado College in 1981, and a PhD. in Geochemistry from Harvard University in 1990. Geochemical News recently caught up with him via email about the state of his field, the byzantine funding mechanisms of the National Science Foundation, and some near-future research challenges.

1. What is your specialty in the broad field of geochemistry?

I'm interested in lots of things, but most of the ones I actually know something about revolve around low temperature geochemistry and surface processes. I've been interested for a long time in how the Earth's geochemical cycles are coupled to the thermal and tectonic evolution of the Earth, and how all of those are coupled to biological evolution. I have studied these processes on both small spatial and time scales (modern fluxes between rocks, soil, the atmosphere, and plants at the plot scale) to large ones (impact of the Himalaya on geochemical fluxes, or evolution of the Earth's atmospheric O_2 levels).

2. What questions are you trying to answer in your current research?

Currently I am thinking a lot about a few things in particular. We're working on understanding metamorphic CO_2 fluxes produced by the India-Asia collision. This has me really excited because, at least where we've looked so far they turn out to be large relative to the weathering consumption of CO_2. This is an unexpected but fascinating result with potentially far-reaching implications. So we're trying to understand how the fluxes vary along the Himalayan arc, and whether they vary systematically with uplift rate, heat flow or other variables. I'm also working on weathering fluxes from volcanic terranes. We think that they are much more efficient sinks for CO_2 than typical cratonic environments, and also more likely to be sensitive to climate. But one problem is that hydrologic fluxes in volcanic terranes are hard to constrain because there are so many small streams rather than a few big river networks, and because there is so much groundwater transport relative to surface water. So there are just some fundamental things we need to understand and quantify better.

We're also trying a different approach to understanding how biogeochemical cycling in the oceans may have worked in the past. Our views of ocean biogeochemistry are so grounded in our understanding of the modern oceans (necessarily), but that makes it very hard to think through the implications of conditions far from today's. We've been working on simulation tools that we can use to understand the behavior of the coupled CNPOS systems in ancient oceans under quite different conditions than we have today, such as low oxygen, or different circulation regimes, or different mechanisms of biogenic particle export and so on. We felt that we really need to rethink the structure of models for this application. The simple ones that people have been using for these sorts of questions aren't quantitatively adequate, and the "sophisticated" ones based on OGCMs are probably inappropriate for many Deep Time problems. This is work "in silico", but we think will be very helpful in posing and testing new hypotheses about biogeochemical dynamics through time.

3. Please give us a short synopsis of your research approach, the types of samples you collect, your analytical methods, and the types of data you work with.

I'm sure that many geochemists would answer this question the same way. Ideally, I like to do some preliminary pondering, and then move to doing some simple calculations or modeling to get a little bit more formal framework of how I think a system or problem behaves. If that appears promising, then we try and design an "experiment" of some kind, often by sampling rocks, soils, stream waters or what have you, and making a set of measurements. The thought process I have just described sounds vague and tentative - and initially, at least, it is. My experience has often been that very good ideas grow out of intuitive thoughts about a larger problem. The hard part is taking those ideas and refining them so that they can be usefully tested. But the real trouble is taking a perfectly good idea and framing it in the format required to write a successful grant proposal. The collective wisdom of the reviewing and panel communities has settled on a very narrow definition of what makes a good proposal, at least in the U.S. In my view this definition is not wrong, but is simply too restrictive. God forbid you should go to NSF with nothing more than a really good idea that you want to work on! Or, what about a proposal with a really interesting problem that you want to explore but without a section on "expected results". The whole concept of "expected results" in a basic research proposal just depresses me - I refuse on principle to write that. And what if you should forget to specify what plastic your bottles are made out of or some other really vital intellectual tidbit? Well, in that case this is obviously not serious science! So, as you can see, I'm not very optimistic about the current system's ability to support really new approaches. I don't blame the funding agencies for this - to quote Walt Kelley and his comic strip Pogo "we have seen the enemy and they is us". We, as proposers, get feedback of this kind, and too often turn around and apply the same ill-considered standards to other people's proposals. I think we as a community can and should do better. It won't by itself change the amount of money NSF or DOE or USDA or whoever has, but it would improve the intellectual climate and eventually, perhaps even the quality of work that was supported. The idea that NSF should fund "transformative" research is nice, but the reviewing and funding apparatus has to really support that before it can become anything other than pretty words. "Incremental" would better describe the current system.

I'd like to see longer grants to reasonably well-established investigators so that they could spend less time writing proposals and more doing science. In my personal view I'd like to see five year proposals from senior investigators that outlined a set of really interesting questions and an overall approach, without wasting space on minor details. Do some of my colleagues with a decade or two of excellent scientific productivity really still need to convince me that they can do good lab work and know how to analyze basic geochemical data? I would hope not, but as it stands now, they have to include all that or risk being told that they haven't provided enough "specifics". I'm personally willing to make a bet that, if we give people like Bernhard Peuker-Ehrenbrink or Kate Freeman or Joel Blum (just to pick on them - wink!) funding on a five year basis for a clearly articulated set of great questions, we can safely assume that they'll figure out the field and lab protocols necessary to generate very interesting science. Instead, we require mindlessly detailed 2 year time lines, "expected results", and logistical plans that only a bureaucrat could love. I don't think we, as a community, are better off for it.

Interestingly enough, NSF decided to make such 5 year grants available, but to only a few new investigators. That's a nice concept, but in my experience it doesn't work so well. A depressingly large number of young people waste valuable time writing 5 year plans for expensive proposals that are statistically very unlikely to get funded. Not only do they have to give a detailed science plan, but they also have to fit in some sort of revolutionary education plan in the same 15 pages. That's a tall order for

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any PI, but especially a new one - unrealistically so in many cases. In my personal view, it makes more sense to have young PIs focus on shorter time frames. This allows them to really focus on well-defined questions on which they can build, and fits with their promotion time scales. We should instead think about longer-term support for people who have demonstrated long term excellence. Others may disagree, but it's time we as a community talked about this.

4. Can you name three fundamental and as yet unanswered questions in your field?

1. How does the long-term carbon cycle feedback really work? The basic idea that has been around for over a century is that weathering rates change as a function of climate on the long time scale, but the details of how this really works are still not clear. We clearly need a means of stabilizing the Earth's climate on long time scales. The very influential models of the 1980's showed quantitatively that this could work with some simple assumptions. A lot of good work since then has unsurprisingly made it clear that "it's not quite that simple". I think the principles are valid, but we have a lot to learn about the mechanisms.

2. How do major orogenic events (the India Asia collision, the Pan-African orogeny...) impact the carbon cycle? They change weathering rates, degassing rates, and impact the organic carbon sub-cycle in multiple ways. We know that the answer to this question is quite different from what many people thought even 15 years ago. But we're still short of having a good quantitative description of how this works.

3. What is going on with the weird and wonderful changes in climate, evolution and biogeochemistry in the Neoproterozoic and Cambrian? The data and chronology have greatly improved over the last decade, both in quantity and quality and the addition of new geochemical tools like MIF sulfur and metal stable isotopes. But we don't have a very synthetic understanding of how and to what extent all the changes we see are linked mechanistically. With dramatic climate swings, major evolutionary milestones, and large amplitude geochemical variations, there's a lot to explain, and modern analogs only get you so far.

5. We know that you are not a prognosticator, but you do know where the field is heading. What, in your opinion, are three important questions that you and your colleagues will be asking in three years?

In three years we'll probably be asking sort of refined versions of the same kinds of questions. Three years is pretty short, and I've learned the hard way that progress just isn't as fast as I always hope it will be. Being a researcher means being an eternal optimist despite good empirical evidence that we should in fact be pessimists. But, overall, I'm excited to see some of the new geochemical tools starting to move from the "exploration" stage to the "application" stage. For a long time I was made quite uneasy by confident predictions that all sorts of new systems were going to be opened up by new instrumentation such as multi-collector ICP-MS, and that this was going to really revolutionize the field. That has proven to be a more difficult road than I think many people understood, but now more and more good work actually addressing interesting large problems is appearing. One other area in my own field that seems ripe for innovation is the link between the geochemistry of organic compounds and metals. At the moment there is need for improved analytical methods as well as a better process-based understanding of how ligands interact with both major elements like Al in soils as well as very minor ones like Fe in seawater, and how these metal-ligand complexes interact with mineral and bacterial surfaces. These are not easy problems, but I believe there is still a lot more to be learned by diving into them.

The Geochemical Society · 5241 Broad Branch Rd, NW · Washington, DC 20015-1305

Constantly on my mind

"In most fields of discourse [constant] is an antonym of 'variable', but in mathematical parlance a variable may sometimes also be called a constant."
-Wikipedia, the free encyclopedia

Speak constant and enter

By the dictionary definition, a constant is basically an indestructible fact of nature. Constants are supposed to be those absolute--unchangeable figures and numbers that pin down our scientific theories and laws of nature, defining the reliability and accuracy of our view of the world and the nature of its reality. Though, in an absolute scientific sense of fallibility, nothing can be absolute, not even a constant. For all I understand, the very basic principles of scientific inquiry and way of thinking encourage us to yield from strong dogmatic authorities like constants.

Nevertheless, scientific research is most reliable on constants. The National Institute of Standards and Technology provides a listing of recommended values for 326 fundamental physical constants for research use. For the scientific method, it seems, it is convenient to be able to say that there is something that sticks. Constants are essential for parametrization and calculation of fundamental equations defining the basis of most scientific theories. Constants are found so important that they are even given names in the eponymic convention to honor their founders. It is one of the most convenient ways to get your name in the history books; find a species, write a symphony, or come up with a constant of nature.

Was Einstein right?

One of the most famous constants in modern science is not necessarily famous for being important but for the fact that it was found by Albert Einstein. Probably the greatest scientific mind in of the 20th century needed a constant to complement his work, so he just made one up. He was reluctant to use it and could not even tell the precise value of it, but realized it was necessary to make his theory work. This parameter became known as the cosmological constant or the scale factor, whichever you prefer to call it. Einstein himself later called it the biggest mistake of his life. Nowadays the value of this cosmological constant is a matter of great debate and commotion. By telling its precise value, cosmologists could unravel the fundamental structure of the whole universe.

Constantly at the speed of light

Another crucial value of constant nature in Einstein's work was the speed of light. It is a controversial constant, firstly because Einstein's definition of it gave birth to something that is the modern opposite of things absolute, the theory of relativity. Secondly, it has been suggested that there is a slight possibility it is not a constant at all.

Constants of nature also have a peculiar way of being defined by other constants. A practical example can be drawn from the standard definition of the unit of length in the SI system. The unit, meter, was traditionally defined by the French Academy of Sciences as a commonly agreed length marked on a platinum-iridium rod acting as a prototype of a meter. Nevertheless, since 1983 the meter has been defined as the distance light travels in a vacuum in 1/299,792,458 seconds. Here we don't run in to that much of trouble as a meter is not a true constant occurring in nature. It has no physical meaning. It is just a convention that has been defined to unify the practice of measuring distance and length. Right?

A more abstract example of a constant reliant in its definition on the speed of light, from a field more relevant to geochemistry, is the fine structure constant (α). α is defined by three other constants of nature, Dirac's constant ($\hbar$, which is actually also in turn defined by another constant, the Planck's constant h), charge of an electron (e) and by the speed of light in a vacuum(c_0).

$$\alpha = e^2/\hbar c_0$$

Peculiarly enough, some studies of the fine structure constant have concluded that its value has not remained constant through time. This in turn has been taken as an indication that some of the other constants defining α must not be constants. The main suspect to have caused the alleged variation has surprisingly been the speed of light. Especially particular studies on the Oklo natural reactor have shown possible variation in α leading to a healthy debate which at the current moment seems to be in favor of constants still retaining their constant status. But we are left with a doubt, is speed of light constant after all. If it has indeed changed it might be responsible for the suggested change in α, which in turn has an effect on other constants that are dependent on the electromagnetic forces in the nucleus of an atom.

1, 2, 3, decay!

One of these constants is crucial for the particular field of isotope geology - the decay constant (λ). Determination of decay constants for different isotopic systems and improvement of their accuracy have been tasks taken very seriously for they define and justify a field of scientific inquiry which has become very prestigious during the last fifty years. The theoretical potential of very powerful isotopic research tools is restricted by the limited knowledge of the fundamental mechanics behind the phenomena of radioactively decaying elements.

Decay constants are actually more than crucial, they are the solid backbone of modern uniformitarianism itself. Macroscopic geological phenomena can be thought of as not being that absolute by geochemists. Changing rate of change is accepted, everyday compromises are done and quantitative models are constantly modified to better explain observed processes. But decay constants, no way! A constant is a constant. Assuming otherwise would bring down the sound basis of geochronology and a whole field of geoscientific reasoning.

Decay constant under fire...

During the recent decades the advancement in ICP-MS technology has revived the analytics of Lu-Hf isotopes which can be used as a tracer method of petrologic processes in much of the same way as the well-established Sm-Nd method. Particularly the laser ablation in-situ study of zircon, which has the capacity to retain detailed petrologic information about the source characteristics and magmatic evolution of its crystallizing melt has proved to be of great interest to igneous petrologists. The method has encountered also several analytical problems but one of the most fundamental dilemmas of these studies is that the decay constant of Lu-Hf system can't be agreed upon. The rising research activity regarding the isotope system has also revived interest in not just obtaining a reasonably justified value for the decay constant but also questions about methodological legitimacy to answer the set challenges.

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For an isotopic system to deliver reliable and comparable petrologic information, its reference values must be justified.

Several attempts using diverse techniques have been made but the variation between experimental results remains significant and no consensus exists among practitioners on the field. Isochron methods utilized in study of extraterrestrial samples seem to yield consistently higher values for λ_{Lu-176} to ^{176}Hf than the determinations done on terrestrial samples. The range obtained in these studies is far greater than what could be expected on the basis of experimental error estimates and has given a cause for much heated debate.

Some researchers argue for terrestrial samples to be used in determining the λ for further study of other terrestrial samples. Some authorities on the other hand question the age determinations - also reliant on a another set of decay constants - of the terrestrial samples used for cross calibration and prefer the λ values obtained from studies of chondritic meteorites. A different approach of direct counting methods can also be used to back up the terrestrial values but still the coincidence of the results is not good enough for most of the skeptics. Some researchers even utilize the average value for the whole range of determined values in their studies arguing that it is the only truly justified value.

So, the rate of decay of ^{176}Lu to ^{176}Hf is under heavy debate but the research is nevertheless going on. Every published study utilizes its own set value for the decay rate and calls it a constant. Can these numbers really be called constants?

A blind trust to a justified belief is as the value of a constant is to a...?

Constants come in many sizes and shapes, most of them are not that important, as some of them mean (literally) the world to someone. Nailing down any constant and relying on it in one's research can in the end be reduced to a justified belief to invariability of a certain natural phenomenon.

The common thing to all constants of nature seems to be, that none of them are absolute in any other than conventional meaning, being culturally defined as one. How do the constants of nature then differ from the man-created meter? What more are natural constants than mere numbers that are agreed (or disagreed) upon? Constants of nature don't really live up to their name... or do they?

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