

## Geochemical Self-Organization

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Volume 15, Number 4, December 1988

URI: [https://id.erudit.org/iderudit/geocan15\\_4con02](https://id.erudit.org/iderudit/geocan15_4con02)

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### Publisher(s)

The Geological Association of Canada

### ISSN

0315-0941 (print)

1911-4850 (digital)

[Explore this journal](#)

### Cite this article

Fowler, A. D. (1988). Geochemical Self-Organization. *Geoscience Canada*, 15(4), 284–286.

papers dealing with various aspects of metallogeny in France were presented: associated with Paleozoic granites, uranium and Au-U in shear zones.

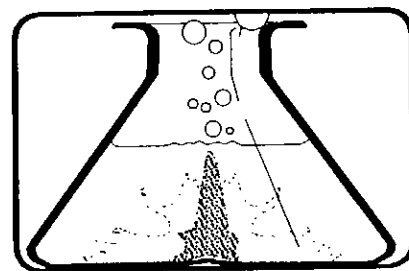
The next session dealt with the application of geophysics to terrane analysis. H. Perroud (France) review Variscan paleomagnetic data and concluded that reliable data for the Paleozoic are far from complete and limitations of the method may not allow distinctions between neighbouring terranes to be made. Other papers in this session applied various types of geophysical data (gravity, magnetic, seismic) to tracing terrane boundaries beneath overstepping sequences both on- and off-shore, the most comprehensive of which was presented by J.P. Lefort (France) to be published shortly as a book.

A whole day was devoted to terranes in the Variscan belt in France. Only in the northern part of the Armorican Massif are the Cadomian (Late Precambrian) terranes preserved without the obscuring effects of the Variscan orogenesis. Here, from NW to SE are (1) a Late Precambrian magmatic arc built upon a 2000 Ma old basement, bordered by (2) a back-arc basin all thrust southward over (3) a continental margin sequence bordering (4) a continental block with (5) a turbidite basin on its southern side. The southern part of the Armorican Massif and all of the Massif Central were involved in strong Variscan orogenesis involving mainly E to W nappe transport and cut by large generally dextral transcurrent shear zones and faults. At least six major nappes have been recognized in the Massif Central (P. Ledru and A. Autran, France) which may correlate with nappes in the southern Armorican Massif (M. Colchen, P. Bouton, P. Meidom and D. Poncet, France; P.L. Guillot and J.P. Floc'h, France; P. Matte, France). The nappes from bottom to top consist of: (a) Early Paleozoic volcanic arc (Thiver-Payzac Unit); (b) ophiolite (Genis Unit); (c) Late Precambrian-(?)Paleozoic miogeocline; (d) Late Precambrian-Early Paleozoic continental rise (Lower Gneiss Unit); (e) Early Paleozoic peri-arc basin (Upper Gneiss Unit); and (f) Carboniferous rift volcanic and sedimentary rocks. Dismembered ophiolites also occur at the base of nappes (d) and (e) (G. Dubuisson, C. Mercier and J. Girardeau, France). D.S. Santallier and others (France) showed that the metamorphism associated with the Variscan Orogeny progressed from high pressure during the Ordovician and Silurian through medium pressure in the Devonian-Carboniferous to low pressure in Late Carboniferous times. While the high-pressure metamorphism is attributed to subduction processes, the latter two are accompanied by widespread plutonism (A. Ploquin, J.M. Stussi, A. Bourguignon and M. Cuney, France; G. Vitel, France; J.L. Bouchez, France). Papers were also presented on correlatives in the Maures and Tanneron Massifs (M. Seno and A. Vauchez, France); in the French Alps (R.P.

Menot, France), in the eastern Alps (F. Neubauer, Austria) and in the Ardennes (F. Meilliez, France, A. Blicq and L. Andre, Belgium). Papers on correlatives in the Bohemian Massif (J. Chaloupsky, Czechoslovakia; G. Hirschmann, W. Germany; P. Matte, France, and P. Rajlich, Czechoslovakia; E. Stein and C. Wagener-Lohse, W. Germany, K. Weber and A. Vollbrecht, W. Germany; W. Franke, J. Schmoll, C. Reichert, G. Dohr and H.J. Durbaum, W. Germany; P. Fluck, J.B. Edel, A. Pique, J.L. Schneider and H. Whitechurch, France), the Schwarzwald and Vosges (G.H. Eisbacher and E. Luschen, W. Germany; A. Krohe, W. Germany), and in the Variscan foreland (W. Brochwicz-Lewinski, Poland; B. Le Gall, France; F. Bernard, B. Doligez, A. Mascle, M. Cazes and T. Rossi, France; C. Bois, O. Gariel, B. Pinet, J.P. Lefort, J.C. Sibuet and M. Cazes, France) were also presented. Other sessions included papers on UK, Eire, Scandinavia, Iberia, Northwest Africa and the Appalachians. Two publications are planned: (1) a book on "The Pre-Mesozoic Terranes in France and Correlative Areas" in English; and (2) a special issue of *Tectonophysics* with a selection of papers from the conference.

One afternoon was devoted to discussion of the various thematic maps being prepared by participants of the project, e.g., terranes, metamorphism, structure, igneous petrology, geophysics. It is a tribute to the linguistic capabilities of French-speaking geologists that the papers at the conference were presented in English which added greatly to dissemination of information around all regions in the circum-Atlantic — congratulations and thanks.

Accepted 6 October 1988.



## Geochemical Self-Organization

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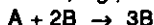
The conference Geochemical Self-Organization was held June 26-July 1, 1988 at The University of California, Santa Barbara campus. The goal of the conference was to bring together scientists working on non-equilibrium, and self-organized geologic systems. This report gives a brief overview of self-organization and focusses on the Canadian participation at the conference.

Self-organization can be defined as the spontaneous transfer of a system from an unpatterned to a patterned state without the intervention of an external template. A familiar example is convection in a fluid, the so-called Benard instability, where at a particular vertical temperature gradient billions of molecules become arranged into coherent macroscopic convection cells, rather than a collection of independent molecules vibrating on a scale of  $10^{-8}$  cm. Geological examples thought to be due to self-organization include disequilibrium textures such as spherulites, layering in certain sedimentary, metamorphic and igneous rocks and ore deposits, zoning in some crystals, the patterns of concretions, dendritic drainage patterns, and frost-heaved patterned ground. What leads to self-organization? Non-equilibrium, feedback, and noise.

Nicolis and Prigogine (1977) show that entropy change ( $dS$ ) can be characterized by two terms, entropy production  $dSp$ , and entropy exchange  $dSe$ . The production term represents entropy produced within the system through irreversible processes and is therefore invariant in sign, whereas entropy exchange can be either positive or negative e.g., a reversal of the direction of heat exchange of a system with its environment. For isolated systems  $dSe = 0$  by definition. In contrast, open systems can in principle have an associated entropy flow such that  $-dSe = dSp > 0$ , and the final state may be more ordered than the initial state (Nicolis and

Prigogine, 1977). This flow gives rise to self-organized or dissipative structures and must occur during non-equilibrium conditions, otherwise  $dSp$  and  $dSe = 0$ . At equilibrium, potential functions such as Gibbs free energy ( $G$ ) can be used to locate the equilibrium, however beyond equilibrium in open systems they can not be used (Coveney, 1988). Prigogine (1967) has described new functions to characterize non-equilibrium open system thermodynamics that relate  $dSp/dt$  to the product of the fluxes (e.g., heat transfer, diffusion) and the thermodynamic force driving the fluxes. Close to equilibrium the fluxes and forces are linearly related resulting in what is termed linear non-equilibrium thermodynamics where the system is characterized by a minimum in entropy production. Further from equilibrium this regime becomes unstable and the system is characterized by non-linear behaviour. Typically this behaviour is shown on so-called bifurcation diagrams (Figure 1).

Feedback provides a source of non-linearity. The simplest form of which is autocatalysis, characterized by reactions in which the presence of the product is required for its own synthesis, e.g.,



Reactions such as this introduce power terms into the rate equations making them non-linear differential equations.

Noise or random fluctuations play an important role in the evolution of systems far from equilibrium. For instance, how does one predict the path that the system at point  $\lambda_c$  (Figure 1) will follow as  $\lambda$  is increased? One can't; in general either evolutionary path is equally probable because random fluctuations cause perturbations to the system and thereby the selection of one path or another.

In addition, the path of a non-equilibrium system beyond the linear branch may become incredibly sensitive to external fields, effectively removing the random element and yielding a single pre-determined path. This is termed "assisted bifurcation" and a familiar example is the self-organized, clockwise rotation of a fluid as it drains a reservoir in the northern hemisphere. Neither the static "equilibrium" fluid prior to draining, nor the fluid under-going laminar flow show the macroscopic effect of the Coriolis acceleration.

Sensitivity to external fields may produce oscillatory behaviour. Prigogine and Stengers (1984) describe an isomerization reaction  $A \rightarrow B$ , where  $A$  also enters into the reaction  $A + \text{light} \rightarrow A^* \rightarrow A + \text{heat}$ . The reverse reaction of  $B$  to  $A$  requires heat and therefore in the presence of light the formation of  $A$  is catalyzed by its own presence. Along the thermodynamic branch the concentration of  $A$  increases linearly with light intensity. Beyond the critical point multiple stable states are possible and fluctuations in the light intensity can cause hops from one state to another. Depending upon the shape of the bifurcation curve of a system hysteresis as well as oscillations may occur (Figure 2).

Far from equilibrium cross-catalytic reactions (a product of reaction #1 is a reactant in simultaneous reaction #2 and produces more of the reactant for reaction #1) may exhibit periodic behaviour (e.g., Prigogine and Stengers, 1984). At equilibrium the reaction reaches a steady state and therefore a constancy in concentration. The addition or removal of reactants or products drives the system from equilibrium and at a critical threshold the concentration of species will oscillate with a definite sustained periodicity

known as the "limit-cycle". This astounding behavior is illustrated by famous reactions such as the "Brusselator" and the "Oregonator". They are truly self-organized and are produced by a coupling of reaction and diffusion kinetics. Winfree (1972) has described a simple recipe for such a self-organized system that can easily be carried out and I highly recommend. This reaction oscillates from red to blue and produces bands that sweep across the system.

There are a few studies in geology that have related growth patterns to self-organization. Amongst these are the work of Haase *et al.* (1980) and Allègre *et al.* (1981) concluding that oscillatory zoning of plagioclase is a result of the coupling of diffusion and kinetics. Boudreau (1984) has suggested that layering in some igneous systems may be due to a feedback-rich mechanism termed "Ostwald Ripening" whereby larger crystals grow at the expense of smaller ones. Similarly Morse and Casey (1988) have shown how the process can explain diagenetic mineral paragenetic sequences that can not be rationalized in terms of equilibrium thermodynamics. Ortoleva (1987) has documented how precipitation dissolution kinetics coupled to transport mechanisms in porous media can lead to feedback and the production of regularly spaced bands. Although definitive models of numerous self-organized systems in geology remain obscure, it is likely that the study of coupled reaction-transport and feedback mechanisms will lead to understanding. It is clear that the study of the kinetics of coupled reaction-transport systems will likely explain many of the systems that have been "mis-behaved" in terms of equilibrium thermodynamics. The following briefly describes the Canadian input to the conference.

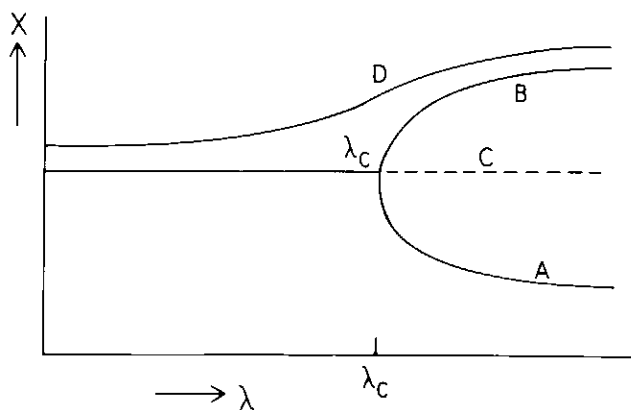


Figure 1 Bifurcation diagram,  $\lambda$  the control parameter, is a measure of the distance from equilibrium (e.g., undercooling in a dynamic crystallization experiment) and  $x$ , a state variable (e.g., crystal composition or less rigorously a quantitative measure of texture) The horizontal solid line is the linear or so-called thermodynamic branch. Beyond  $\lambda_c$ , the bifurcation point, the system has two stable states, and one unstable state (dashed line) for any given value of  $\lambda$ . Curve D schematically represents the path of a similar system, although under conditions of "assisted-bifurcation".

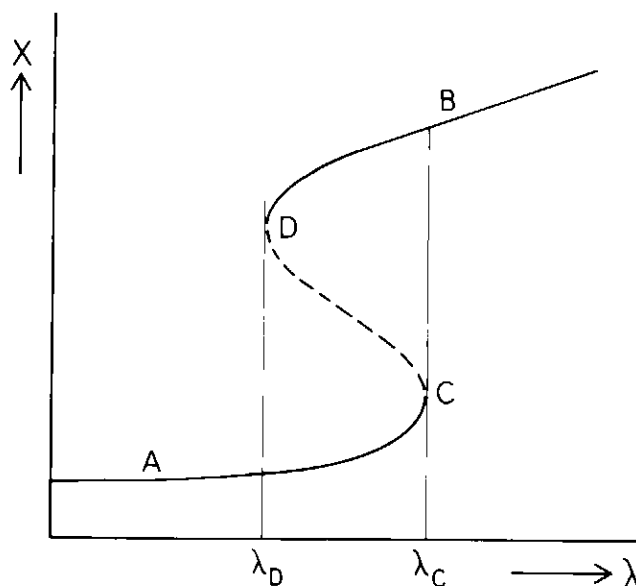


Figure 2 A continued increase in  $\lambda$  along branch A to C results in a hop to branch B. Similarly a decrease in  $\lambda$  along branch B to D results in a hop to branch A. Note the potential for hysteresis between  $\lambda_c$  and  $\lambda_D$ .

John Chadham of the Department of Mathematics and Statistics, McMaster University stressed the importance of non-linear feedback mechanisms in the study of reactive flows in porous media with illustrative examples of ore deposit genesis, the enhancement of reservoir permeability, and waste storage. He discussed some of the modelling techniques available, but cautioned that the mathematics involved in the study of these systems is on the fringe of mathematical analysis. However, the theoretical and computational models that have been developed thus far, allow one to treat these systems in a unified way.

Hans Machel, Department of Geology, Univ. of Alberta, demonstrated that metastable marine, and diagenetic carbonate-solution systems are characterized by bulk solution disequilibrium rather than by equilibrium processes. The data of depth micro-profiles of crystals, the application of diffusion models for crystal dissolution and precipitation, and the calculation of trace element distributions suggest that the recrystallization of these carbonates may result in geochemical patterns indicative of bulk solution disequilibrium. This disequilibrium may be far more common than previously recognized and is inconsistent with existing geochemical diagenetic models, and interpretations of trace element and isotope patterns in marine and diagenetic carbonates. This seriously compromises the concept of cement stratigraphy and other popular geochemical models commonly used in carbonate diagenesis.

Current research on the modelling of disequilibrium textures in igneous rocks by Tony Fowler, Geology, Univ. of Ottawa, Gene Stanley, Physics, Boston University, and Gérard Daccord, Dowell-Schlumberger was presented by Tony Fowler. Many of these textures are self-organized and consist of radiate and or branching arrays that are scale-invariant. Sections of these can be unambiguously quantified as two-dimensional fractal objects. Unlike compact objects whose mass is a function of the cube of the radius, the mass of these objects is a function of a smaller power, the fractal dimension. Among the examples presented a section of a large disequilibrium clinopyroxene that might otherwise be described as ramifying, plumose, branching, harrisitic or Willow-Lake-textured was shown to have a fractal dimension of 1.60. The morphologies of many minerals far from equilibrium are fractal. Experimental work has demonstrated that a variety of morphologies in the olivine and plagioclase systems can be formed by varying the supercooling. The fractal research will allow for a quantitative and precise classification of the textures and lead to a better understanding of experimental results, and more meaningful comparisons of these to natural textures.

Disequilibrium crystal growth simulations performed by modifying the basic Diffusion Limited Aggregation (DLA) algorithm were presented. The analogue between DLA and disequilibrium crystal growth is appropriate as these crystals are grown at high undercoolings where diffusion in the silicate liquid is inhibited and is the rate-limiting and morphology controlling factor. The simulation results are fractal and remarkably similar to the natural images. Feedback occurs as the branches are self-perpetuating in the sense that the probability of a random walker (or structural unit in a silicate melt) travelling down the fiord between the branches is very small. Therefore the growth of the branches is amplified. Modification of the algorithm yields results strikingly similar to, and with similar fractal dimensions to natural crystals, and will help understand the physical processes involved in growth.

Current work on plagioclase zoning by T.H. Pearce and A.M. Kolisnik of the Laser laboratory, Queen's University was reported by Tom Pearce. These new observations using two interference imaging techniques (laser transmission and reflected light Nomarski) indicate the complexity of the oscillatory zoning phenomenon. The plagioclase crystals studied are from glassy basalts, andesites, dacites and rhyolites from Hawaii, Mexico, the High Cascades, Lesser Antilles and the Aegean. Zoning patterns of plagioclase in different rock types appears to be distinctive and easily recognized. In basalts crystals tend to have a normally zoned baseline ornamented by superimposed Type 1 (small An%) oscillations. The Type 1 zones are typically euhedral but some evidence of solution and convolute zoning occurs locally. This zoning is likely due to self-organization in the local system immediately surrounding the crystal. In basalts, it is not uncommon for adjacent crystals to have similar zoning stratigraphies. Plagioclase from andesites typically has incredibly complex zoning patterns and rarely do any two adjacent crystals have a similar zoning stratigraphy. Rounded surfaces clearly due to resorption and discontinuous zoning patterns with abrupt jumps of 10 to 30 An% ("calcic spikes") are typical. These complex patterns are consistent with some sort of mixing event. It is not always clear if the liquids are locally or externally derived (*i.e.*, back-mixed andesite/dacite or basalt). Work continues on this potentially valuable method of interpreting the growth histories of individual crystals in different magmatic regimes.

Paul Williams, Department of Geology, Univ. of New Brunswick, gave a detailed account of observations of mineral differentiation that give rise to such structures as metamorphic layering, pressure shadows, and the larger fold related domains common to many metamorphic rocks. The differentiation is particularly well developed in pro-

grade greenschist facies rocks probably reflecting a dependency on fluid transport. The mineral domains generally show a close correspondence to deformational domains which in turn reflect the heterogeneity of the deformation, and it was suggested that fluid migration is channelized into the more strongly deformed areas in a self-perpetuating process. Solution and precipitation of quartz and carbonate appear to be particularly important and there is evidence that these minerals are commonly transported out of the system rather than simply being reorganized locally. Volume losses as high as 50% have been suggested for large bodies of rock, some regional in scale. Such migration provides a source of cement for overlying sediments.

The conference proceedings are to be published in *Geology Reviews* and a second Geochemical Self-Organization Conference is planned for November 1990. Thanks are due to Edward Hearn for preparing the figures.

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Accepted 14 October 1988.