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Hydrothermal mineralising systems as chemical reactors: Wavelet analysis, multifractals and correlations

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ABSTRACT

Hydrothermal mineralising systems are discussed as large, open flow chemical reactors held far from equilibrium during their life-time by deformation and the influx of heat, fluid and dissolved chemical species. As such they are nonlinear dynamical systems and need to be analysed using the tools that have been developed for such systems. Hydrothermal systems undergo a number of phase transitions during their evolution and this paper focuses on methods for characterising these transitions in a quantitative manner and establishing whether they resemble either abrupt or continuous (critical) phase transitions or whether they have some other kind of nature. Critical phase transitions are characterised by long range correlations for some parameter characteristic of the system, power-law probability distributions, so that there is no characteristic length scale, and a high sensitivity to perturbations. The transitions undergone in mineralised hydrothermal systems are: (i) widespread, non-localised mineral alteration involving exothermic mineral reactions that produce hydrous silicate phases, carbonates and iron-oxides, (ii) strongly localised veining, brecciation and/or stock-work formation, (iii) a series of localised endothermic mineral reactions involving the formation of non-hydrous silicates, sulphides and metals such as gold, (iv) multiple overprinting repetitions of transitions (ii) and (iii). We quantify aspects of these transitions in some gold deposits from the Yilgarn craton of Western Australia using wavelet transforms. This technique is convenient and fast. It enables one to establish if the transition is multifractal (and if so, quantify the multifractal, or singularity, spectrum) and to determine the scale dependence of long range correlations or anti-correlations. Other aspects of the spectrum enable quantitative distinctions between sub-critical, critical and super-critical systems. The availability of long drill holes with detailed chemical analyses and mineral abundances derived from hyperspectral data enables individual ore bodies to be characterised rapidly in a quantitative manner and constraints placed on whether the various transitions are possibly critical or of some other form. We also present some simple nonlinear models, including numerical simulation, self-organised branching and multiplicative cascade processes that produce the multifractal character and correlation scaling relations observed in these data sets. Distinctions between systems that are strongly and weakly mineralised are discussed.

1. Introduction and statement of the problem

The formation of hydrothermal systems, and their associated patterns of rock alteration, veining, brecciation and mineralisation, presents one of the most scientifically challenging parts of the geosciences. These systems are examples of the ultimate in full coupling between mechanical, hydrological, thermal and chemical processes and operate as highly nonlinear entities. An understanding of the processes involved in the formation of these systems is of fundamental economic importance in a world that has a continuing demand for metals in a scientific and industrial environment where discovery rates are declining and the cost of discovery is increasing. For over 100 years scientific investigations of hydrothermal systems have been directed at studies of individual mineral deposits at ever increasing detail but still with no unified view of what controls the location, size and grade of individual deposits. More recently (Occhipinti et al., 2016) the discipline has adopted a systems approach where the mineral deposit is viewed as part of an integrated crustal or lithospheric scale system in which a number of processes associated with fluid production, transport and mixing and metal deposition are considered together. Still the fundamental principles and parameters that define location, size and grade have not emerged in any coherent manner. For the most part, the traditional view of mineralising systems has been grounded in concepts associated with linear system behaviour, a view that is embedded in the conceptually and mechanically unrealistic *source-transport-trap* concept of mineral systems. The outstanding example of linear system behaviour is an assumption that chemical equilibrium thermodynamics controls what we see.

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We present the view that hydrothermal mineralising systems are archetypal, large, nonlinear, non-equilibrium, evolving systems in which a number of interacting parts and processes result in highly irregular spatial and temporal distributions of alteration mineral assemblages, of deformation such as fracturing, veins and breccias, at a number of scales, and of mineralisation and sulphides. The evolution of these systems involves a number of phase transitions such as unaltered → altered, undeformed → brecciated, and un-mineralised → mineralised. These features are some of the hallmarks of systems that are now widely studied under the umbrella of *Nonlinear Dynamical Systems*, a subject that has evolved significantly over the last 120 years since the pioneering works of Poincaré (1892); Gibbs (1902; *Dover rendition*, 1960); Lotka (1910, 1920); Onsager (1944); Prigogine (1955); Lorenz (1963) and Mandelbrot (1974). The literature associated with Nonlinear Dynamical Systems is now too large to adequately review or summarise here and much new material is emerging each month particularly in the fields of non-equilibrium critical systems and pattern formation in very large non-equilibrium systems (Cross and Greenside, 2009).

Hydrothermal mineralised systems are *stochastic* in nature. By this we mean that they exhibit great irregularity in the spatial distributions of alteration mineral assemblages, mineral chemistry, mineralisation, fracturing, veining and brecciation. The term *stochastic* is used to mean that probability distributions need to be used to describe such spatial distributions rather than simple mathematical relations. The basic question we ask is: Are these stochastic distributions the result of chance where the concentration of one mineral or the density of veining or of brecciation is unrelated to a neighbouring one? Or is the apparent irregularity the result of an underlying deterministic process that produces patterns that are apparently stochastic but are intrinsically deterministic? If the latter is true, then spatial correlations in alteration, mineralisation and veining/brecciation should exist and one might expect that large, high grade mineral deposits are characterised by long range correlations. Moreover, processes operating at a range of spatial scales from lithospheric plumbing system scales to the nano-metre chemical reaction scale promote interactions and correlations at an array of spatial scales. Hence one expects the resultant signatures in hydrothermal systems to scale in different ways at different spatial scales and hence to be multifractal in nature.

We consider hydrothermal mineralising systems as large open flow chemical reactors held far from chemical equilibrium by flows of heat, fluids and chemical components. At the same time mechanical disequilibrium is maintained by the flow of momentum, expressed as veining and brecciation. Closed chemical systems must always evolve (Ross, 2008, p4) to an equilibrium state (which is itself a stationary state where no evolution of the system occurs) whereas open flow systems can evolve to one or more non-equilibrium stationary states and remain there, or switch from one non-equilibrium state to another, for as long as the flows are maintained (Ord et al., 2012). These are characteristics of nonlinear dynamical systems.

The tantalising question is: What processes generate a large, high grade mineral deposit as opposed to a small disseminated deposit? In keeping with our view of mineralising systems as chemical reactors we propose that the following conditions need to be met to generate a large high grade deposit:

- Processes and geometries for efficient fluid mixing must exist.
- A plumbing system exists such that as much as possible of the reactive material in the system can be accessed. This means that optimal fluid and heat flow geometries must be developed that do not involve channelling of fluids into a small number of conduits.
- Within the system, intimate access to nutrients must exist.
- Large contact areas between reactants and nutrients must be generated during the fluid mixing process. This means that although the flow might remain laminar a pore or fracture geometry exists that enables chaotic flow to develop (Ottino, 1994; Lester et al., 2012). We emphasise that the term *chaotic* in this sense does not necessarily imply turbulent flow (Hobbs and Ord, 2015, pp 390–393).
- The shape and size of the system has to optimise heat loss so that maximum possible yield results (Aris, 1961).

These are the conditions that a chemical engineer would take into account in designing an efficient high yielding chemical reactor. In addition a chemical engineer would need to optimise the net present value (NPV) of the reactor cost given other constraints imposed upon her. Luckily we do not need to consider such monetary issues but perhaps mother Nature does. Perhaps She (being intrinsically a thermodynamicist) requires that the equivalent of NPV (entropy production) is maximised given the constraints on the system (which may for instance be pre-existing crustal structure)? Or perhaps She requires that the configuration of the plumbing system maximises entropy? These are overarching, fundamental issues regarding hydrothermal systems that need to be addressed in the future.

One way of expressing the criteria for producing a high yielding mineralising reactor is that of Bejan and Lorente (2008) in the form of their *Constructal Law* which they claim is a fundamental law of Nature: *for a finite-size flow system to persist in time* (*to live*) *it must evolve such that it provides greater and greater access to the currents that flow through it.* Although this seems to be an excellent description of the plumbing system that must develop in order to optimise fluid and heat transport in a high yielding mineralising system it does not address some of the other dot points mentioned above. It does however imply that we should be able to observe long range spatial correlations in the patterns of alteration, mineralisation and deformation we see in mineralised systems. We return to the Constructal Law in the discussion.

Another way of tackling the question is to explore the system of mathematical equations that would describe the operation of mineralising systems and see if we can distil from these some useful basic principles that enable us to understand these systems a little better. The operation of a chemical reactor is described by a set of mathematical equations that cover the dot points mentioned above. These equations (see Appendix A) are fully coupled to each other and describe the following processes. By *fully coupled* we mean that each process has a first order feedback on all of the other processes.

- Mass balance.
- Heat transfer.
- Fluid transport including the coupled mechanics of permeability generation and destruction.
- Chemical reaction kinetics including the *chemical* mechanisms of permeability generation and destruction.
- Mechanical deformation including the *mechanical* mechanisms of permeability generation and destruction.

In keeping with a large number of natural nonlinear dynamical systems the solutions to these equations are invariably chaotic (Aris, 1978; Lynch et al., 1982; Gray and Scott, 1994; Burghardt and Berezowski, 1996; Berezowski, 2000; Berezowski, 2014; Berezowski et al., 2000) and because of the very large number of thermodynamic states generated by chaotic behaviour (Beck and Schlögl, 1993) the systems are intrinsically *multifractal*. Just as the concept of *entropy* arises in statistical mechanics (Sethna, 2011) from any physical system because of the very large number of atomic states the concept of a *multifractal singularity spectrum* arises in chaotic systems because of the very large number of chaotic states (Beck and Schlögl, 1993).

A multifractal in our context is a geometrical pattern made up of a number of spatially intertwined fractals each with its own fractal dimension. An additional characteristic feature of such systems is that their evolution is punctuated by a series of phase transitions where the system changes its qualitative behaviour (Sethna, 2011). At these phase transitions long range correlations develop where the system has no intrinsic length scale (in other words, the correlations are