

TOWARDS A NEW PETROLOGY

GIUSEPPE LUCIDO

Istituto di Mineralogia, Petrografia e Geochimica, Università di Palermo, via Archirafi 36, 90123 Palermo, Italy

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Abstract: In the first part of the paper the study of rocks from the late century up to now is briefly outlined. In particular, it is shown that classic petrology is passing through a crisis. In order to resolve this crisis, a completely different type of approach is proposed. In the second part of the paper, by introducing new principles in petrology, it is shown that it is possible to study a rock at the same molecular scale of an aqueous solution, partial melt, or magma from which it originated. Furthermore, because statistical physics builds a bridge between molecular properties and macroscopic phenomena, it is possible to understand the mesoscopic behaviour of outcrops. The route traced out is that of the "reductionism and complexity". In the final part of the paper, the future direction of research is indicated. Special emphasis was placed on the "liquid → solid interface" and on the role of the "colloidal state" during petrogenesis. It is suggested that petrologic systems result from two different effects: one deterministic and the other probabilistic.

Key words: petrology, new frontiers, molecular-scale, complexity, statistical physics.

Introduction

Petrology is a branch of science that deals with the origin, spreading, structure and history of rocks. The object of petrology is to study changes that occur spontaneously in rock masses: liquid magmas which solidify, solid rocks which melt partially or totally, sediments which undergo chemical or physical transformations.

In the late century the study of rocks was carried out by naturalists, who were often satisfied with a brief and superficial examination of the hand-specimen by naked eyes. With time, the study of rocks became more and more extensive until it has grown into a new discipline, essentially having a systematic and descriptive character, the petrography. It's possible to fairly assert that the foundation of petrography as a true science begins about one and a half centuries ago, in consequence of the Sorby's (1856) studies which really showed the advantages one may take of the microscopic examination of thin sections.

As more became known about mineral composition and rock textures, chemically experienced researchers such as Washington (e.g. 1901, 1913, 1922, 1923) began to concentrate on chemical composition of rocks - for many years through routine analysis of a dozen oxide components. Contemporaneously, the great progress of physical chemistry and in particular the thermodynamic study of heterogeneous systems and the precise information obtainable from the phase rule (e.g. Goldschmidt 1911; Bowen 1925) were rapidly carried away and applied to petrography. So it was that petrography originated petrology.

The petrologic researches provided a rich harvest of data and gave a very important contribution to the Earth sciences. They had the majority of the rocky formations as object, ocean and sea bottoms, evidently, included. Except a few tropical and polar regions, these researches were indeed extended to various geologic formations of the Earth. So, today, we may state that classic petrology is passing through a crisis. In order to come out of this crisis "a completely different type of approach" is essential; in other words, it is necessary to introduce new principles in petrology.

New principles in petrology

Molecular size

Petrologists cannot and must not forget that petrology is a geological science. However, even though petrology must address all its efforts to the resolution of geological problems, it is necessary that it no longer starts, so to say, from the outcrop, that is, from a mesoscopic scale to then proceed to the microscopic scale, but it must start from a molecular scale to then arrive at the mesoscopic scale. In this way the range of size to be searched for in a rock has completely changed. From this new viewpoint, in its approach, petrology no longer attends upon, so to say, geology, but it will wonderfully come back to geology later, at the time of checking, verification, elaboration, and correlation. Thus the rocks are always the tools by means of which a petrologist must try to get the first principles to synthesize on a large scale.

Fortunately there is the possibility to accurately define the structure of any rock in terms of knowledge of the site of the atoms making up the sample and of their relative temporal evolution, that is, in terms of space-time correlation functions (Triolo & Triolo 1992). This definition corresponds to the knowledge of the boundary conditions necessary to solve the equations of motion of the system of atoms making up the rock. Considered the great number of atoms of any rock-sample, such a definition of structure could seem utopian if the "statistical mechanics" from one hand and the "Fourier inversion techniques" from the other were not of great help to us. Statistical mechanics enables us to average the behaviour of systems with a great number of atoms, so the above-mentioned definition of structure becomes operative; the technique of the Fourier transform plays the leading role because it lets us link up the statistical mechanics, the structure definition, and the experiment (Triolo & Triolo 1992). The correlation functions are, in fact, the Fourier transforms of the structure functions,

experimentally measurable by scattering experiments of electromagnetic waves or particles, for example, X-rays, photons, electrons, and neutrons. The use of the tool depends on the problem to be solved and on the system under examination.

Principles of statistical physics applied to the study of rocks and their precursive fluids

Basic approaches to study bulk fluids

The study of fluids as precursors of rocks has become more and more important in any petrogenetic process, whatever the nature of the fluid may be, magma, aqueous solution, or partial melt. Firstly, it is therefore necessary to study the nature, structure and behaviour of bulk fluids. The physical and chemical properties of a bulk fluid are now relatively well known, as a number of methods have been developed to study its structure and thermodynamics. These methods can be classified in three main groups: a- hard sphere models, b- Monte-Carlo or molecular dynamics simulations, and c- perturbation theories. As regards the hard sphere models, the basic idea is that the structure is determined largely by the form of the repulsive potential between the molecules. Since all real potentials are strongly repulsive at short separations, the molecules can be considered in first approximation as perfectly hard spheres. Such an approach is presently the only way to obtain an estimate for the interfacial free energy. Differently from this approach, the advantage of the computer experiments is that they apply on well defined (although not necessarily real) systems. In other words, it is possible to vary molecular mass and intermolecular potential, which cannot be modified in a real experiment. The two common techniques for computer simulation are: the molecular dynamics method and the Monte-Carlo method.

The molecular dynamics method (Alder & Wainwright 1957) consists of integrating the coupled differential equations which describe the behaviour of the system. These equations derive from the application to each molecule of the principle of Newtonian mechanics: $f_i = m_i g_i$, where g_i is the acceleration of the molecule labelled i , m_i is its mass, and f_i is the force applied to it by the intermolecular potential of the neighboring molecules (Bonissent 1982). A molecular dynamics simulation is a deterministic process. This is not the case for the Monte-Carlo method (Metropolis et al. 1953), which involves a probabilistic element. According to this method, successive configurations are generated by a random displacement of the molecules, usually one at a time. The configurations are accepted or rejected, depending on the difference in potential energy ΔU_{ij} between the two successive configurations labelled i and j . The new configuration is accepted with the probability $\exp(-\Delta U_{ij}/K_B T)$, where K_B is the Boltzmann constant and T the absolute temperature (Bonissent 1982). This algorithm generates an ensemble of states of the system (Wood & Jacobson 1957), such that the occurrence of a given state i is proportional to its thermodynamic probability $\exp(-U_i/K_B T)$.

Finally, perturbation theories (e.g. Barker & Henderson 1967; Weeks et al. 1971; Abraham & Singh 1977) use the principles of statistical mechanics, without any model considerations, or computer simulation. They take advantage of the relative simplicity of the equations for the hard sphere fluids and of the fact that the realistic intermolecular potentials are sufficiently

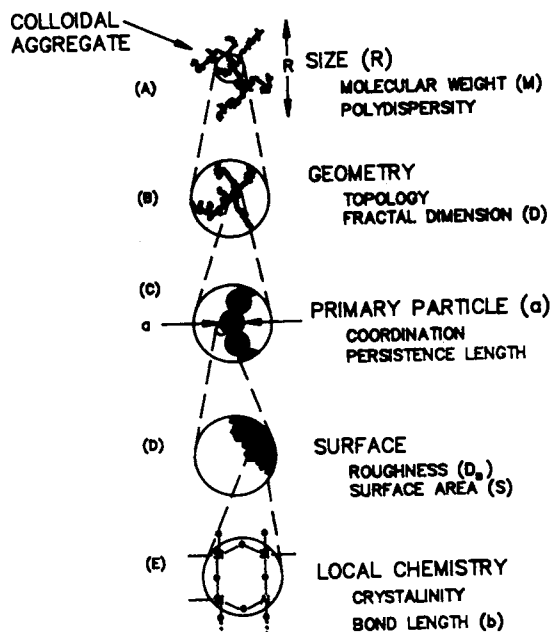


Fig. 1. Structure of a colloidal aggregate viewed on five length scales. Proceeding down the figure one sees finer detail of the structure (Schaefer 1987).

close to the hard sphere potential.

Concluding, each one of the above methods provides information on the structure and related thermodynamic behaviour of a fluid. In particular, we must direct our common efforts towards the liquid \rightarrow solid transition. So doing, we may study a rock at the same molecular scale of an aqueous solution, partial melt, or magma from which it originated.

Fractal approach to study rocks at the molecular scale

It is possible to introduce in the molecular-size range of a rock the fractal concept of Mandelbrot (1977). This author coined the term fractal from the Latin adjective fractus (broken); he introduced this term specifically for temporal or spatial phenomena that exhibit partial correlations over many scales. For the purpose of this paper, a fractal object can be described by two properties. The first one is self-similarity, which means that a detail of the fractal object is structurally identical to the whole, or in other words, that the structure of the object is independent of the characteristic length scale of observation. Though these words are easy to say or write, they imply a property that is very different from anything encountered in everyday life. For example, when a structure can be studied by examining it in a magnifying glass or microscope, the structure cannot be fractal, because the fact that these magnifying devices are useful implies that the magnified structure looks different from that seen by the naked eye. In our case this means that a rock can be considered as a fractal object and be described only within some spatial range. The second property which characterizes the fractal object is its fractal dimension D (Mandelbrot 1977), defined as the exponent of the dimension R in the relation $M = \text{const } R^D$, where M represents the mass. For a rod-like object, the mass scales as the size to the first power ($D = 1$); whereas for

a two-dimensional disk-like object, the mass scales as the second power of the radius ($D = 2$). For a sphere, clearly, the mass scales as the cube of the radius ($D = 3$). For fractal objects this dimension need not be an integer.

Fig. 1 shows, in real space, the structure of a two-dimensional colloidal aggregate (typical of silica spheres at an air/water interface) at five levels of magnification (Schaefer 1987). At the lowest level of magnification, shown in Fig. 1 (A), structural parameters include the size of the ramified irregular object, R , its molecular weight, M , and, if there is a distribution of sizes, the width of this distribution (Martin 1986). If one focuses in on the aggregate by magnifying the center portion as illustrated in Fig. 1 (B), all information about size, molecular weight, and polydispersity is lost. On the first level of magnification, structural parameters of interest concern the geometry and topology of the backbone. In this structural regime the concept of fractal geometry is valuable because, on intermediate length scales, aggregate structures often are self-similar. As before mentioned, self-similar structures are geometrically identical under different degrees of magnification. It should be evident from Fig. 1 (A-C) that self-similarity exists only over a limited range of length scales. The finite size of the object certainly limits self-similarity on large scales. On small scales, self-similarity often gives way to compact structures as indicated in Fig. 1. In going from Fig. 1 (B) to Fig. 1 (C), the self-similar nature of the fractal backbone is lost and one actually resolves the structure of the individual colloidal particles (monomers characterized by a dimension a) which make up the aggregate. On the length scale of Fig. 1 (C), structural parameters include this primary particle size, the coordination number of the particles, and the persistence length which describes the degree of linearity of the arms of the cluster (see Hurd & Schaefer 1985; Dimon et al. 1986). Further detail of the primary particles is revealed in Fig. 1 (D) which actually resolves the surface structure of these particles. In this regime structural information includes the roughness, characterized by a surface fractal dimension D_s and the surface area S (Pfeifer & Avnir 1983; Bale & Schmidt 1984). Obviously if one continues the magnification process, ultimately, the local chemistry of the material is resolved as illustrated in Fig. 1 (E). On this length scale, structural information includes the crystallinity of the object and the details of the local bonding (Schaefer 1987).

In conclusion, fractal geometry very often summarizes and averages over very complex structural details. By applying fractal theory to rocks, already really applied to sedimentary rocks (e.g. Pape et al. 1983; Avnir et al. 1984; Katz & Thompson 1985; Hall et al. 1986; Krohn & Thompson 1986; Wong et al. 1986; Wong 1987; Thompson et al. 1987), it is possible that the surface characteristics (depending on the physical state of matter) and the random distribution of the molecular aggregates may offer important clues about petrogenetic processes (see e.g. Lucido et al. 1988, 1989, 1991).

Future direction of research

Liquid \rightarrow solid interface

Liquid - solid interfaces occur abundantly in petrologic systems. They have been sadly neglected even though these interfaces play an important role in many sedimentary and metamorphic phenomena and particularly in magmatic phenomena. The study of a system composed of a liquid and a solid phase, and an

interface, presents some specific difficulties. One is the fact that the two phases can coexist only at the melting point. In such a system, the problems arise mainly from the presence of the liquid. The structure of the liquid is perturbed at the interface by the solid, and this affects the thermodynamic properties of the system. Fortunately, the applications of the above-methods developed for bulk fluids, a- hard spheres models, b- Monte-Carlo or molecular dynamics simulations, c- perturbation theories, give information concerning the structure and thermodynamics of the liquid - solid interface.

We now consider the case in which the composition and temperature fields evolve by diffusion (Tait & Jaupart 1990). The solidification front can either be flat or very commonly it develops so that a third phase occupying a transition region between a layer of interely solid material and an entirely liquid layer forms. The case of a flat interface is illustrated in Fig. 2 in which the diffusive profiles of composition and temperature developed ahead of the interface are sketched. Note that the compositional boundary layer is much thinner than the thermal boundary layer because of the general fact that the thermal diffusivity in the liquid is much greater than the compositional diffusivity. Fig. 2 also illustrates the idea of constitutional supercooling which can lead to instability of the planar solidification front and formation of an intermediate phase (Tait & Jaupart 1990).

In the following section it will be reported, as an example, the case of magmas, but evidently this new way of thinking may be extended to both metamorphic and sedimentary processes.

Magma liquid immiscibility near the monotectic

In the case of magmas, we must see what the fluid was and what happened to this fluid before it solidified, namely, before

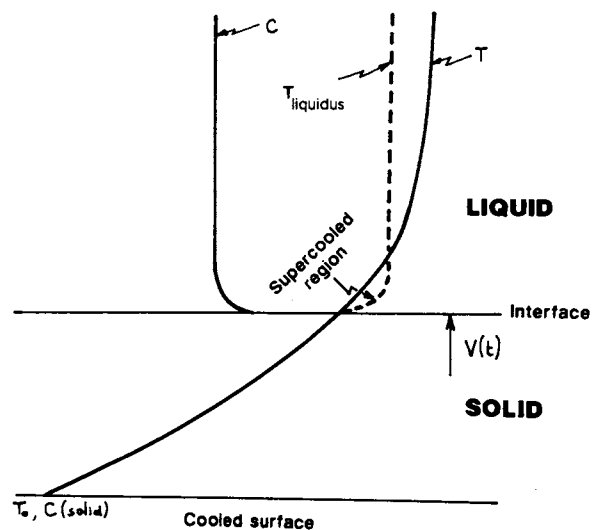


Fig. 2. Illustrating the diffusive profiles of temperature and composition developed during solidification of a binary system with a planar solid/liquid interface. Note that the thermal boundary layer is considerably thicker than the compositional boundary layer. A gradient in the local value of the liquidus temperature is also induced on the same scale as the compositional boundary layer. The phenomenon of constitutional supercooling is shown: the hatched area is such that the liquidus temperature is locally higher than the actual temperature (Tait & Jaupart 1990).

originating the rock. In particular, we must direct our common efforts towards the liquid \rightarrow solid transition. Here we shall refer to the case of a simple fluid or binary system. This is adequate for illustrating many of the basic physical principles involved in the crystallization of multicomponent magmas (see Lucido 1990). In this respect, there is a geologically reasonable range of compositions and temperatures below the monotectic (Fig. 3) where the equilibrium solid cannot form directly from an undercooled single-phase liquid, although such solid is the equilibrium phase (Lucido 1992). Thermodynamics requires a precursor reaction, either a prior liquid-liquid phase separation or a simultaneous formation of the second liquid, if the solid is to form (Cahn 1969). Fig. 3 has been divided into five regions bounded by the liquidus, spinodal, and binodal curve and by the monotectic line. In region I of Fig. 3 solid cannot form from a single-phase liquid, however, after liquid-liquid phase separation it becomes stable. In region II the solid can form before the precursor reaction, but only a small free energy change is available. After the precursor reaction, the liquid-liquid phase separation has increased the free energy change available for solid formation. In region III, solid is metastable in contact with a single-phase liquid and will crystallize from it but will dissolve in the presence of two liquids. In region IV, liquid-liquid phase separation causes a decrease of the free energy available for formation of solid. Finally, region V is bounded by the spinodal. Within it, a single-phase liquid is unstable with respect to continuous separation into two phases. Solid cannot form from a single-phase liquid in this region unless the other liquid forms (Lucido 1992). In conclusion, the combined regions I, II and V of Fig. 3 show the range over which crystallization is prevented or retarded until after a precursor reaction. Although the free energy of the fluid decreases during the precursor reaction, liq-

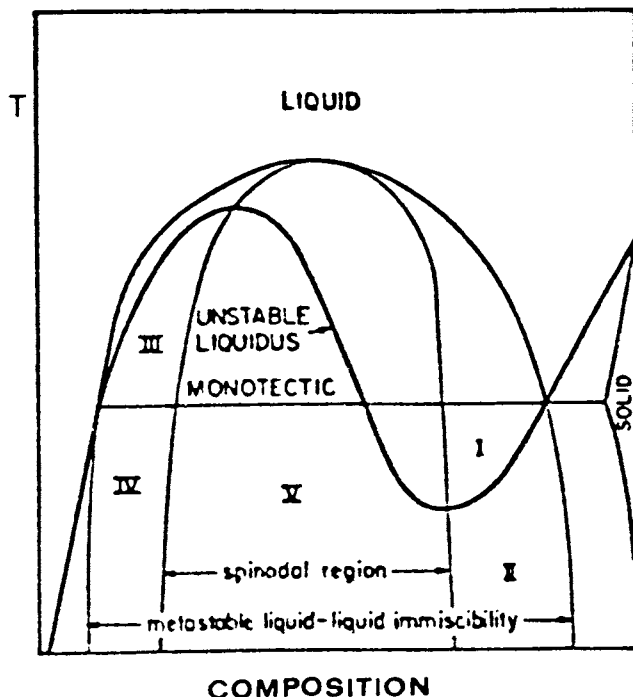


Fig. 3. Composition-temperature diagram illustrating stable, metastable, and unstable equilibria. Three phases, two liquids and one solid, appear near the monotectic line. The liquidus is a single continuous curve with metastable and unstable portions (after Cahn 1969).

uid-liquid immiscibility makes possible reactions that were not possible before (see Fig. 4). We think that this behaviour is a very important factor in the crystallization of magmas and in their fractionation trends; it is still a virtually unexplored research-field (see e.g. Lucido 1987, 1989, 1990, 1992).

Formation of colloids

One of the objectives of this paper is to draw attention to one factor which has been neglected and which might play an important part in helping us to understand the history of rocks better: the role of the colloidal state.

Colloid systems may be defined as systems containing at least two components: a- a continuous dispersion medium and b- disperse phase. Freundlich (1926) suggests that they may be divided into two classes, called lyophilic (solvent loving) and lyophobic (solvent fearing), respectively. The lyophilic colloid solution is thermodynamically stable since there is a reduction in the Gibbs free energy when the "solute" is dispersed. The strong interaction between "solute" and solvent usually supplies sufficient energy to break up the disperse phase ($\Delta H < 0$) and there is often an increase in entropy as well. For the lyophobic colloid, the Gibbs free energy increases when the disperse phase is distributed through the dispersion medium so that it is a minimum when the disperse phase remains in the form of a single lump. A lyophobic colloid can, therefore, only be dispersed if its surface is treated in some way that causes a strong repulsion between the particles. In this way the particles can be prevented from aggregating (or coagulating) for long periods, although it must be emphasized that they are still thermodynamically unstable and the barrier to coagulation is merely a kinetic one

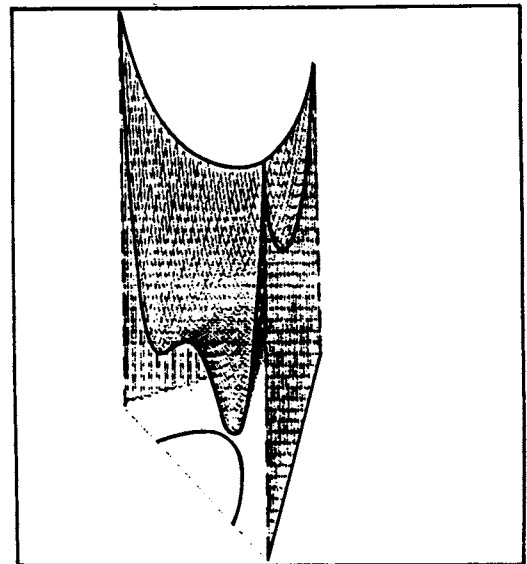


Fig. 4. Hypothetical behaviour of free energy for a three-component system, at different temperatures, and phase diagram in the concentration space. This illustrates the rich possibilities of immiscibility and formation of supramolecular structures in many-components systems (Palma Vittorelli 1989).

(Hunter 1987). Given geological time they will ultimately form an aggregate!

Barring a few notable exceptions, the colloidal state of matter has been considered by soil chemists and clay mineralogists only. Roth (1983), taking into account the petrogenetic role of coagulates $\text{SiO}_2/\text{Al}_2\text{O}_3$ and sorbates (K, Na, Ca, Mg and Fe), indicated the possibility of explaining pre-crystallization phase of magmatites, volcanics and some metamorphites in terms of colloids. Colloid formation during magmatic and metamorphic processes in the Earth has not been previously reported in the literature, with the exception of the Sicilian basaltic magma (Lucido & Triolo 1989). While studying phase separation phenomena in the basaltic magma of Sicily, Lucido & Triolo (1989) found that at the liquid \rightarrow solid interface, near the metastable liquid immiscibility region, colloid formation occurred. Furthermore, very recently, considering magma as a simple and homogeneous fluid characterized by density fluctuations in critical conditions, Lucido (1993) proposed a new theory of the origin of the Earth's continental crust. According to this theory, the formation of the sialic crust of the Earth involved a colloidal process ensuing from the spontaneous emulsification of the "truly primitive" basic magma.

Concluding, over the past fifty years or so, colloid science has undergone something of a revolution, transforming itself from little more than a collection of qualitative observations of the macroscopic behaviour of some complex systems into a discipline with a solid theoretical foundation. It can now boast a set of concepts which, if used judiciously, can go a long way towards providing an understanding of the many interesting behaviour patterns exhibited by rocks and their precursors.

Conclusions

By introducing molecular size in petrology, and by considering that statistical physics builds a bridge between submicroscopic (molecular) properties and macroscopic phenomena, the following interesting conclusions can be drawn for petrologic systems.

1- With the aim of verifying the existence of a universal scaling law, it is possible to introduce in the igneous, metamorphic and sedimentary rocks a new concept of structure in terms of space-time correlation functions.

2- Starting from the principles of statistical physics and chemistry of submicroscopic (molecular) systems, it is possible to understand the mesoscopic behaviour of an outcrop and also to understand the relationships with the neighbouring rocky complexes.

3- By computer simulation, it is possible to calculate the exchanged energies in the processes of formation and/or destruction of molecular structures and in the processes of immiscibility of complex fluids (see Fig. 4). In particular, it is possible to get information about liquid immiscibility to which undergo magmas as a function of pressure and temperature.

4- Liquid \rightarrow solid interface has been sadly neglected, even though this interface plays an important role in many magmatic, metamorphic and sedimentary processes. Fortunately, the applications of the methods developed for bulk liquids give information concerning its structure and thermodynamics.

5- The great and rapid progress of physical chemistry of colloids (already partially applied to sedimentary rocks) cannot be without a future line of development. It must be used and applied to petrology; in this way it will fill a "very important gap"

existing in scientific literature. Colloid formation is a field with a bright future and unlimited potential.

From the foregoing, it appears evident that "the course traced out is that of reductionism and complexity". That is, it is a question of going back to the molecular interactions the petrologic phenomena and of applying to the structures of rocks the statistical mechanics of the disordered systems. The final check point will be the observation of the system at mesoscopic scale and the comparison with the prediction of theory. In practice, it is necessary to realize that the systems of petrologic interest are the result of two different effects: one deterministic and the other probabilistic. The global behaviour results from the relative effect of one behaviour on the other.

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