FRACTALITY AS A WORKING TOOL FOR PETROLOGY: SMALL-ANGLE NEUTRON SCATTERING EXPERIMENTS TO DETECT CRITICAL BEHAVIOUR OF MAGMA

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Abstract: Viewing magma as a natural critical fluid decomposing spinodally, it is possible to consider a magmatic rock as a system formed by one phase embedded in a background of the other phase. Furthermore, because critical phenomenology predicts fractal structures in magmatic rocks, the possibility of inquiring into magma behaviour on cooling it is offered. Small angle neutron scattering experiments have been carried out on several magmatic rocks from different localities. The results obtained, if on the one hand indicate the utility of the fractal concept in studying very complex structural details in magmatic rocks, on the other hand, confirm the validity of the small angle neutron scattering technique, for this kind of study. In particular, the analysis of the scattering data excludes the possibility that, at the molecular level, our volcanic and plutonic rocks are fractal volumes; no inference is possible about the particle-matrix interface. In addition, comparing between themselves the experimental results of the plutonic rocks with those of the volcanic ones, it is possible to notice a quite similar behaviour for all samples. This fact, clearly suggests that magma behaviour on cooling occurs by means of a unique and iterative mechanism. This is consistent with the hypothesis that the scattering originates from droplets with similar surface fractal dimension and cooled at or near critical condition by spinodal mechanism.

Key words: magma, fractals, neutron scattering, spinodal decomposition.

Introduction

In recent years much attention has been paid to fractal objects. Mandelbrot (1977) has introduced the term "fractal" specifically for temporal or spatial phenomena that exhibit partial correlations over many scales. Fractal theory covers by now a long list of different subjects ranging from simple geography to the study of mathematical models. A few examples are landscapes (Mandelbrot, 1982), percolation on fractal lattices (Havlin et al., 1983), turbulence (Hentschel and Procaccia, 1983), rings of Saturn (Avron and Simon, 1981), and polymer models (Havlin and Ben-Avraham, 1982; Magid et al., 1983).

Self-similarity and Hausdorff dimension

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 characteristics length scale of observation. This means that, in the real world, an object can be considered as a fractal object and be described by a fractal dimension only within some spatial range (domain). The fractal structure is easily observed by small angle neutron scattering (SANS) if self similarity holds in some spatial range below 1000 A.

The second property which characterizes the fractal object is its fractal or Hausdorff dimension, D (Mandelbrot, 1977), defined as the exponent of the dimension R in the relation $M \propto R^{-D}$, where M represents the mass. For a linear fractal function, the Hausdorff dimension D may vary between 1 (completely differentiable) and 2 (so rough and irregular that it effectively takes up the whole of a two-dimensional topological space). For surfaces, the corresponding range for D lies between 2 (absolutely smooth) and 3 (infinitely crumped). In particular, for spatial dimension equal to 3, a mass fractal can be called a volume fractal, and a boundary fractal can be called a surface fractal. This fractal dimension very often summarizes and averages over very complex structural details.

Fractals in the Earth sciences

The application of the fractal theory to the Earth's sciences is currently an intense field of research (e. g. Mandelbrot, 1975; Burrough, 1981; Bale and Schmidt, 1984; Brown and

Scholz, 1985; Turcotte, 1986; Plotnick, 1986). In particular, the importance of the fractal properties in many sedimentary rocks has been acknowledged by many scientists (e. g. Pape et al., 1983; Avnir et al., 1984; Katz and Thompson, 1985; Hall et al., 1986; Krohn and Thompson, 1986; Wong et al., 1986; Wong, 1987; Thompson et al., 1987). In igneous petrology, however, such a field of research is prenatal. In magmatic rocks the study of fractals is useful if some physical property of a rock can be related to the fractal dimension. This is, for example, the case for magmatic rocks originated from a critical fluid, where the critical behaviour predicts a fractal nature.

Fractal nature of critical phenomena

Critical fluids

The point on a temperature-pressure diagram that describes the physical conditions under which solid, liquid, and gaseous phases can coexist at equilibrium in a single physical system is the so-called "critical point". In 1967 Fisher proposed that, in the neighborhood of a critical point, a condensing droplet's area and volume are related by a formula equivalent to $area^{1/D} = volume^{1/3}$. To illustrate this relation, he considered the condensation of vapor into liquid. A gas consists of isolated molecules well separated from one another, except for occasional clusters which are bound together more-or-less tightly by the attractive forces. Clusters of different sizes are in mutual statistical equilibrium, associating and disassociating, but even fairly large clusters resembling "droplets" of liquid have a small chance of occurring (Mandelbrot, 1982). For a large enough cluster, the surface area is fairly well defined. The surface of a cluster gives it stability. If the temperature now is lowered, it becomes advantageous for clusters to combine to form droplets and for droplets to aggregate, thereby reducing the total surface area and hence lowering the total energy (Mandelbrot, 1982). If conditions are favorable, the droplets grow rapidly. A macroscopic droplet's presence indicates that condensation has taken place! Fisher (1967) evaluates D analytically without concern for its geometric meaning, but it is unavoidable that the underlying droplet surfaces are fractals of dimension D (e. g. Mandelbrot, 1982; Peliti and Vulpiani, 1985). In particular, the analytic characteristics of a physical system in the neighborhood of a critical point are scaling, therefore governed by power laws.

In conclusion, building up a picture in which the fluid is made up of droplets whose surface is a fractal object of dimension $D > 2$, it is possible to explain some properties of the critical point of fluids. In particular, let us now see what is the behaviour of such a fluid if it is a magmatic fluid.

Petrologic model of magma critical behaviour

Singularity of the critical region

Statistical mechanics studies on phase distributions and textural characters of magmatic rocks give valuable clues as to the possible behaviour of natural magmas. On the basis of modern statistical mechanics approximations and considering peculiar textural characters of some igneous Sicilian rocks, Lucido and Triolo (1983; 1984) outlined the behaviour of

a magma in the proximity of the critical region. These authors describe the structure of magma in terms of probability density functions of the position of the particles (ions and/or non ions) making up the magmatic fluid. For a magmatic fluid very much away from the critical region, the correlation length ξ (in a sense this quantity is the spatial scale of a fluctuation) is of the order of a few molecular diameters. By approaching the critical region ξ will increase up to several hundred molecular diameters and hence the magmatic fluid will actually behave in a peculiar way, in a sense will act like an unstable system with high compressibility and long range correlation between the particles. From a physico-chemical standpoint this amounts to a magmatic fluid in which the behaviour of any portion of the whole fluid is strictly related to the behaviour of any other portion of the fluid itself. This means that by approaching the critical surface liquid phases will behave less and less as liquid and more and more like a mixture of liquid and gas phases, owing to the increase of ξ (Lucido and Triolo, 1984). Let us now see what happens on leaving the critical conditions by cooling the magmatic fluid. The effect of decreasing the temperature is also to decrease the fluctuations characterising the critical behaviour. In particular, the above correlation length will decrease and the magma will behave more like a fluid with short range correlation (liquid).

Liquid immiscibility in critical conditions

Very recently on the basis of the known physico-chemical data on phase transformations and taking into account the thermodynamic behaviour of a fluid near the critical region, a new view of magma is proposed (Lucido, 1989). According to this view magma is a natural critical fluid which decomposes by spinodal mechanism. Within the spinodal region the second derivative of the free energy with respect to the concentration $(\delta^2 G/\delta X^2)$ is < 0. Now if the molar free energy curve remains continuous with continuous first derivatives, we have a spinodal in such a magma at all temperatures above the critical consolution temperature. In such a case the two phases in equilibrium are closely related and one phase makes the other by continuous composition changes (Cahn, 1968). Above the monotectic temperature the two-liquid equilibrium and the spinodal have a metastable continuation and thus we have a spinodal reaction leading to two related metastable phases (see Chan and Goldburg, 1987) in a liquidus plus solid portion of the diagram. That is, there is a closely related new phase in the melt corresponding to a liquid in density or nearest neighbor distance. Starting from a magma in the critical condition, as temperature decreases, a second liquid phase at different density initially separates on a submicroscopic scale, and then, coalescing, collects into larger differentiated clusters that are first tiny globules (Lucido, 1989). In other words, in time, as temperature of magma decreases, a continuous liquid fractionation occurs. More specifically, the fluctuations of the spinodal type involving high surface charge ions will be more effectively damped than the others, and so there might be a tendency of the fluid to split into two immiscible liquids, one enriched in high charge ions and the other enriched in low charge ions. That is, the magmatic fluid will tend to separate into two immiscible liquid fractions, one enriched in elements like iron, magnesium, calcium, titanium etc., and the other enriched in low charge density ions and elements having the tendency to

form polymeric networks (Lucido and Triolo, 1984). So, as a consequence, we find droplets of dark (Fe rich) phase dispersed in a clearer phase richer in elements forming framework structures or vice-versa. In confirmation of this, the existence of droplets of one phase embedded in a background of the other phase has been recognized as the source of an essential singularity on the coexistence curve (e. g. Fisher, 1967; Langer, 1967; Domb, 1976; Binder, 1976; Glimm et al., 1976; Bruce and Wallace, 1981). "Hence one can view a magmatic rock as a random two-phase system. The disordered nature of this system suggests that it can be used to study statistical physics and to understand how randomness on a microscopic (molecular) scale can furnish clues as to the underlying mechanisms". With regard to this, microscopic nature is a rich source of experimental evidence of fractal objects: prominent examples are critical fluctuations in phase transitions (e. g. Bak, 1982; Suzuki; 1983; Pfeifer et al., 1984). On this subject, it is important to remember that fractal surfaces themselves may be frozen critical phenomena and that fracture of a magmatic rock can be understood as a critical phenomenon, too (Pfeifer et al., 1984).

In conclusion, by considering the above petrologic model and by analogy with other critical fluids, we should expect to find droplets of magmatic fluid whose "surface" is a fractal object of dimension D. Because neutrons probe all of the hidden interior structure of a magmatic rock, SANS seemed to us a powerful method to test this hypothesis. Very recently, SANS experiments have been carried out on volcanic rocks from different localities (Lucido et al., 1988). The cooling of magmas yields volcanic or plutonic different types of rocks depending on the environmental conditions; in this paper we apply the concept of fractal dimension D to some plutonic rocks by means of small angle neutron scattering. In this way, one of the main purposes of the paper is to compare the results previously published for volcanic rocks with those here presented for plutonic ones.

Experimental

The rock samples were mounted on cylindrical aluminium holders about 1" in diameter. The area of the samples exposed to the neutron beam was about 1 cm^2, while the thickness was such as to allow transmission of about 0.5. Scattering intensities were recorded using the 10-m ORR SANS camera of the National Center for Small Angle Scattering Research at Oak Ridge Research Reactor (ORR) in Oak Ridge (USA) and the 30-m SANS camera of the National Center for Small Angle Scattering Research, at the High Flux Isotope Reactor (HFIR) in Oak Ridge (USA). We used neutrons of 4.75 A wavelength with a sample to detector distance of 4.56 m for the ORR camera and varying between 3 and 19 m for the HFIR camera. Since this geometry gave us a range of momentum transfer between 0.003 A^{-1} and 0.12 A^{-1}, the scale length investigated is between about 50 A and about 1000 A. During measurements the samples were kept at room temperature. Correction for detector background and sensitivity, conversion of scattering patterns to radial averages and calculation of scattering cross sections per unit volume $d \Sigma (k)/d \Omega (cm^{-1})$, were obtained from these averages by calibrations with secondary calibration standards provided by the National Center for Small Angle Scattering Research.

Theory

Before giving the scattering equations for fractals we shall introduce a few generalities describing the experimental method we used; more details can be found elsewhere (Wong, 1985, 1987; Wong et al., 1986; Wong and Bray, 1988; Schaefer et al., 1984; Teixeira, 1988; Triolo et al., 1985).

In a neutron scattering experiment, a monochromatic beam of neutrons of given wavelength, λ, and intensity, 10, impinges upon the samples and is therefore scattered. The incident and the scattered beams are characterized by their wavenumbers. The scattering event is characterized by the momentum transfer k (the difference between the wavenumbers of the scattered and unscattered neutrons), and by the energy transfer (their energy difference). For elastic scattering, the momentum transfer is related to λ and to the scattering angle. 2Θ, by means of the equation $k = 4 \pi sin \Theta / \lambda$. If the scattering objects are noninteracting, monodisperse spheres of radius R, and scattering density ϱ imbedded in a dispersing medium with scattering density ϱ_o, then the scattered intensity as a function of k is given by:

$$I(k) = V^2 (\varrho - \varrho_o)^2 [3 (sin(kR)-kR cos (kR))/k^3 R^3]^2 \quad (1)$$

where V is the volume of the scattering particle.

At large $k R$, equation (1) shows a k^{-4} dependence (Porod law) typical of systems characterized by a sharp interface separating two media with different scattering density (Debye et al., 1957; Porod, 1982). For a diffuse interface, negative deviation from the limiting slope can be found. A different slope can be expected for fractal objects. The analysis of this slope, in certain cases, yields information on the fractal coefficient. For this reason we have applied (Lucido et al., 1988) the scattering equations derived (Wong, 1985, 1987; Wong et al., 1986; Wong and Bray, 1987, 1988) for a two phase model of non-interacting objects having fractal surface or fractal volume. In fact, the above-mentioned petrologic model shows that upon cooling, the magmatic fluid from which the rocks originated will tend to separate into two immiscible liquid fractions, one enriched in high charge density ions, and the other enriched in low charge density ions and elements having the tendency to form polymeric networks. We shall therefore assume that our rocks are made by non-interacting, monodisperse, spherical particles embedded in a uniform matrix. The surface between the particle and the matrix is supposed to be rough; of course, the smooth interface will simply be characterized by a small degree of roughness. The result obtained (Wong, 1985) is equation (2):

$$I(k) = A[1/k^4 + a/k^{3+x}] \quad (2)$$

where A is merely a collection of constants and depends on the dimension, concentration and composition of the dispersed phase, x is a universally positive parameter which characterizes the surface roughness, while a is a function of the roughness of the interface between the particles and the matrix ($a = O$ for smooth surfaces).

For the case of self-similar fractals for which $x = 3-D$, D being the fractal dimension, an entirely equivalent equation has been previously reported by Schaefer et al., (1984). Therefore, in order to get information on the fractal dimension of a scattering object, scattering results must be obtained

in a range of k values. From a plot of $\log I(k)$ vs. $\log k$, a power law exponent can be derived. As we shall see in the next section, such an exponent may be related to the fractal dimension D of the scattering objects.

Discussion

If the scattering intensity of a rock sample follows a k^{-4} dependence, one can conclude that only the first term of equation (2) is important, and hence that $a = O$. Roughness is therefore unimportant in the mentioned length scale. However, $I(k)$ may possibly obey a single power law with exponent other than -4. If the scattering does not originate from the contrast at the interface particle-matrix, but rather from the particle volume, and if the volume is fractal, then $I(k)$ should obey a single power law dependence with exponent > -3 (Schaefer et al., 1984). Wong and coworkers (Wong, 1985, 1987; Wong et al., 1986; Wong and Bray, 1987, 1988) have shown that if $I(k)$ obeys a single power law dependence with exponent < -4 or if a portion of the scattering curve obeys a single power law dependence with exponent < -4, then the surface is a fractal. However, as already said, a diffuse interface may cause small negative deviations from Porod law. We simulated this condition and found that such deviations, even in the most dramatic case, should not be greater than about 0.2 units. In any other case roughness is the dominating factor in determining the scattering intensity. If the scattering particles are interacting monodisperse spheres, a first order correction to equation (2) can be obtained by multiplying it times the structure function of the system (Triolo et al., 1985). For size-polydisperse particles, the analysis of the slope of $\log I(k)$ vs. $\log k$ might still allow us to distinguish whether or not the particles have a fractal volume, but no conclusion will be possible on the interface (Wong, 1985, 1987; Wong et al., 1986; Wong and Bray, 1987, 1988; Martin, 1986). At the moment, there is no treatment for the polydispersity related to a distribution of fractal dimensions within the sample case, unless the polydispersity of fractal dimension is a consequence of the size polydispersity (Martin, 1986).

Comparison between plutonic and volcanic rocks

We first discuss the results obtained for the plutonic rocks here reported. Figs. 1 through 3 show log-log plots for our samples. The numbers shown above each line are simply archive numbers for the plutonic rocks described in Tab. 1. As above 0.07 A^{-1} the scattered intensity becomes of the same order of the background, only the portion of data between 0.003 A^{-1} and 0.07 A^{-1} are shown in the log-log plots. In this k range, the intensity of all our abyssal samples obeys a single power law dependence with the exponents shown in Tab. 1. The exponent of the power law varies between -3.4 and -4.0 for all samples. The values of the power law exponents are such that polydispersity should not be so large as to prevent us from concluding that in no case do the scattering particles have fractal volumes.

Let us now see the results recently published by Lucido et al. (1988) for some volcanic rocks from different localities. Tab. 2 shows power law exponents for the scattering intensity of 18 volcanic rock-samples. In the k range 0.007–0.1 A^{-1} the intensity of all volcanic samples obeys a single power-law dependence. The exponent of the power law varies between
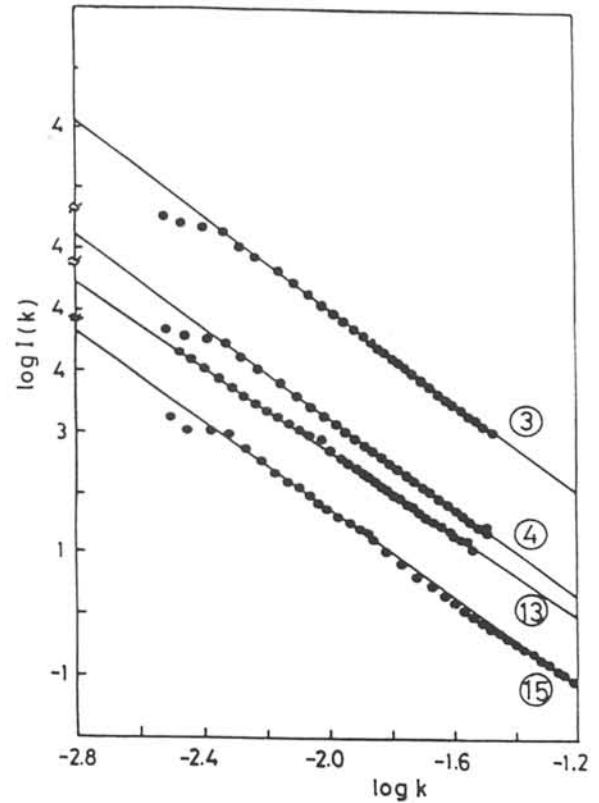

Fig. 1. $log_{10} I(k)$ vs. $log_{10} k$ plots of our plutonic rock samples. Each curve is identified by the archive number of the rock.
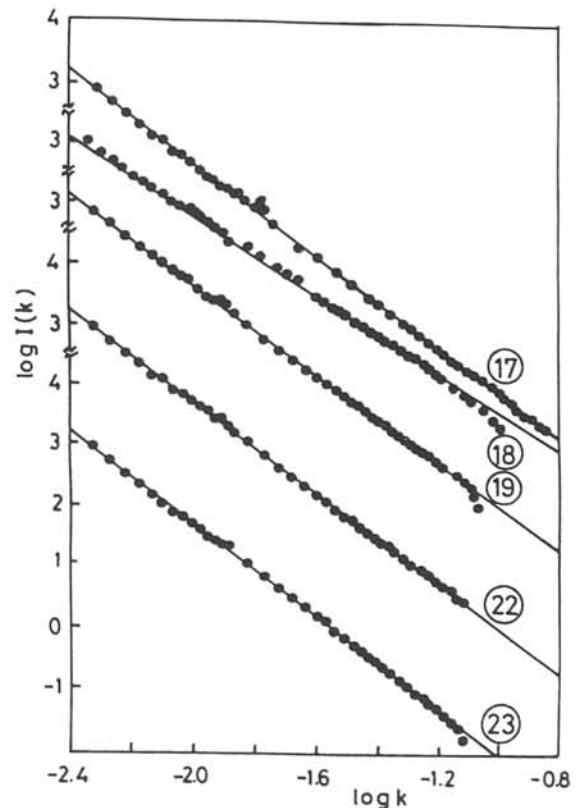

Fig. 2. $log_{10} I(k)$ vs. $log_{10} k$ plots of our plutonic rock samples. Each curve is identified by the archive number of the rock.
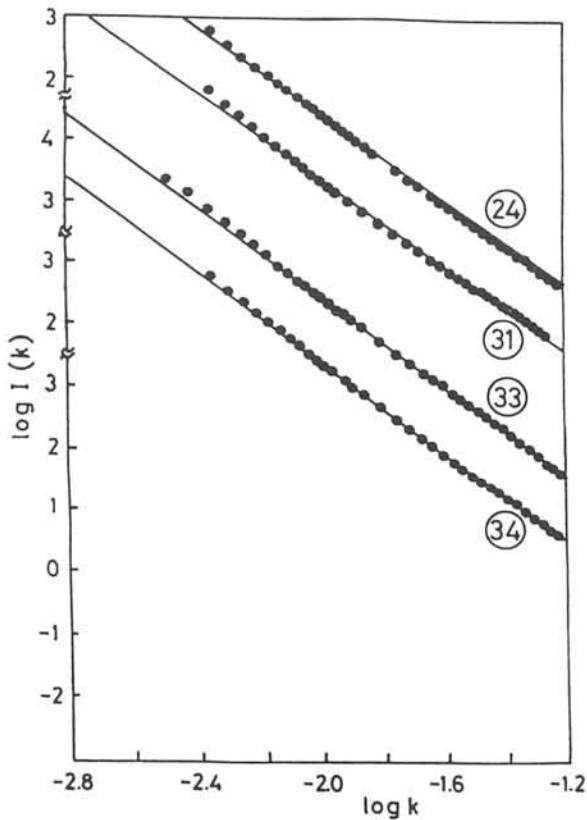
Fig. 3. $log_{10} I(k)$ vs. $log_{10} k$ plots of our plutonic rock samples. Each curve is identified by the archive number of the rock.

Fig. 4. Frequency bar-graph of the power-law exponents (abscissas) of volcanic (light-grey shadow) and plutonic (dark-grey shadow) rocks.

−3.5 and −4.0 for all samples. Now, if we compare the power law exponents of the plutonic rocks (see Tab. 1) with those of the volcanic ones (see Tab. 2), surprisingly no difference can be found! We observe that all magmatic (plutonic and volcanic) samples, fall within a narrow range of values, that is, it is possible to notice a quite similar behaviour. Fig. 4 exhibits histograms that better visualize this similar behaviour. Likewise the plutonic rocks, the values of the power law exponents of the volcanic rocks are such that polydispersity
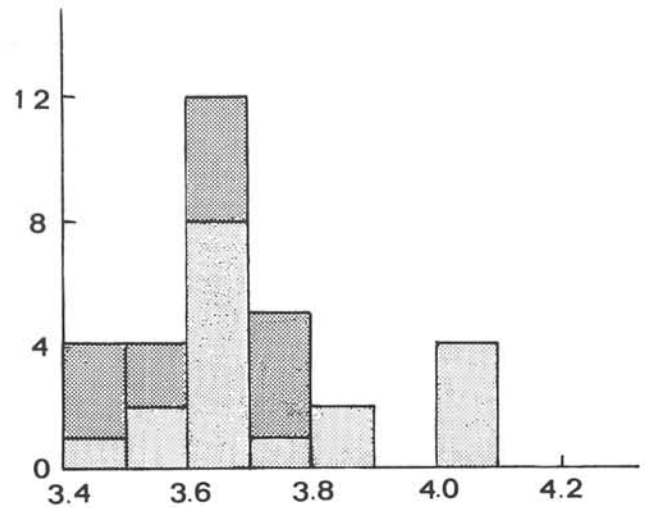
should not be so large as to prevent us from concluding that in no case do the scattering particles have fractal volumes.

If we assume that the particles are monodisperse, then we can conclude that the interface roughness is of dominant importance for all plutonic and volcanic samples. Vice-versa, if we assume that the samples are size polydisperse, then the total interface of each sample, rather than the single particle--matrix interface, could be a fractal (Wong, 1985, 1987; Wong et al., 1986; Wong and Bray 1987, 1988). In both cases, the dependence of $I(k)$ on k would be the same; in this way no inference is possible about the interface. However, although no inference is possible about the particle-matrix interface, the observed power law exponents suggest that there is a "unique and iterative mechanism" which regulates magma behaviour on cooling. This is consistent with the hypothesis that the scattering originates from droplets (produced by density fluctuations) having similar surface fractal dimension, spinodally cooled at or near critical condition. Such a mechan-

Table 1. Power-law exponents for the scattering intensity of the plutonic rocks studied. Sample numbers are archive numbers.

Sample	Rock type	Locality	Power-law exponent
3	Spotted anorthosite	Rustenburg Pt mine, Merensky Reef, Bushveld Complex (South Africa)	−3.77 ± 0.02
4	Giant mottled anorthosite (with intercumulus pyroxene clusters)	Rustenburg Pt mine, Merensky Reef, Bushveld Complex (South Africa)	−3.68 ± 0.01
13	Leucocratic granite (fine-grained rock)	Tichka Massif, South-Western High Atlas (Morocco)	−3.63 ± 0.03
15	Granodiorite with scattered dark spots	Tichka Massif, South-Western High Atlas (Morocco)	−3.65 ± 0.03
17	Dioritic rock, associated with sample 18	Tichka Massif, South-Western High Atlas (Morocco)	−3.71 ± 0.03
18	Gabbroic rock, associated with sample 17	Tichka Massif, South-Western High Atlas (Morocco)	−3.44 ± 0.02
19	Diorite (coarse-grained rock)	Tichka Massif, South-Western High Atlas (Morocco)	−3.64 ± 0.02
22	Amphibole-biotite granodiorite	On the road to Burgos' Forest, Burgos (Sardinia)	−3.78 ± 0.02
23	Leucogranitic rock	Below Burgos village, Burgos (Sardinia)	−3.79 ± 0.02
24	Magmatic rock having „emulsion like" structure	On the road Ozieri-Mores, to the South West of Ozieri (Sardinia)	−3.48 ± 0.01
31	Basic nodules-bearing granite	Scopetu quarry, La Grucitta, Gallura (Sardinia)	−3.45 ± 0.03
33	Granitic rock	Punta Falcone (Sardinia)	−3.60 ± 0.02
34	Gabbroic basic rock	Punta Falcone (Sardinia)	−3.60 ± 0.01

Table 2. Power-law exponents for the scattering intensity of the volcanic rocks studied by Lucido et al. (1988)

Sample	Rock type	Locality	Power-law exponent
1V	Rhyolitic inclusion in soda-trachytic ignimbrite	Cala Cinque Denti, Pantelleria	-4.01 ± 0.03
2V	Soda-rhyolitic ignimbrite	Northern coast of Bagno dell' Acqua, Pantelleria	-3.70 ± 0.05
3V	Soda-rhyolitic ignimbrite	Khartibucale, Pantelleria	-3.72 ± 0.04
4V	Hyalopantellerite	Cuddia del Gadir, Pantelleria	-4.05 ± 0.06
5V	Basalt	Contrada Bue Marino, Pantelleria	-3.81 ± 0.02
6V	Vitrophyric inclusion in pumice	Punta Pozzolana, Pantelleria	-3.65 ± 0.03
7V	Granitoid inclusion in soda-trachytic lava	Cala dell'Alca, Pantelleria	-3.66 ± 0.02
8V	Benmoreitic inclusion in hyaloclastite	M. Levante, Linosa	-3.94 ± 0.05
9V	Gabbroic inclusion in hyaloclastite	M. Levante, Linosa	-3.50 ± 0.03
10V	Benmoreitic inclusion in hyaloclastite	M. Levante, Linosa	-3.63 ± 0.06
11V	Granitoid inclusion in hyaloclastite	M. Levante, Linosa	-3.56 ± 0.03
12V	Granitoid inclusion in hyaloclastite	M. Levante, Linosa	-3.65 ± 0.04
13V	Granitoid inclusion in basaltic rock	M. Vulcano, Linosa	-3.61 ± 0.04
14V	Basaltic rock	M. Vulcano, Linosa	-3.64 ± 0.01
15V	Monchiquite	Western Otago, New Zealand	-3.61 ± 0.06
16V	Camptonite	Western Otago, New Zealand	-3.59 ± 0.02
17V	Monchiquite	Western Otago, New Zealand	-4.00 ± 0.05
18V	Basaltic sphere	Unknown; found on the slopes of M. Caputo, a calcareous formation near Palermo	-4.1 ± 0.1

ism can be evidently interrupted in a different way in different physical environments: the difference is mainly due to changes in equilibrium dependent on the rates of diffusion of the component species and on the degree of supersaturation of the magmatic fluid.

Conclusions

By introducing in the molecular domain of a magmatic rock the Mandelbrot concept of fractal dimension by means of SANS, and by considering the new viewpoint of magma such as critical fluid decomposing spinodally, the following interesting conclusions can be drawn for natural magmatic systems.

1 – Fractal concepts are useful in interpreting complex situations, like particle aggregation and frozen critical phenomena, in magmatic rocks.

2 – Small angle neutron scattering is a powerful method for studying fractality in rocky igneous complexes.

3 – The analysis of the scattering curves, based on the fact that the experimental power law exponents lie between -3 and -4, in agreement with our expectation excludes the possibility that the scattering particles of our plutonic and volcanic rocks are fractal volumes.

4 – The exponents in the resulting power law share all characteristics of exponents for "a unique and iterative mechanism". In our experiments, in fact, volcanic and plutonic rocks surprisingly have quite similar behaviour. We think that this similar behaviour is due to the fact that the surface of the droplets of magmatic fluid had a defined and similar fractal dimension D, produced by density fluctuations on cooling. In corroboration of this view, using absorption techniques, Carnaham et al. (1968) found fractal surfaces ($D = 2.88$ and 2.73) for two granitic rocks from Nevada.

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