Properties of a fractal aggregate

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Julien's paper described the physical nature of a fractal aggregate which is a system of solid particles in the process of coalescence. The present paper supplements this description and, in the author's opinion, provides a fuller description of the physical properties of such an object. We shall begin by noting that a fractal aggregate or, as it is frequently called, a fractal cluster is one of several possible types of cluster ¹ with fractal structure. Such a cluster can consist of separate solid particles, which coalesce in accordance with a certain law. Such aggregates are formed in a solution during formation of a gel, i.e., of a cluster consisting of particles that have merged (sol); similar aggregates form in smokes and in fogs, in the course of relaxation of a metal vapor, during formation of films on the surface by condensation from a jet containing aerosols, etc. Thus, a fractal cluster (aggregate) is formed in the course of growth of a cluster when solid particles merge. The physical nature of formation of a fractal cluster governs its structure and properties.

We shall compare a fractal cluster with a percolation cluster, which also has a structure but appears in different physical processes. We shall discuss the simplest model of the structure of a percolation cluster. We shall consider a cubic lattice and link some of the neighboring lattice sites. A percolation cluster consists of such links joining the lattice sites. We shall introduce a probability *p* that the neighboring lattice sites are linked, so that 1-*p* represents the probability that the neighboring sites are not linked.

Regions linked in this way form clusters with a fractal structure. As the probability p increases, the characteristic dimensions of such clusters increase and at some critical value $p = p_c$ an infinite cluster is formed, which is important in transport processes. This infinite cluster has fractal properties, but it differs fundamentally from a fractal cluster formed during growth processes. In the case of a fractal cluster its average density p falls on increase of its size R in accordance with the expression $p \propto R^{D-d}$, where d is the dimensionality of space and D is the fractal cluster of infinite size tends to zero. A percolation cluster can become infinite only if its density exceeds a certain critical value. This funda-

mental difference between the properties of the percolation and fractal clusters reflects the difference between the physical processes which they represent.

One of the main parameters of a system with a fractal structure is its fractal dimensionality D defined by

$$C(\mathbf{r}) = \frac{\langle \rho(\mathbf{r}') \rho(\mathbf{r}' - \mathbf{r}) \rangle}{\langle \rho^2(\mathbf{r}') \rangle} \sim r^{d-D},$$
(1)

where **r** is the coordinate; C(r) is the correlation function; $\rho(\mathbf{r})$ is the density of matter; *d* is the dimensionality of space; the averaging is carried out in the coordinate space. The main methods for determination of the fractal dimensionality of a cluster follow from the above expression.

Mathematical models are available for the description of possible physical situations in the aggregation of particles into a cluster. Table I gives the values of the fractal dimensionality of clusters formed as a result of different cluster growth mechanisms. These mechanisms can be distinguished on the basis of the following parameters. Firstly, a cluster may grow by attachment of single particles (clusterparticle aggregation) or it may form by joining of two clusters, which in turn are the results of aggregation of clusters of smaller dimensions (cluster-cluster aggregation). Secondly, coalescing particles or clusters may move in different ways in space (they may be exhibiting Brownian or rectilinear motion) and this is reflected in the compactness of the cluster. Thirdly, the compactness of a cluster depends on the probability coalescence of particles in contact. The lower this probability the deeper the penetration of particles into a cluster or of a cluster into a cluster, i.e., the more compact is the final cluster. The results given in Table I apply to the three-dimensional case and describe different physical situations. The errors in Table I represent usually the statistical error of the results of different calculations.

The growth of a fractal cluster simulates a number of physical processes and structures. They include breakdown of an insulator, hydrodynamic structures formed on injection of a liquid or a gas under pressure into a more viscous liquid, and structures in front of a wave of crystallization of an amorphous material. The similarity between these processes and structures is reflected in the fact that under cer-

Aggregation model	Fractal dimensionality
 Particle-cluster, rectilinear paths Particle-cluster, Brownian motion Cluster-cluster, rectilinear paths Cluster-cluster, Brownian motion Cluster-cluster, low coalescence probability 	$\begin{array}{c} 3\\2.46\pm0.05\\1.94\pm0.08\\1.77\pm0.03\\2.02\pm0.06\end{array}$

tain conditions they can be described mathematically in the same way.

We can demonstrate this similarity by considering the process of formation of a fractal cluster when single particles become attached and these particles diffuse in space. The process can be described conveniently by the Monte Carlo method specifying random motion of particles attached to a cluster and assuming that these particles reach a cluster consecutively. In the case of a cluster with an infinitely large number of particles we can employ a different description in which a cluster is considered as an entity with a boundary to which a flux of particles is directed. The attachment of these particles causes motion of the boundary of a cluster, i.e., the cluster grows.

Bearing in mind the diffusive nature of the particle motion, we can describe the flux of particles to a cluster by $\mathbf{j} = -\mathcal{D} \nabla N$, where \mathcal{D} is the diffusion coefficient of the particles and N is their density. Under steady-state conditions this gives the Laplace equation for the particle density:

$$\Delta V = 0. \tag{2}$$

This equation should be supplemented by the boundary condition governing the velocity of a boundary v:

$$v = -an \nabla X, \tag{3}$$

where a is a coefficient and **n** is a unit vector directed at rightangles to the boundary.

The boundary condition (3) creates a "broken" boundary. In fact, the smaller the radius of curvature of the boundary, the faster its growth. This gives rise to an instability which accelerates the growth of a boundary as the radii of curvature of its parts decrease, i.e., this process results in the breakup of the scale of the boundary. We obtain a regular pattern by introducing the minimum radius of curvature of the surface, which corresponds to the size of a single particle. In this description the structure of the cluster surface corresponds to that obtained by a different description when the growth of a cluster occurs as a result of attachment of individual particles. The fractal properties of the cluster are the same in both descriptions.

The similarities between the growth of a fractal cluster and the above processes and structures are manifested by the same mathematical description based on Eqs. (2) and (3). However, the physical quantities occurring in these equations are different for each process or structure. In the case of electrical breakdown the relevant quantity is the electric potential, in the case of hydrodynamic structures ("viscous fingers") it is the pressure, whereas in the process of crystallization of an amorphous substance it is the temperature. In view of the different physical nature of these processes and structures, they not only have some similarities but also specific distinguishing characteristics. We shall demonstrate this in the case of hydrodynamic structures.

Hydrodynamic structures ("viscous fingers") appear when a less viscous liquid is injected into a more viscous one under pressure. This process occurs most simply in a Hele-Shaw configuration representing two parallel plates between which there is a viscous liquid and the distance between the plates is many times less than their dimensions. A less viscous liquid is introduced through an aperture in the upper plate and the velocity of its motion is given by



FIG. 1. Structure of a "viscous finger" formed on injection of air into a liquid crystal under various experimental conditions [V. Horvath, J. Kertész, and T. Vicsek, Europhys. Lett. 4, 1133 (1987)].

$$\mathbf{v} = -\frac{b^2}{12\eta} \,\nabla p,\tag{4}$$

where η is the viscosity of the more viscous liquid and p is the pressure. This condition is equivalent to the boundary condition of Eq. (3) and if we add also the equation $\Delta p = 0$, we can see the similarity in the mathematical description of the process of growth of a fractal cluster and the formation of hydrodynamic structures. However, the minimal scale selected in these cases is different and therefore under some conditions a fractal cluster may differ physically from a "viscous finger."

In the course of formation of "viscous fingers" the phenomenon of capillarity is of fundamental importance:

$$N_{\rm cap} = \frac{v\eta}{\sigma} \,. \tag{5}$$

where v is the velocity at the tip of a "finger," η is the viscosity of the more viscous liquid, and σ is the surface tension at the boundary between the liquids. Injection of a less viscous liquid into a more viscous one breaks up the boundary between them as the liquids move. The minimum scale of the boundary can be estimated from

$$\lambda \sim b N_{\rm cap}^{-1/2} \tag{6}$$

if the motion is two-dimensional, i.e., the scale is greater than the distance between the plates $(N_{\rm cap} \ll 1)$. Hence, it follows that depending on the conditions during formation of "viscous fingers," we can expect different types of morphology (Fig. 1). Under certain conditions the structure may be fractal.

We can thus see that there are differences in the nature of a fractal cluster and a hydrodynamic structure of the "viscous finger" type. In the case of a fractal cluster during growth limited by the diffusion of particles, the resultant cluster has a fractal structure and the minimum scale of this structure corresponds to the size of the particles forming it. In the case of a "viscous finger" the minimum scale of the structure is governed by the parameters of the process and the structure of the boundary between the liquids is not always fractal.

It therefore follows that a fractal cluster is a physical object that forms during growth when solid particles coalesce, it has specific physical properties, and bears similarities to a number of physical processes and structures.

¹⁾Usually a cluster is the name for a large number of bound atoms or molecules which retain their individuality within this system. Recently, this term has been extended to systems comprising a large number of bound macroscopic particles.