## Impact of Fractal Geometry on Permittivity and Related Quantities

# Dubravko Risović,\*,† Blaženka Gašparović,‡ and Božena Ćosović‡

Molecular Physics Laboratory and Center for Marine and Environmental Research, Rudjer Bošković Institute, POB 180, HR-10002 Zagreb, Croatia

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A new physical model of relative permittivity and derived quantities of fractal structures, contrary to the current concept, predicts their dependence on the fractal dimension (D) and on the extent (R) of the considered structure. The scaling of considered quantities predicted by our theory is  $\sim R^{D-d}$ , where d is dimension of Euclidean space. Hence, fractality, a feature often found in physical, chemical, and biological systems, influences also basic properties of such systems that, so far, were believed to be material and not structure related. The theory has been experimentally verified by electrochemical measurements of capacitance of adsorbed layers of nonionic surfactant Triton-X-100 and linoleic acid that have fractal structure. The experimental results, substantiating theoretical predictions, are presented, and the influence of relevant parameters is discussed.

#### Introduction

The dielectric features of material, relative permittivity, susceptibility, polarizibility, and related properties such as capacitance play a significant role in various chemical, physical, and biological systems and on scales ranging from microscopic to macroscopic. Among other things, capacitance is linked to the knowledge of the fluctuations of several physical quantities, e.g., voltage and electromagnetic field fluctuations, 1-6 dipole moment,<sup>5-7</sup> pH, and charge,<sup>8</sup> and also to polarizibility and dielectric dispersion of colloidal and polyelectrolite systems.<sup>5-7</sup> Hence, it is crucial for understanding of biological systems that include ionic channels and cell membranes<sup>9-11</sup> but also other systems such as adsorbed and spread films. 12-14 On the other hand, it has been shown that many of these systems are fractal structures or exhibit fractal behavior<sup>15</sup> and can be described with effective fractal dimension D. Therefore, it was worthwhile to examine the possible influence of fractal geometry on fundamental and derived electric properties. In that context, we examine the relative permittivity and specific capacitance of fractal structures and their dependence on geometrical features. The notion of these quantities in a classical (nonfractal) system implies that they are constant and characteristics of the material of the considered system be it physical, chemical, or biological.

Starting from considerations of fundamental material properties, we have developed a general theoretical model that predicts dependence of relative permittivity and consequently  $C_{\rm s}$  of fractal structures (e.g., adsorbed layer, molecular or particle aggregates, porous material, etc.) on structure size and fractal dimension. These theoretical predictions have been experimentally verified by electrochemical measurements of capacitance of adsorbed layers possessing fractal structure, in conjunction with the recently introduced method the for determination of fractal dimension of such layers. This specific testbed was chosen because structural and dynamic properties of adsorbed molecular films are of both fundamental and applied interest in diverse areas. These systems were studied with various tech-

niques, such as optical, <sup>17–20</sup> electrochemical, <sup>21,22</sup> atomic force microscopy, <sup>23,24</sup> etc. Although, most of the adsorption studies deal with the fractality of the surface, <sup>25–28</sup> there is also evidence of a fractality of the adsorbed layer itself. <sup>16,29,30</sup>

The approach used in the study of fractal structures is based on the possibility to describe quantitatively complex objects that are statistically scale-invariant, physical realizations of mathematical fractals that appear the same on all length scales. This property manifests itself as a power-law-scaling ratio that characterizes one or more features of an object or a process carried out near or at the object:

feature 
$$\sim \text{scale}^{\Delta}$$
 (1)

Here "feature" should be considered in the broadest sense (e.g., surface area, scattered light intensity, the rate of reaction, etc.), and the "scale" could be particle size, pore diameter, scattering vector, cross-sectional area of an adsorbate, or layer thickness. The exponent  $\Delta$  is a parameter, which indicates how sensitive the considered feature is to changes in the applied scale. This noninteger exponent  $\Delta$  has the meaning of dimension, which Mandelbrot<sup>15</sup> termed "fractal". Extension of this concept, coming from the recognition that effective geometries of various structures and processes can be described in terms of fractal geometry, results in an effective fractal dimension  $D_{\text{feature}}$  or simply D.

The geometrical properties of material may play significant role in a various processes and reflect themselves on macroscopic level properties. On the other hand, various processes especially self-organizational processes may result in a fractal structures with unusual features on the micro- and macroscopic level. Hence, we had assumed that geometrical features, such as fractality, should reflect itself also in basic material properties, such as dielectric properties.

### **Theory**

To develop relation between basic dielectric properties and fractal geometry we proceed by examining the basic molecular properties of such structures.

<sup>\*</sup> To whom correspondence should be addressed. Phone: +385-1-456-11-22. Fax: +385-1-468-01-12. E-mail: drisovic@rudjer.irb.hr.

<sup>†</sup> Molecular Physics Laboratory.

<sup>‡</sup> Center for Marine and Environmental Research.

The relative permittivity of a medium (dielectric constant),  $\kappa$ , is given by

$$\kappa = 1 + \frac{P}{\epsilon_0 E_{\rm e}} \tag{2}$$

Here, P is polarization (electric dipole moment per unit volume),  $E_{\rm e}$  is the external field, and  $\epsilon_0$  is permittivity of free space ( $\epsilon_0 = 8.854 \times 10^{-12} \text{ F/m}$ ).

For a medium characterized with polarizibility  $\alpha$  and molecular density (number of molecules per unit volume) N, the polarization can be written as<sup>31</sup>

$$P = \frac{N\alpha E_{\rm e}}{1 - \frac{N\alpha}{3\epsilon_0}} \tag{3}$$

Introduction of eq 3 into eq 2 and retaining only the linear term of expansion gives

$$\kappa \approx 1 + \frac{N\alpha}{\epsilon_0} \tag{4}$$

This relation is generally valid (in a week polarizability approximation) and does not imply any structural effects.

Now, if we consider a fractal structure made from molecules, particles, etc. (either in three or two dimensions), then the number of particles, n, within any space of size R is  $n(R) \sim R^D$ , where D is the fractal dimension. The average density of particles (number of particles per unit surface or volume) is given by

$$N \propto n(R)/R^d \tag{5}$$

where d is the dimension of a space containing considered fractal structure.

Hence, for a fractal structure of size, R, the density, N, varies

$$N \propto R^{D-d} \tag{6}$$

For D = d, we obtain the familiar result that the average density is independent of considered size.

Now, using eqs 4 and 6, the dielectric constant of the fractal structure is given by

$$\kappa - 1 \propto R^{D-d} \tag{7}$$

where, the proportionality constant includes also the term  $\alpha/\epsilon_0$ . This relation clearly shows that the relative permittivity is not only a characteristic of the material but for fractal structures also depends on extent and D of the considered structure. Consequently, for a fractal structure, all physical quantities related to the relative permittivity, such as polarization and specific capacity, shall exhibit similar scaling.

To verify this theoretical prediction, we now apply this theory to an electrochemical system. We shall consider the capacitance of adsorbed layers that exhibit fractal properties. Such systems are usually modeled by a parallel plate capacitor. Consider a capacitor that between its plates contains a material characterized by  $\kappa$ . Its capacitance, C, is enhanced in regard to the capacitor without the material (vacuum) whose capacitance is  $C_0$ :

$$C = C_0 \kappa \tag{8}$$

A similar relation holds for the specific capacitance (capacitance per unit area):

$$C_{\rm s} = C_{0\rm s} \kappa \tag{9}$$

If we consider the  $C_s$  of a fractal structure (e.g., fractal layer adsorbed on an electrode surface or a capacitance of a fractal membrane), using eq 7 and including the appropriate proportionality constant, one obtains

$$C_{\rm s} = C_{0\rm s} + KR^{D-d} \tag{10}$$

where  $K = kC_{0s}$   $\alpha/\epsilon_0$ , and k denotes the proportionality constant in eq 6. Insight into the physical meaning of the proportionality constant K can be attained through dimensional analysis of eq 10. For d=2, it follows that  $[C_s]=[C_{0s}]+[K]L^{D-2}$ . The dimension of a quantity is denoted by square bracket. Hence, the dimension of  $[K]L^{D-2}$  should be equal to the dimension of the capacitance, or  $M^{-1}L^{-2}t^2q^2L^{-2}=[K]L^{D-2}$ , where M, L, t, and q are dimensions of mass, length, time, and charge, respectively. Therefore,  $[K]=[C]L^{-D}$  with  $[C]\equiv M^{-1}L^{-2}t^2q^2$  (dimension of capacitance). Hence, K has a dimension of capacitance per unit fractal space ("area") of dimension D and could be called "the specific fractal capacitance".

Hence, for D = d, the specific capacitance of the considered system shall be constant and independent of size. This result complies with the classical notion and definition of specific capacitance. However, for a fractal structure with  $D \neq d$ , the capacitance per unit surface area,  $C_s$ , is size dependent and scales as  $R^{D-d}$ .

Now, we apply this theory to an electrode/electrolyte interface with adsorbed layer on the hanging mercury drop electrode (HMDE). The considered system is spherical; hence, in the exact treatment, the relation for the capacitance of a spherical capacitor should be applied. However, because of different scales of involved dimensions (radii of mercury drop and thickness of the Helmholtz layers), the model of parallel plate capacitor represents a good approximation.

The differential capacitance, C, of the interface, in approximation of parallel plate capacitor, is given by

$$C = \frac{\epsilon_0 \kappa A}{l} \tag{11}$$

where A is area and l is separation of the plates.

The specific capacitance is given by

$$C_{\rm s} = \frac{C}{A} = \frac{\epsilon_0 \kappa}{l} \tag{12}$$

Use of eq 12 instead of eq 9 leads to the modified eq 10 adequate for the considered electrochemical system:

$$C_{\rm s} = \frac{\epsilon_0}{I} + \frac{\alpha}{I} k R^{D-d} \tag{13}$$

Designating  $\epsilon_0/l = C_{0s}$  and  $(\alpha k/l) = \beta$ , a specific equation for  $C_s$  of the fractal interface, an equivalent to the general eq 10, is obtained:

$$C_{s} = C_{0s} + \beta R^{D-d} \tag{14}$$

This equation, derived from basic electrical properties of a fractal structure, is a more detailed and specific equivalent of eq 1, which represents the general principle of fractal analysis approach. This relation clearly shows that for a certain material

the  $C_s$  depends on a structure size and has a power-low dependence on difference of fractal and Euclidean dimension.

In our case, the considered fractal structure is the layer adsorbed on the surface of the mercury drop electrode. The size of adsorbed structure is determined by the size of the electrode surface. If the electrode surface is increased, the size (area) of adsorbed structure is increased. Hence, because the size of considered structure (R in eq 14) is described by the area of the mercury drop electrode surface for our case, eq 14 becomes

$$C_{\rm s} = C_{0\rm s} + \beta \left(\frac{A}{4\pi}\right)^{(D-2)/2}$$
 (15)

Here,  $A = 4\pi r^2$ , is the electrode surface area and r is the radius of mercury drop.

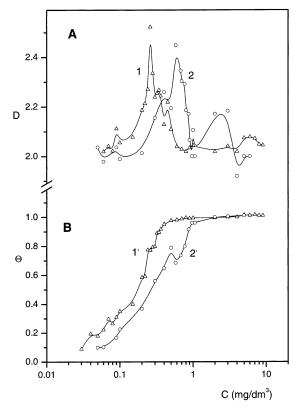
## **Experimental Section**

As a testbed for this theory, the behavior of the specific capacitance of the adsorbed layer of selected organic substances that exhibit fractal structure <sup>16,32</sup> was examined. The adsorption was studied by electrochemical method, and the fractal dimension was determined by using the method of scaling of mercury drop electrode. <sup>16</sup>

To prove the theoretical prediction that the scaling is independent of material (molecular species) or conditions leading to formation of fractal structures, we have included measurements on different molecular species and layers formed under different experimental conditions. So, the capacitance of adsorbed fractal layers of two different substances, nonionic surfactant Triton-X-100 (T-X-100) and linoleic acid (LA), was investigated. Capacity current curves were recorded by phase sensitive alternating current voltammetry (90° out of phase). The  $C_s$  values were calculated from measured capacity currents obtained for each bulk concentration. The measurements were performed at different experimental conditions: mass-transfer controlled adsorption of T-X-100 in a stirred solution and diffusion controlled adsorption of LA, for 1 and 3 min accumulation times, respectively. Different experimental conditions were included in this study because it has been found that morphology of the fractal adsorbed layer is a result of the growth mechanism which is influenced by hydrodynamics (stirring or diffusion) and by the structure of the solution (monomers and dimers).33

The fractal dimension (*D*) was determined from the size scaling of the hanging mercury drop electrode. <sup>16</sup> In this method, *D* is related to the capacitive current that is proportional to the electrode surface area ( $A = 4\pi r^2$ ), described by the electrode radius *r*, through the relation  $i_c \sim A \sim r^D$ . *D* is obtained from the slope of the log-log plot of  $i_c$  vs *r*. The results for fractal dimension determination were elaborated for the electrode potential -0.6 V, representing the approximately nonpolar electrode. For the purpose of fractal analysis, the size of the mercury drop electrode in the voltammetric measurements was varied stepwise over nine sizes corresponding to the electrode surface area in the range of 0.88 to 3.82 mm<sup>2</sup>. A measurement was performed each time with a freshly prepared new drop of a selected size that did not change during the measurement.

T-X-100 (Rohm and Haas, Italy) and LA (Fluka, Switzerland) were used without further purification. Mercury was purified by double distillation under reduced pressure. NaHCO<sub>3</sub> (Merck) was used without prior purification. NaCl (Kemika, Croatia) was purified by prolonged heating at 450 °C. All solutions were prepared with deionized water obtained with the Milly-Q water system (Millipore, Switzerland). A carbonate buffer was used



**Figure 1.** (a) Dependence of adsorbed layer fractal dimension on bulk concentration: curve 1, T-X-100, 1 min stirring ( $\triangle$ ), and curve 2, LA, 3 min diffusion (O); (b) The corresponding total fractional electrode coverage for T-X-100 (curve 1) and LA (curve 2) vs respective bulk concentration. Results are elaborated for the electrode potential E = -0.6 V. Symbols denote measurement points, whereas lines represent the fit with the B-spline.

to maintain pH 8.3. The other experimental conditions and the equipment used were the same as in ref 16.

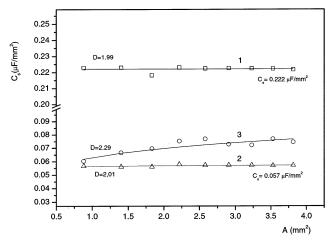
### **Results and Discussion**

The changes of the adsorbed layer fractal dimension with increase of T-X-100 and LA bulk concentrations, corresponding to different fractional electrode coverage, and for different adsorption regimes are shown in Figure 1. The observed variations of D depend on the details of the adsorption process.

In Figure 2, the dependence of  $C_s$  on electrode size is shown for systems with dimension D = d = 2 and for a system with fractal dimension  $D \approx 2.3$ .

We have considered a pure electrode surface and a surface with adsorbed layer with D=2. For such a system, our theory predicts constant, size independent  $C_{\rm s}$ . The measurements in the pure electrolyte (0.55 M NaCl) are included to demonstrate the precision of measurements and agreement with theory of a nonfractal, well-known, and controlled system. For the pure electrolyte, the measured  $D=1.999\pm0.005$ , reflects a smooth electrode surface. The average measured value of  $C_{\rm s}$  is 0.222  $\pm$  0.001 F/m² complying with typical experimental values of such systems. The slight deviation of the measured points from the straight line represent the scatter of the measurement data due to manual adjustment of mercury drop size.

Curve 2 represents  $C_s$  obtained for adsorbed layers of T-X-100 with bulk concentration of 4.0 mg/dm<sup>3</sup>, with  $D \approx 2$ . As we can see, the obtained  $C_s$  value is constant and independent of size. This is in perfect agreement with our theoretical prediction for structures with D=d=2 and also corresponds to the generally accepted concept of size-independent  $C_s$ . The



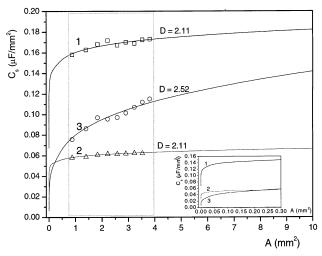
**Figure 2.** Dependence of  $C_s$  on the mercury drop area for the systems with  $D \approx 2.0$  (curves 1 and 2) and for the adsorbed layer with fractal dimensions  $D \neq 2$  (curve 3). Curve 1 ( $\square$ ) represents pure electrolyte, and curve 2 (△) represents adsorbed layer of T-X-100, at bulk concentration 4.0 mg/dm<sup>3</sup>. The average  $C_s$  values are 0.222  $\pm$  0.001 and  $0.057 \pm 0.0005 \,\mu\text{F/mm}^2$ , for electrolyte and T-X-100, respectively. Curve 3 (O) corresponds to linoleic acid, D = 2.29, at a bulk concentration of 0.77 mg/dm<sup>3</sup>. Symbols denote measured values, whereas lines represent corressponding theoretical values calculated from eq 15.

decrease of capacitance of the adsorbed layer in respect to that of the pure electrolyte is a consequence of the adsorption of organic molecules on the electrode that have lower dielectric constant than the electrolyte. Hence, the increase in electrode surface coverage, as described by the corresponding isotherm, results in a decrease of the capacitance in a nonlinear fashion.

Curve 3 represents the measured data and corresponding fit with eq 15 for a system with distinct fractal dimension D =2.29 obtained for the adsorbed layer of LA at bulk concentration  $0.77 \text{ mg/dm}^3$ . In the fit of measured  $C_s$  vs A data, the corresponding experimental D value has been used along with  $C_{0s}$  and  $\beta$  as fit parameters in the least-squares method. In the fit procedure, the values of D and A were also allowed to vary slightly within the limits corresponding to the respective standard deviations. The experimental data (depicted by symbols) are well fitted with the theoretical curve based on eq 15.

Theoretical considerations (eq 15) predict that the specific capacitance  $(C_s)$  of the fractal layer is not constant but changes with structure size (in our case with electrode size, i.e., adsorbed layer area) in a way determined by a fractal dimension regardless of other parameters governing the adsorption. Hence, it is confirmed that, for adsorbed layers with  $D \neq 2$ , the measured values of specific capacitance show size and D dependence as predicted by our theory.

The applied experimental electrochemical method is limited to only 1 order of magnitude in size range, but a lot of other systems that are of interest are on a larger or a much smaller scale of size ranges (e.g., in biophysics, nanomaterials, etc.). Therefore, to predict the behavior of a system in a much broader size range than that measured, one has to extrapolate from the measurement data. In Figure 3, the behavior of the  $C_s$  vs A in measured and extrapolated size ranges is depicted (here A denotes representative size of any considered structure). The boxed area represents the experimentally investigated size range. Here, the  $C_s$ 's of different T-X-100 layers are shown. Curves 1 and 2 represent the fit of measured data with eq 15, for two adsorbed layers that have same  $D \neq 2$ . However, these layers are obtained at the different adsorption stages of the layer formation, corresponding to the T-X-100 bulk concentrations



**Figure 3.** Dependence of  $C_s$  on the structure size and fractal dimension, extrapolated for the extended size range from the measured data of T-X-100 (symbols) fitted with eq 15 (lines). Depicted are curves corresponding to D = 2.11, at bulk concentration 0.09 mg/dm<sup>3</sup> (curve 1), D = 2.11, at 0.5 mg/dm<sup>3</sup> (curve 2), and D = 2.52, at 0.26 mg/dm<sup>3</sup> (curve 3). The boxed area represents the experimentally investigated size range. Inserted is the enlerged small size range comprising A = $0-0.3 \text{ mm}^2$ .

0.09 and 0.5 mg/dm<sup>3</sup>, respectively (cf. Figure 1). Although the D and size range are the same, it can be seen that the curvature of curve 1 is greater than curve 2. This is a consequence of the difference in factors  $\beta$  ( $\beta = \alpha k/l$ ) entering into the expression for the curvature and corresponding to different average layer thickness. Curve 3 corresponds to the fit for the layer with the highest observed D, obtained at the bulk concentration 0.26 mg/ dm<sup>3</sup> T-X-100. It can be seen that for the investigated size range the rate of change of  $C_s$  (curve inclination) is greater for higher D as predicted.

The dynamics of change of  $C_s$  with changes in D is different and depends on a considered size range. As the structure size range decreases, the rate of  $C_s$  change increases. With the increase of the fractal structure size, the changes are more pronounced for the systems with higher fractal dimension.

It is worth noting that the fit of experimental data with eq 15 facilitates the evaluation of the average layer thickness, *l*, from the C-axis intercept. For example, for a T-X-100 layer obtained at a bulk concentration of 0.26 mg/dm<sup>3</sup> with D = 2.52 (curve 3 in Figure 3), the fit to the measured data gave an intercept value of 0.00529  $\mu$ F/mm<sup>2</sup>, wherefrom l = 1.67 nm, what is compatible with T-X-100 molecule size. This fit is very sensitive with regard to the experimental data scatter that results from experimental and measurement errors. Hence, to exactly calculate l, the measurements must be done very precisely, especially with regard to mercury drop size determination, and if possible should be extended to more than 1 order of magnitude.

### **Summary and Conclusions**

The behavior of relative permittivity and consequently specific capacitance of fractal structures with regard to its size and fractal dimension is considered. The theory of relative permittivity of fractal structures is developed. It predicts that relative permittivity (dielectric constant,  $\kappa$ ) is not constant but rather depends on size and fractal dimension of considered structure. As a consequence of this dependence, the specific capacitance,  $C_s$ , of fractal structures is not constant for a given material but exhibits dependence on fractal dimension and size of the considered structure. This theory has been experimentally verified for fractal structures of adsorbed layers of different organic substances (linoleic acid and nonionic surfactant Triton-X-100) applying electrochemical measurements. Complying with the theoretical prediction for nonfractal and fractal systems with D=d=2, the measured  $C_{\rm s}$  values, obtained for the pure electrolyte (nonfractal system) and for the adsorbed layers with fractal dimension D=2, proved to be size-and dimension-independent. For fractal systems with  $D\neq 2.0$ ,  $C_{\rm s}$  is D- and size-dependent as predicted. The rate of change of  $C_{\rm s}$  depends on the magnitude of the fractal dimension D, size of the fractal structure, and factor  $\beta$  comprising the material and geometrical features.

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