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**STRUCTURAL AND TOPOLOGICAL CONTRIBUTIONS TO THE ELECTRONIC  
PROPERTIES OF INHOMOGENEOUS MEDIA**

*City University of New York*

**Ph.D. 1986**

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STRUCTURAL AND TOPOLOGICAL CONTRIBUTIONS  
TO THE ELECTRONIC PROPERTIES  
OF INHOMOGENEOUS MEDIA

by

MICHAEL KRAMER

A dissertation submitted to the Graduate Faculty in  
Physics in partial fulfillment of the requirements for the  
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1986

This manuscript has been read and accepted for the Graduate Faculty in Physics in satisfaction of the dissertation requirement for the degree of Doctor of Philosophy.

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## ABSTRACT

### STRUCTURAL AND TOPOLOGICAL CONTRIBUTIONS TO THE ELECTRONIC PROPERTIES OF INHOMOGENEOUS MEDIA

by

MICHAEL KRAMER

Adviser: Professor Micha Tomkiewicz

Recent theoretical developments in the theory of inhomogeneous media have enabled Physicists to explore various properties of random composites. We demonstrate how these techniques can be applied to the treatment of electrochemical systems, specifically porous structures. A Random Network Model is introduced to represent the porous electrode immersed in electrolyte, and using numerical simulation the a.c. impedance behavior of the system is explored. We explore how various compositional and structural parameters contribute to the electrical properties of the system. The topology of our model is analyzed, and the effect of fractal structures on impedance behavior is considered. To examine the electrode topology during electrode operation, another model is introduced, the dissolution-precipitation model. We show how the model correctly predicts the concentration profile of dissolved particles in solution and how the reaction produces fractal structures on the surface of the model electrode.

Dedicated to my dear wife, Debby

and to

Yehoshua, Yair and Shoshana

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## Introduction

The focus of Physics in the 19th century was on the world of the visible and experiential - gravity, mechanics, electricity and magnetism. The Newtonian and Maxwellian theories are extraordinarily successful in describing the world of our senses. With the advent of the twentieth century and the birth of quantum mechanics much of the physicist's attention shifted to the opposite extreme - the microscopic world of atoms and subatomic particles. The quantum theory of solids has brought with it a deep understanding of the behavior of materials from first principles. The behavior of electrons in the solid's crystal potential, the interaction of electromagnetic radiation with the solid, the identification of phonons and their effect on elastic, thermal and electrical properties are all relatively well understood and have sparked our technological explosion.

The existence of multi-phase granular systems presents new challenges, due to the fact that neither of the above approaches are suitable for an accurate treatment of this problem. To approach the electronic properties of the system- on the one hand, each phase of the material can be well characterized by local electronic properties (the inhomogeneity is manifest on a length scale larger than the electronic mean free path). On the other hand, the system is macroscopically inhomogeneous, and horrendous boundary conditions confront anyone who would attempt to solve Maxwell's equations

across the different phases. Recent studies into phenomena based on random aggregation and the clustering of small particles have demonstrated that by utilizing statistical techniques, the aggregate properties of composite materials may be derived from the local behavior of the system's constituents.

This work is an attempt to apply the study of inhomogeneous media to electrochemical systems, particularly porous structures, and to explore the contributions of the electrode morphology to the electronic properties of the system. The basis of the research is a computer model of the electrode system that captures the composite media character of the electrode. Chapter I presents an overview of the theory of conduction in heterogeneous media, and Chapter II describes the porous electrode-electrolyte system and explains the applicability of the composite media description. Chapter III introduces the Random Network Model- the structure of the system, the impedance characteristics and the relationship between them. Chapter IV details an alternative simulation for the construction of structures on the surface of porous electrodes and sketches how this may be used to further explore the system.

## Electronic Conduction in Composite Media:

### Theoretical Tools

#### A. Introduction

Electronic conduction in randomly disordered systems has been explored using the tools of percolation theory. If the system under study is composed of an insulating phase and a conducting phase, then by varying the composition of the system, a sharp change in the conductivity of the system is observed. The critical point (of volume fraction) at which this occurs is termed the percolation threshold and is determined by the dimensionality of the system. Percolation is actually a general process that applies to many systems in which a random media can exhibit long range connectivity. If the composition of the system is varied, the point at which this long range connectivity disappears is the percolation threshold. In this manner, percolation has been used as a model to describe the flow of liquid in a porous media, stochastic star formations in spiral galaxies, and dilute magnetic systems, to name just a few areas of application<sup>1</sup>.

Percolation can be illustrated by considering a lattice of  $N$  sites (which we will consider in the limit as  $N \rightarrow \infty$ ), in which some of the sites are randomly selected as being "allowed", and the remaining sites are vacant. In addition to this site percolation, one can formulate the problem in terms of allowed and missing bonds to arrive at bond percolation. If  $c$  is defined as the concentration of

allowed sites, then when  $c$  is very small, allowed sites occur as scattered small islands in the lattice. As  $c$  increases, the clusters grow larger and larger, until we reach  $c^*$ , where the mean cluster size approaches the size of an infinite cluster (i.e. it provides a path of connectivity through the whole space). If  $P(c)$  is the ratio of the number of sites in the infinite cluster to the number of sites on the lattice,  $P(c)$  becomes the probability that a given site will be part of the infinite cluster. A plot of  $P(c)$  as a function of  $c$  for a 3D lattice is shown in Figure 1.1, and the existence of a critical point is seen from the shape of the curve as  $c \rightarrow c^*$ . Near threshold (from above), the behavior of  $P(c)$  may be characterized by a power law,

$$P(c) \propto (c - c^*)^s, \quad (1.1)$$

where  $s$ , for a cubic lattice, has been found to be approximately

$$0.3 \leq s \leq 0.4,^2$$

and  $c^* \approx .3$  (site percolation)<sup>66</sup> and  $.25$  (bond percolation)<sup>67</sup>.

An alternate quantity of interest is the correlation length,  $\xi$ , which measures the average cluster size. As the concentration approaches the critical concentration, the formation of the infinite cluster described above is represented by the divergence of  $\xi$ , such that

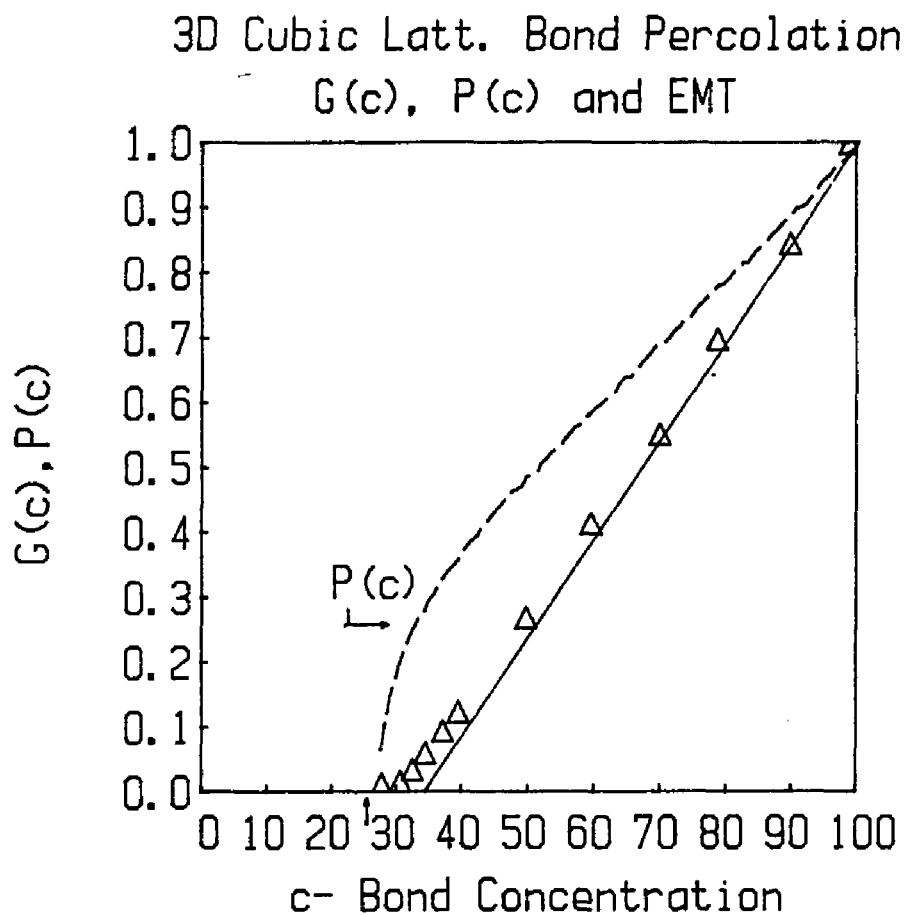


Figure 1.1 - 3D Bond Percolation, after Kirkpatrick (Ref. 3).  $c$  is the % bond concentration of conducting bonds. The data for  $P(c)$  (dotted line) and  $G(c)$  (triangles) are obtained by a computer simulation on a cubic lattice of size  $15 \times 15 \times 15$ .  $P(c)$  is the probability of a given site being part of the infinite cluster,  $G(c)$  is the conductance of the system, and the solid line is the Effective Medium Prediction for  $G(c)$ . The arrow under the x axis corresponds to the location of  $c^*$ , the percolation threshold.

$$\xi = \xi_0(c-c^*)^{-\nu} \quad (1.2)$$

where  $\xi_0$  is the size of the granules of which the composite material is made. As one approaches the percolation threshold, the correlation length becomes the dominant length scale of the system. The exponent  $\nu$  will prove to be of some theoretical importance, as we shall see later.

It has been shown, however, that  $P(c)$  is not suitable for representing the dependence of the conductivity on the composition of the system<sup>3</sup>. Although an allowed site may be part of a cluster, it is not necessarily a participant in the transport properties of the system. Many cluster "dead ends" will increase the probability of an infinite cluster, but will not themselves be participating in conduction. In order to formulate the percolation problem in terms of conductivity,  $\sigma(c)$ , Kirkpatrick<sup>5</sup> introduced the Random Resistor Network Model.

#### B. The Random Resistor Network Model

We assume that the system is microscopically inhomogeneous but that the dimensions of the conducting regions are large with respect to the electronic mean free paths. We may therefore define a local conductivity,  $\sigma(\vec{r})$ , at a position  $\vec{r}$  in the material. The system is considered to have two phases of microscopic conductances  $\sigma_1(\vec{r})=0$ , and  $\sigma_2(\vec{r})=1$ , and we suppose that we have a detailed knowledge of how

the two phases are arranged. We can calculate the macroscopic conductivity of the system using the Ohm's law equation

$$\mathbf{j}(\mathbf{r}) = -\sigma(\mathbf{r}) \nabla V(\mathbf{r}) \quad (1.3)$$

and the continuity equation

$$\nabla \cdot \mathbf{j}(\mathbf{r}) = 0. \quad (1.4)$$

By introducing a cubic mesh of points  $\{r_i\}$  with spacing  $\Delta r$ , these continuum equations may be solved using a finite difference approximation:

$$\sum_j g_{ij} (V_i - V_j) = 0 \quad (1.5)$$

with  $g_{ij} = \Delta r \sigma[(r_i + r_j)/2]$ , and  $i, j$  being neighboring sites on the mesh. Kirkpatrick noted<sup>5</sup> that Equation 1.5 is identical to the Kirchoff's law equations for a 3D network of resistors, and Equations 1.3-1.5 may be solved by numerical simulation. We define a cubic mesh of resistors, and using a random number generator a specified fraction,  $c$ , of these resistors are assigned conductivity 1 while the remaining resistors are assigned conductivity 0.  $\sigma(c)$  as a function of the relative concentration  $c$ , is also shown in Figure 1.1. Again it is determined that near the threshold (as approached from above),

$$\sigma(c) \propto (c - c^*)^t \quad (1.6)$$

where  $t \approx 1.94 \pm 1$  for both bond and site percolation<sup>6,8</sup>. (The value of  $c^*$  remains  $\approx .25$  (bond percolation) and  $.3$  (site percolation)). Thus far the numerical simulations considered only discrete lattice percolation; by introducing correlations between neighboring bonds, Webman, Jortner and Cohen<sup>6</sup> transformed the problem into a continuous percolation model, and showed that  $c^* \approx .15$  (bond percolation).

A generalization of the above result<sup>7,8</sup> is for  $\sigma_2 = b$  with a probability  $c$  and  $\sigma_1 = a$  with a probability  $(1-c)$ , where  $a$  and  $b$  are both greater than zero. If we assume that  $a \ll b$ , then we have:

$$\sigma \propto b (c - c^*)^t \quad \text{for } c > c^* \quad (1.7a)$$

$$\sigma \propto a (c^* - c)^{-s} \quad \text{for } c < c^* \quad (1.7b)$$

Equation 1.7b may be explained by observing that although  $a$  is the low conductance phase of material, at very low concentrations of "b" the current is forced to traverse a path through "a". As the concentration of "b" increases, the current shorts through clusters of "b", resulting in an apparent divergence in the conductivity as one approaches threshold ( $c^*$ ). At  $c = c^*$ , however, there is no divergence of the conductivity due to the fact that the current does not follow all the twists and turns of the conducting phase and travels through the more numerous paths of the "a" phase, even though it is more poorly conducting. (There is also no divergence

in the correlation length). If we assume that at  $c^*$  the power laws in Equations 1.7 are no longer applicable, and  $\sigma$  is almost constant in a small region  $\delta = |c - c^*|$ , we may equate Equations 1.7 at  $c = c^* \pm \delta$ :

$$\sigma = a\delta^{-s} = b\delta^t$$

we find that the "crossover region" is defined by

$$\delta = \left(\frac{a}{b}\right)^{1/(t+s)}$$

and that near  $c^*$ , the conductivity is defined as:

$$\sigma \propto a^u b^{1-u}, \quad u = t/(s+t). \quad (1.7c)$$

Thus far we have considered only systems containing a mixtures of pure conductors and non-conductors. Bergman and Imry<sup>9</sup> considered the case of a heterogeneous mixture of a conducting phase and an insulating dielectric phase. The bulk effective dielectric constant,  $\kappa_e$  is

$$\kappa_e = \epsilon_e + \frac{4\pi\sigma_e}{i\omega} \quad (1.8)$$

where  $\epsilon_e$  and  $\sigma_e$  are the bulk static dielectric constant and conductivity respectively, and  $\omega$  is the frequency of the excitation signal. For  $\kappa_1 = \epsilon_1$  and  $\kappa_2 = 4\pi\sigma_2/i\omega$ , with  $|\kappa_1| \ll |\kappa_2|$  and near

the percolation threshold  $c_2 \rightarrow c^*$ ,  $\sigma_e$  scales as  $(c - c^*)^\alpha$  while  $\epsilon_e$  increases as  $(c - c^*)^{-\gamma(1-\beta)}$ . They postulated that the large enhancement of the dielectric constant as one approaches  $c^*$  from below is due to the presence of long paths of conducting material separated by thin barriers, whose effective capacitance is enormous. In addition, at  $c=c^*$ ,

$$\sigma_e \sim \omega^\beta \quad \text{and} \quad \epsilon_e \sim \omega^{\beta-1}, \quad \text{where } \beta=0.73 \pm 0.05 \quad . \quad (1.9)$$

Equations 1.7 present a scaling behavior for the conductivity, and exponents  $s, t$  and  $u$  are the critical exponents that characterize this scaling relation. Values for these exponents have been obtained from numerical simulation, and for 3D bond percolation are:

$$s=.75 \quad t=1.94 \quad u=.67 \pm 0.08 \quad .^{69}$$

A correlation length for the resistor network has been defined by Bass and Stephen<sup>11</sup> as follows: apply a potential  $V_0$  at vertex 0, and measure the average potential  $\langle V_n \rangle$  at vertices  $n$  links away. Then

$$\xi^{-1} = -\lim_{n \rightarrow \infty} n^{-1} \ln \langle V_n / V_0 \rangle \quad . \quad (1.10)$$

### C. Finite Size Scaling and Real Space Renormalization

The existence of the scaling relationships of Equations 1.7 has prompted the application of scaling theories<sup>12</sup> which have been successfully applied to critical phenomena, such as magnetic spin systems (the simplest of them being the Ising Model). Real space renormalization is applied to map the system into regions of physical parameters where the correlations are easy to compute. Following this, one matches these transformed values to the original correlations, and calculates the correlation function. This allows us to identify the correlation length  $\xi$  (which is the scale on which, for a particular concentration, the system first appears homogeneous<sup>13</sup>), and to calculate the critical exponent  $\nu$  from Equation 1.2. (A detailed example calculation is presented in Stauffer, p. 15). This renormalization is allowed due to the divergence of  $\xi$  near  $c^*$ , so that the system's properties are invariant no matter which finite length scales we use to investigate the system. "Thus it should not matter at  $c^*$  on what length scale we are investigating the system; apart from simple scale factors the system looks similar whether we look at it with the eye, with a magnifying glass, or with an optical microscope"<sup>14</sup>.

Systems that exhibit such properties of self-similarity under transformations of scale are prime candidates for classification as fractal systems. In fact, a major theoretical contribution to the study of critical behavior in heterogeneous systems due to Mandelbrot<sup>15</sup> is the concept that the scaling property of the

clusters result from the analytic properties of the cluster's geometry. Although the clusters constitute a random disordered system, when the clusters are treated as fractals their geometric properties become simply describable through their fractal dimension, which is related to the exponent  $\nu$  of Equation 1.2.

#### D. Fractals

The traditional Euclidean geometry defines geometric constructs as occurring in a Euclidean space  $E$  (the dimensions of the space in which it is embedded), and having a topological dimension,  $D_T$  (e.g. a line or a circle have  $D_T=1$ ). In connection with some systems, however, mathematicians have found it useful to define other dimensions, one such being the Hausdorff dimension,  $D$ . The Hausdorff dimension describes how the measure of an object changes under the application of different scales of measurement. In general,

$$D_T \leq D \leq E \quad (1.11)$$

and specifically, Mandelbrot advances the following definition:

A fractal object is one whose Hausdorff dimension is strictly greater than its topological dimension.  $D$  is termed the object's fractal dimension, which is in general not an integer.

The classic example of a fractal dimension is the Richardson Effect (Figure 1.2). If we wish to measure the length of an island coastline, we can take a ruler and start to measure the length of the land-water border. It quickly becomes apparent though, that the resulting length measure is totally dependent on the size of the ruler with which we measured the coast (the resolution of the measurement). As we decrease the size of our measuring stick we are able to measure the coast with finer and finer detail. The result is the Richardson curves of Figure 1.2, where the relationship between the length scale used and the total length is linear when viewed on a log-log plot. If  $\epsilon$  is the resolution of the measurement, and  $L(\epsilon)$  is the total length measured, then

$$L(\epsilon) \propto \epsilon^{1-D} \quad (1.12)$$

where  $D$  is the fractal dimension. Thus, coastlines are representative of a fractal curve. Mandelbrot argues that  $D$  is the only measure of convincing physical significance, since it is the only scale-invariant quantity in the problem. In the case of the coast of Britain, the Richardson slope is approximately  $-.2$ , which means that  $D=1.2$ , which is indeed greater than the topological dimension of the coastline curve ( $D_T=1$ ), thus satisfying the condition for a fractal.

An alternate definition of fractals is the relationship between measure of contained mass and radius, (which implies a relationship

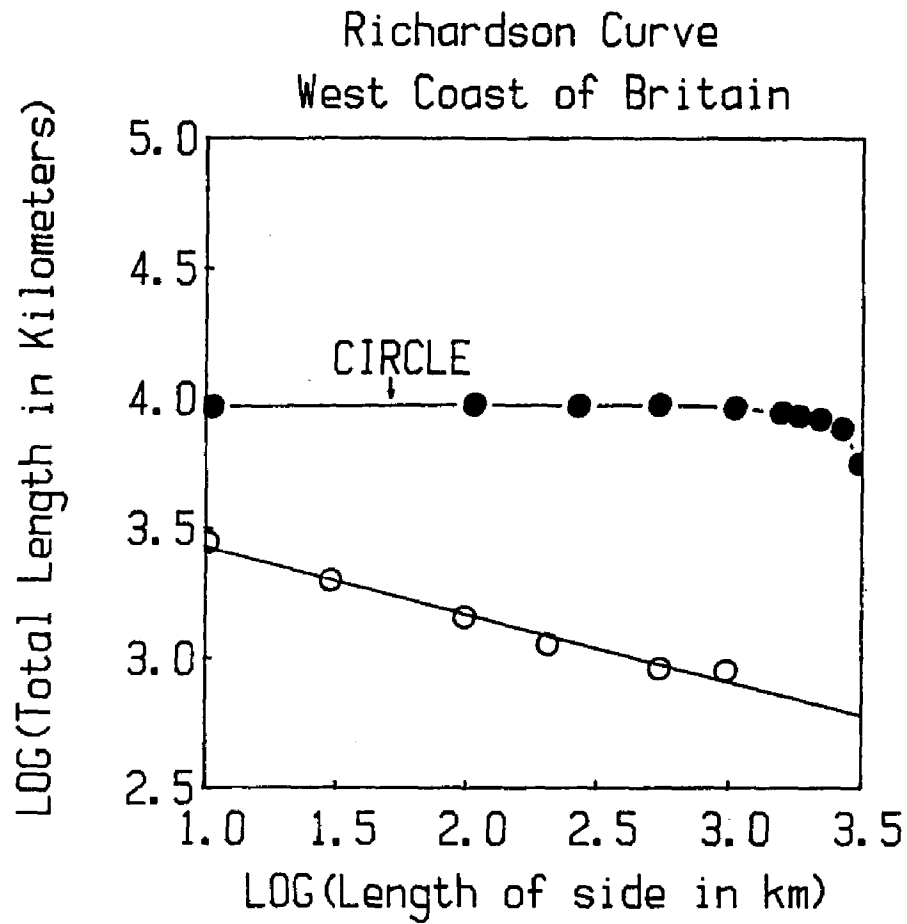


Figure 1.2 - The Richardson curve illustrating that the coast of Britain (outlined points) is a fractal curve. The Richardson Curve for a circle (solid points) has a slope of 0.0, indicating that its effective dimension is equal to its topological dimension. (After Mandelbrot, Ref. 15)

between length and volume, or length and area). The amount of mass,  $M$ , contained in an object of radius  $R$  can be expressed as:

$$M(R) \propto R^D . \quad (1.13)$$

(We are well acquainted with the classic cases for which  $D=3$  (e.g. a homogeneous sphere) or  $D=2$  (e.g. a homogeneous 2-D square plate). It is this relationship which may be utilized to calculate the fractal dimension of a percolating cluster. The straightforward procedure<sup>14</sup> consists of taking circles or spheres of increasing size and measuring their contents or mass (i.e. how much of the percolating cluster do they contain at different sizes), and using Equation 1.11 to directly calculate  $D$ . Kirkpatrick<sup>16</sup> employs a variation of this method for calculating a fractal dimension of the "backbone" of the infinite cluster.

An alternate expression of Equation 1.11 is the scaling property of the density-density correlation function<sup>19,20</sup>:

$$C(r) \equiv N^{-1} \sum_{r'} \rho(r')\rho(r'+r) \propto r^{D-d} \equiv r^{-A} \quad (1.14)$$

where  $d$  is the spatial (Euclidean) dimension ( $E$ ), and  $D$  is the fractal dimension. The density  $\rho(r)$  is defined to be 1 for occupied sites and 0 for empty sites. The calculation is performed by computing  $C(r)$  for a given  $r$  in a given direction over all the particles in the lattice (or in the cluster), and then  $C(r)$  is

averaged over different directions. This is repeated for a number of discrete lengths  $r$ , and the exponent  $A$  is evaluated from the slope of  $\log(C(r))$  plotted as a function of  $\log(r)$ . The utility of this calculation is that it is easy to perform in computer aggregation simulations. This relationship is also directly verifiable using scattering experiments such as neutron or X-ray scattering. The structure factor,  $S(k)$  which is measured in a scattering experiment, is none other than the Fourier transform of  $C(r)$ .<sup>20,21</sup> Thus, we may relate the scattering intensity,  $I$ , and the wavenumber,  $k$ , by

$$I(k) \propto k^{-x} . \quad (1.15)$$

The interpretation of this exponent,  $x$ , is dependant on the scattering system. For "volume fractals" (i.e. polymer-like structures),  $x$  is none other than  $D$  from Equation 1.13; for scattering from surfaces,  $x=6-D$ , where  $D$  is the fractal dimension of the surface ( $D=2$  corresponds to a flat surface). For fractally porous materials<sup>22,23,24</sup>,  $x=7-\gamma$ , where  $\gamma$  is the exponent describing the distribution of pores of radius  $r$ ,

$$P(r) = r^{-\gamma} . \quad (1.16)$$

Real life objects do not exhibit fractal behavior over all length scales. In practice, there is an inner and outer cutoff for  $\epsilon$  and  $R$ , within which Equations 1.12 and 1.13 hold - corresponding to the

range where these systems exhibit self-similarity. In the percolation problem the bounds are from the lattice size,  $a$ , to the correlation length,  $\xi$ .<sup>16</sup>

#### E. Scaling of the Conductance on Percolating Fractal

By combining Equation 1.2 and the conductance scaling relationship of Equation 1.7b, we find that the conductivity may be expressed as

$$\sigma \propto \xi^{-\mu/\nu} \quad (1.17)$$

Using finite-size scaling arguments due to the self-similarity of the system, Palevski and Deutscher<sup>25</sup> show that for a system of size  $L \ll \xi$ :

$$\sigma \propto L^{-\mu/\nu} \quad (1.18)$$

Thus, effective dimensions for the impedance properties of the system may be calculated using methods similar to those for the classic fractal relationships reviewed above. In fact, Palevski and Deutscher measure the dependence of  $R(L)$  as a function of  $L$  (where  $R(L)$  is the resistance in a segment of the percolating gold clusters and  $L$  is length of the side of a square within which they measure the resistance). As they increase the area of the square within which they measure  $R$ , they observe a linear relationship between  $\log$

$R(L)$  and  $\log L$ . Computation of the slope of that line yields the ratio  $\mu/\nu$ .

#### F. Effective Medium Theory (EMT)

While the Random Resistor Network approach treats the electrical properties of the system as explicitly arising from the system's microscopic constituents, EMT takes almost the opposite approach. We consider a composite system made up of two materials having conductivity  $\sigma_1$  and  $\sigma_2$ . Our objective is to calculate  $\sigma_e$ , an effective conductivity of the entire composite. We begin<sup>28</sup> by considering a small (spherical) grain inside the material, and treat that grain as if it is embedded in a homogeneous effective medium, of conductivity  $\sigma_e$ , to be computed self-consistently (see Figure 1.3). Let the field and current density far from the central grain be  $E_0$  and  $J_0 = \sigma_e E_0$ . We can therefore calculate the fields and current density within the grain to be

$$E_{in} = [3\sigma_e / (\sigma_1 + 2\sigma_e)] E_0 \quad (1.19)$$

$$J_{in} = \sigma_1 E_{in} \quad (1.20)$$

where  $\sigma_1$  is either  $\sigma_1$  or  $\sigma_2$ . (the microscopic assumptions and geometrical considerations necessary to produce Equation 1.19 are somewhat controversial, although all derivations produce this result).<sup>26,27,28</sup> The self consistency for computing  $\sigma_e$  comes from the assumption that

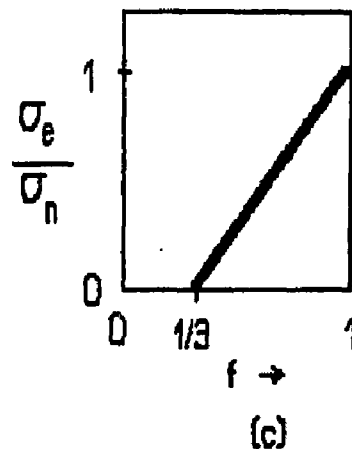
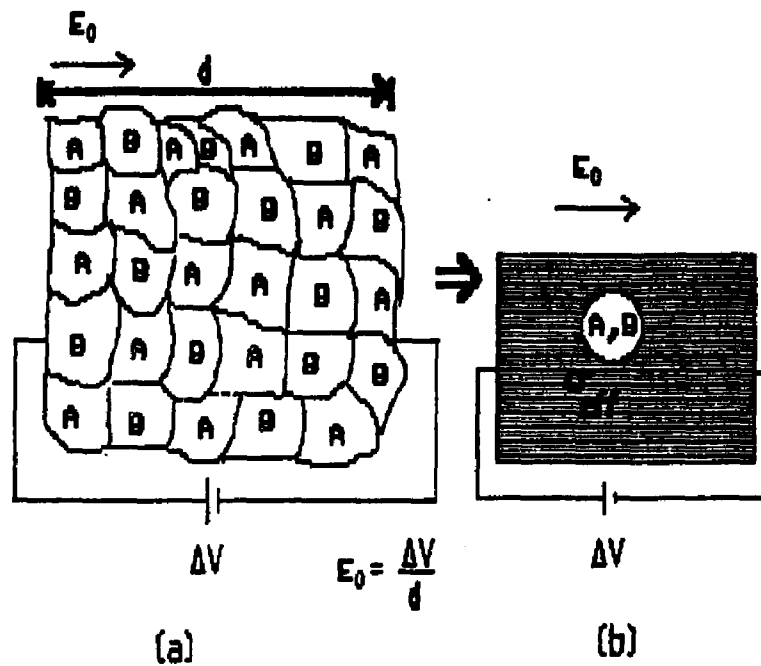


Figure 1.3 - Illustration of Effective Medium Theory. (After Stroud, Ref. 28).  
 (a) Schematic of a two-component composite in an applied electric field,  $E_0$ .  
 (b) An illustration of the self-consistent embedding procedure which defines the effective conductivity,  $\sigma_e$ .  
 (c) Schematic behavior of  $\sigma_e$  as a function of concentration,  $f$  (from the application of Equation 1.23).

$$\langle J_{in} \rangle = \sigma_e \langle E_{in} \rangle . \quad (1.21)$$

Assuming that our composite is made up of the fraction  $f$  of component 1 and  $(1-f)$  of component 2, we can substitute Equations 1.19 and 1.20 into 1.21 to yield,

$$\frac{3f\sigma_1}{\sigma_1+2\sigma_e} + \frac{3(1-f)\sigma_2}{\sigma_2+2\sigma_e} = \sigma_e \left( \frac{3f}{\sigma_1+2\sigma_e} + \frac{3(1-f)}{\sigma_2+2\sigma_e} \right) \quad (1.22)$$

which simplifies to:

$$f \frac{\sigma_1 - \sigma_e}{\sigma_1 + 2\sigma_e} + (1-f) \frac{\sigma_2 - \sigma_e}{\sigma_2 + 2\sigma_e} = 0 . \quad (1.23)$$

Although EMT has proved to be a useful approximation for dealing with systems about which we have very little structural information, the theory fails when a percolating system approaches the percolation threshold. EMT predicts a much higher critical concentration,  $c^*$ , than is observed experimentally or by numerical simulation<sup>3</sup>. This may be explained<sup>29</sup> by noting our bias for spherical conducting geometries. If in fact our system consists of flattened spheroids or elongated conducting components, the probability of connectivity through the system is enhanced at lower conductor concentrations. In a random system, there is no a priori reason to assume the favoring of spherical structures over elongated ones. This illustrates that some knowledge of the internal geometry of a particular system is necessary for the application of EMT such a system.

### G. Summary

We have presented a multiplicity of techniques for the theoretical treatment of conduction in heterogeneous media. The current presentation has centered on a simple application, the impedance characterization of a two-phase random mixture of conductor and non-conductor. It should be pointed out however, that these theories may be adapted to treat more complex systems, not only systems with multiple component phases, but also systems with unique non-random correlations, and systems with specialized geometries. Moreover, although this work focuses on conduction in composite media, these are general tools equally applicable for the treatment of a wide range of transport and static problems in stochastic systems, all revolving around the properties of clustering and aggregation in these systems. A full discussion of all the ramifications of these theories are beyond the scope of this work (and me). In the coming chapters we will employ these concepts and techniques to our problem at hand - the stochastic morphology of the porous electrode and it's contribution to the electronic conduction.

## II. The Chemistry and Physics of Porous Electrodes

### A. Solid-Liquid Interfaces

When two phases of material are brought into contact with each other, the difference in their free energy develops a potential difference across the interface. When these two phases are metal (electrode) and electrolyte, the potential difference induces a surface charge on the metal and region of net charge of opposite sign in the electrolyte. In the Gouy-Chapman (GC) model of the interface, there is a diffuse region of local net charge that extends a distance into the electrolyte. If we imagine a microscopically thin lamina of electrolyte at a distance  $x$  from the interface, utilizing Boltzmann statistics the charge density in that region can be written as

$$\rho(x) = nze \exp[-ze\phi(x)/k_B T] \quad (2.1)$$

where  $n$  is the ion concentration,  $z$  is the net charge on each ion,  $\phi(x)$  is the electrostatic potential at position  $x$  in the solution with respect to the interface,  $k_B$  is the Boltzmann constant, and  $T$  is the absolute temperature. Combining this with the Poisson equation:

$$\nabla^2 \phi = \frac{4\pi}{\epsilon} \rho \quad (2.2)$$

the approximate solution is<sup>30</sup>

$$\phi = \phi_0 e^{-\kappa x} \quad (2.3)$$

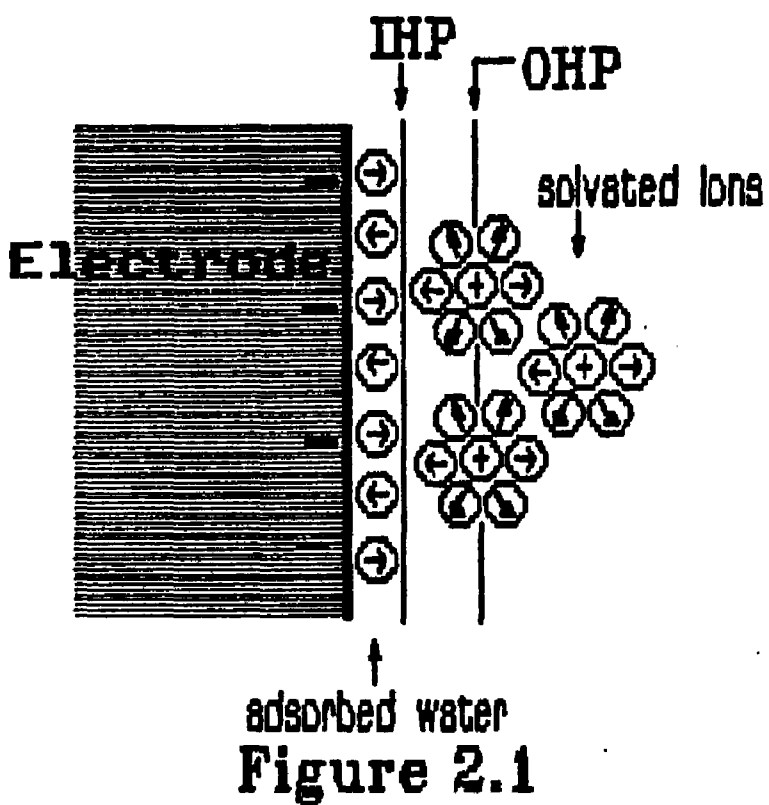
$\kappa$  has units of inverse length and  $\kappa^{-1}$  is the characteristic expanse of the charge distribution. Thus, we can consider all the charge in solution to be located on a plane parallel to the interface and located at  $d_{GC} = \kappa^{-1}$ . The behavior of this double layer is therefore identical to that of a parallel plate capacitor of capacitance

$$C_{GC} = \frac{\epsilon_0 A}{GC} \quad (2.4) \quad d$$

where  $\epsilon_0$  is the permittivity of free space and  $A$  is the cross sectional area of the interface. It is the electric field at the interface, represented by the capacitor, that constitutes the driving force for charge transfer across the interface and which represents the electrode potential (in comparison with other electrodes in solution). Another field at the interface is generated by adsorbed water molecules and unsolvated ions on the interface and it is also associated with a capacitance  $C_H$ , the Helmholtz Capacitance. The total capacitance of the interface is due to two capacitors in series, and the total capacitance is:

$$\frac{1}{C} = \frac{1}{C_H} + \frac{1}{C_{GC}} \quad (2.5)$$

Figure 2.1 is a diagram of the charge distribution at the interface and the location of the two planes of effective charge. At high



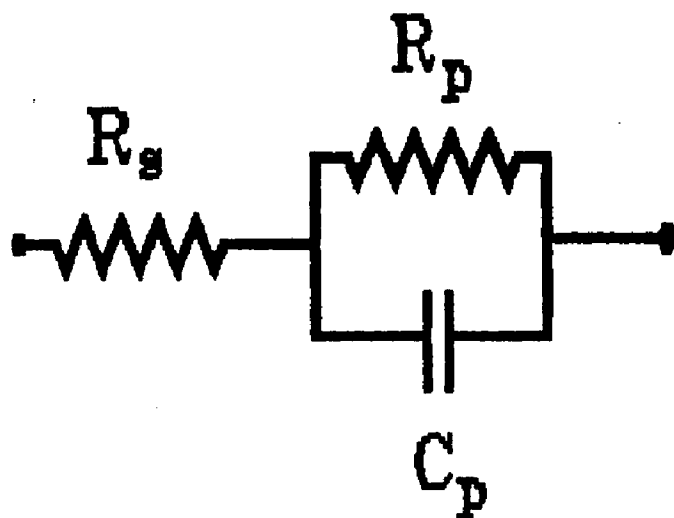
Schematic of the electrified interface. The IHP (inner Helmholtz plane) is due to the adsorbed surface species, and the OHP (outer Helmholtz plane) is due to the diffuse charge distribution in solution.

solution concentrations the GC layer is very narrow, and as a result of the capacitance behavior of Equation 2.4,  $C_{GC}$  becomes large and may be ignored in comparison with  $C_H$ . At low electrolyte concentrations  $C_{GC}$  dominates (the Helmholtz capacitance is independent of concentration).

Thus far we have considered no faradaic current flow across the interface, an assumption of an Ideally Polarizable Electrode (IPE). More generally (depending on the catalytic properties of the metal and on the solution), the metal electrolyte interface is represented by the equivalent circuit model of Figure 2.2, where  $R_p$  represents a leakage current (a deviation from IPE behavior). In Figure 2.2  $R_s$  is the resistance of the metal - which is usually negligible.

If there is an insulator or semiconductor build up on the electrode due to some chemical reaction at the surface, then the metal-solution capacitance remains, but the value will be altered by the dielectric properties of the new layer. In this case, the  $R_s$  of Figure 2.2 is the bulk resistance of the insulating or semiconducting layer on the interface.

The bulk electrolyte solution has an ohmic conductivity due to the movement of ions, and for a given volume of solution we can represent the current carrying capacity of the solution using an equivalent resistance,



**Figure 2.2**

Equivalent circuit model of the solid-liquid interface.

$$R_e = \frac{\rho_e \ell}{A}, \quad (2.6)$$

where the electrolyte volume is defined by the length  $\ell$  and cross-sectional area  $A$ , and the electrolyte resistivity is determined from the relationship<sup>31</sup>

$$\rho_e = \frac{1}{nz\mu} \quad (2.7)$$

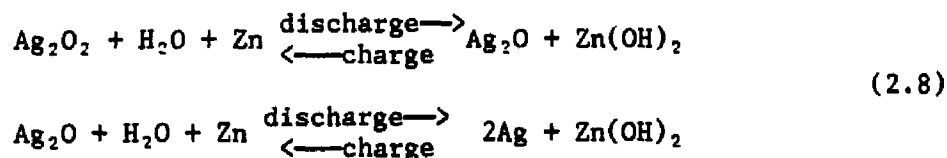
$n$  is the number of ions in the given volume,  $z$  is the charge per ion, and  $\mu$  is the mobility determined by the characteristic species in solution.

A useful experimental technique for the study of electrochemical systems is the use of a.c. impedance measurements<sup>32</sup>. These measurements allow for nondestructive in-situ characterization of the system. Impedance measurements are a natural choice given the theoretical behavior of these systems as simple passive circuit components. Using very low signal a.c. excitations, we can measure both static and kinetic electrochemical processes.

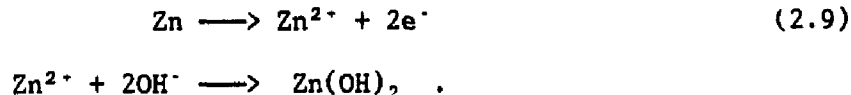
#### B. Typical Application: A Secondary Battery Cell

Perhaps the most important technological application of these electrochemical principles is in battery electrodes. A typical example is the Silver-Zinc cell of Figure 2.3<sup>33</sup>. A silver oxide

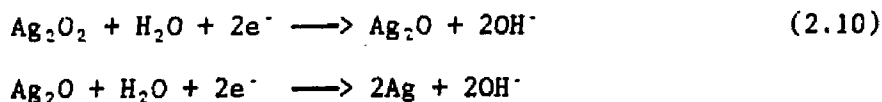
electrode and a Zinc electrode are placed in a solution of KOH. The overall reaction is a two step one<sup>34</sup>:



In the discharge process the Zn electrode is oxidized, liberating electrons to flow through the external circuit, and forming a layer of ZnO on the electrode. The half-cell reaction is:



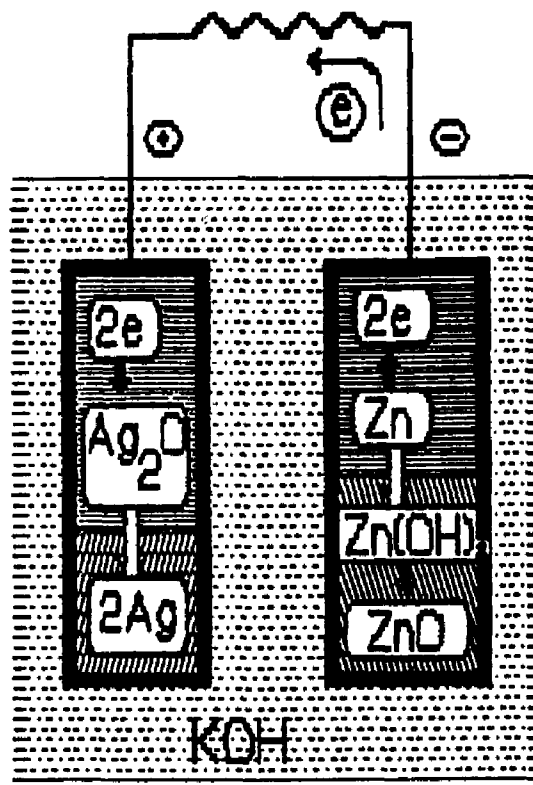
On the silver electrode the AgO is reduced to produce Ag<sup>35</sup>:



or schematically:



Focusing on the Zn electrode, the net result of the discharge process is the buildup of ZnO on the surface of the electrode. The state of charge of the electrode can be determined from the amount of ZnO on the surface, and as previously mentioned this changes the capacitance of the interface.



**Figure 2.3**

A typical secondary Battery cell during discharge.  
 During discharge, a layer of  $\text{ZnO}$  is forming on the anode.

The most important characteristics of any battery are its storage capacity, its energy density and its power density. The energy density can be increased by the choice of two electrodes with large differences in their electrode potential, and by producing a very high current density on discharge. To achieve this, a large real surface area is needed between the metal and the electrolyte which can be obtained by using a porous electrode structure. Indeed, all commercial batteries are based on porous morphology. Although the electrochemical theory discussed thus far assumes flat surfaces, it is a logical starting point for the treatment of rough surfaces associated with porous electrodes. It is clear though, that some corrections and modifications are needed to account for the new morphological effects that accompany the increased current density.

#### C. Porous Electrodes - Classical Approach

DeLevie<sup>36</sup> introduced the first coherent treatment of the effects of surface roughness. He considered a single pore with a uniform cross section, homogeneously filled with electrolyte, and assumed that the resistance of the electrode material is negligible. The actual potentials in the pore are replaced by their average values in planes perpendicular to the pore axis. This may be represented by the transmission line equivalent circuit model of Figure 2.4, which transforms the problem into a one-dimensional one.  $R$  is the solution resistance per unit pore length ( $R$  has dimensions of ohm/cm), and  $Z$  is the impedance of the electrode-electrolyte interface per unit pore length [ $Z$  has dimensions of ohm-cm and  $Z/dx$  is the

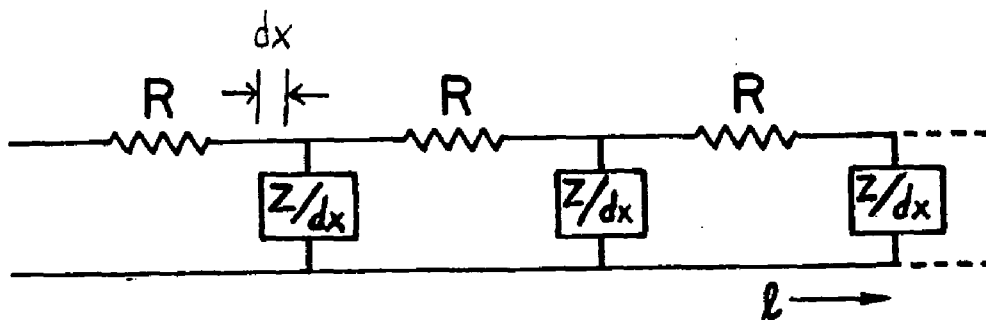


Figure 2.4- DeLevie's single pore transmission line model, for a pore of length  $l$ .  $R$  is the electrolyte resistance per unit length, and  $Z/dx$  is the impedance of the interface (a microscopic section of length  $dx$ ).

interfacial impedance of a tiny expanse of the pore of length  $dx$  - this makes sense if you consider an example of a pure capacitance on the interface of value ( $C$  per unit pore length) ; the impedance of a length  $dx$  of that will be  $1/(i\omega Cdx)$ . Solving the current-voltage equations for the transmission line yields the impedance of the entire pore of length  $\ell$ :

$$Z_0 = (RZ)^{1/2} \coth(\rho\ell) \quad (2.11)$$

where  $\rho = (R/Z)^{1/2}$ .

This leads to the definition of a characteristic penetration depth for the current in the pore,  $\lambda = 1/\rho$ . When  $\ell \gg \lambda$   $\coth(\rho\ell) \approx 1$  which DeLevie refers to as the "semi-infinite pore" where there are effectively no "edge effects" due to the pore end. Another important result of this model is the fact that although  $Z$  represents the interfacial impedance of a unit flat surface, the presence of the pore produces a  $Z^{1/2}$  dependence for the entire rough surface. For an arbitrary a.c. signal, this translates into a frequency dependence of

$$Z \propto \omega^{-1/2} \quad (2.12)$$

and in the literature this has been taken to be the "signature" of a rough surface<sup>37</sup>, as opposed to an  $\omega^{-1}$  dependence for a flat surface. (This signature, however is not unique since one can show, for example, that the Warburg impedance which is a pure diffusion phenomenon unrelated to morphology, is also characterized by  $\omega^{-1/2}$ ).

The definition of penetration depth in Equation 2.11 also illustrates the fact that at high frequencies the a.c. current penetrates very little into the pores. The ratio of  $Z/R$  controls the penetration due to the fact that at  $Z \gg R$ , the current tends to flow through the "top" of the transmission line in Figure 2.4 (through  $R$  exclusively). On the other hand, when  $Z \ll R$ , then the current tends to flow in Figure 2.4, right through the first "down" path that it finds (through  $Z$ ). At high frequencies,  $Z$  is low and the current short circuits "down" early in its flow through the pore.

#### D. Critique of Classical Model- A Search for Alternatives...

There are numerous problems with this approach to rough surface impedance behavior. The most striking difficulty is that there is a large body of experimental evidence to the fact that Equation 2.12 is simply not a unique characterization of surface roughness. Some manifestly rough surfaces have impedance behavior of a smooth surface<sup>38</sup>. In addition, experimental studies have found many different frequency dependences not at all limited to exponents of  $-.5$ .<sup>39,40</sup> A general dependence of

$$Z \propto \omega^{-\beta} \quad (2.13)$$

where  $0 \leq \beta \leq 1.0$ , emerges from the literature, with the deviation from  $\beta=1$  attributed to surface roughness<sup>41,42</sup>. In various studies this fractional frequency scaling has been called a Constant Phase Angle (CPA)<sup>41</sup>, Constant Phase Element (CPE)<sup>43</sup>, or Fractional Power

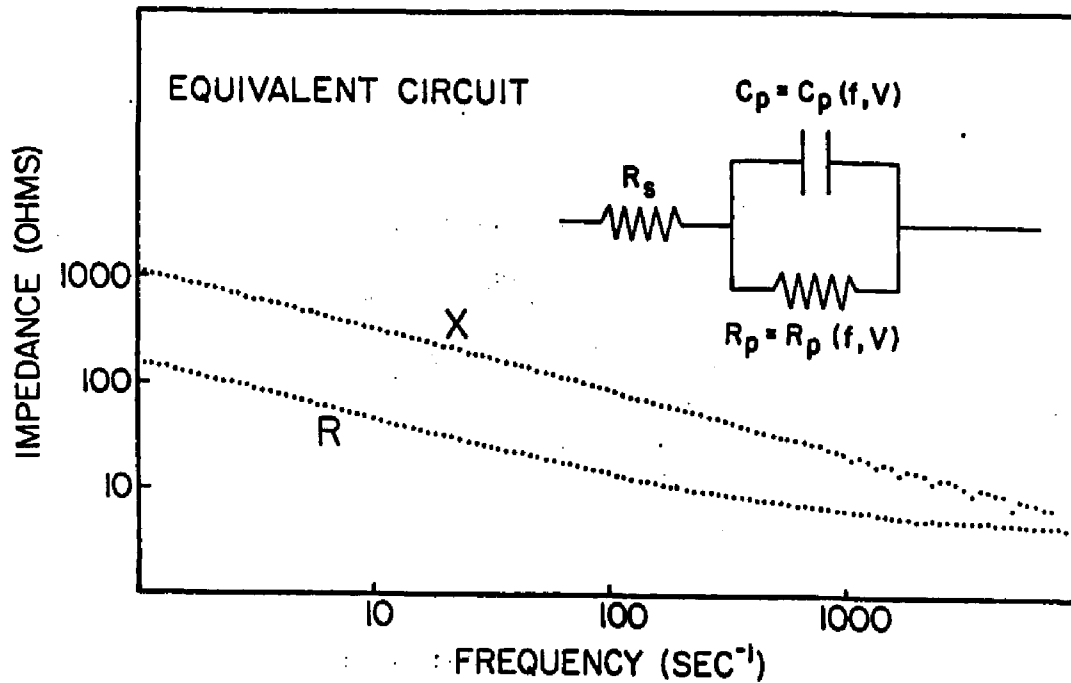


Figure 2.5 - A.C. impedance measurement of polycrystalline CdSe showing that both the real (R) and the imaginary (X) components of the impedance present a power-law frequency dependence. The CPA behavior is evident at low frequencies where R and X have identical frequency scaling exponents.

Frequency Dependence (FPFD)<sup>44</sup>, with little new theoretical light shed on the subject. (We will hereafter refer to this behavior as CPA behavior; the constant phase angle refers to the fact that both the real and the imaginary parts of the impedance scale with the same exponent.)

Lyden, Cohen and Tomkiewicz, in an experimental study on the impedance characterization of polycrystalline CdSe, demonstrated a characteristic scaling (CPA) behavior of the impedance as a function of frequency<sup>45</sup> (Figure 2.5). The authors identified this as a percolation-type behavior which they traced directly to the polycrystalline semiconductor electrolyte interface, with the critical exponents within the range of the Imry-Bergman treatment (see Equation 1.9). The percolation behavior may be associated with the morphology of the system, an analysis which arises from the identification of the interface region as a composite medium. However, the model that was employed<sup>46</sup> to explain the percolation mechanism is very specific to that system and has little general applicability. Despite this fact, the work is most noteworthy because it is a first attempt at applying the ideas of the study of composite media to the porous electrode and rough surface problem.

#### E. ...Which Brings Us Back to Fractals

Very recent studies have shown that CPA behavior arises as a natural consequence of a fractal interface structure.

Liu<sup>47</sup> has constructed a model electrode-electrolyte interface out of a "textbook" fractal structure - a Cantor bar<sup>48</sup> (Figure 2.6a). He assumes that the interface is ideally polarizable and that each "prong" in the bar can be represented by a series R-C element that scales with the "prong" width (Figure 2.6b). By solving for the total impedance of this equivalent circuit he obtains a CPA behavior. The exponent  $\beta$  that he measures (Equation 2.13) is related to the fractal dimension of the bar by  $\beta=1-D$ . It should be noted that the shape of the impedance curve that he obtains is as follows (In chapter 3 we will show very similar results from the Random Network model): On a plot of  $\text{Log}(Z)$  vs.  $\text{LOG}(\omega)$ , at low frequency the real impedance is a constant value, and the imaginary impedance is of slope  $-1$ . In mid-frequency both the real and imaginary slopes are  $-\beta$ . Finally at high frequency the real impedance is again constant, and the imaginary impedance has a slope of  $-1$ . Liu gives a very intuitive explanation of the scaling in the mid frequency range. If we view the a.c. signal in light of it's penetrability (see the description of penetration depth above) then the lower the frequency the finer is the length scale that is spanned by the current. In the limit of high frequency, the current is unable to penetrate the pores at all due to the large length scale that the current averages over. At low frequencies the current is able to penetrate everywhere due to the fact that it scales very microscopic lengths. It is in the mid-frequency regime in which the length scale of the "current yard stick" is varying, and that is when it is measuring the interface length with varying length yardsticks and the

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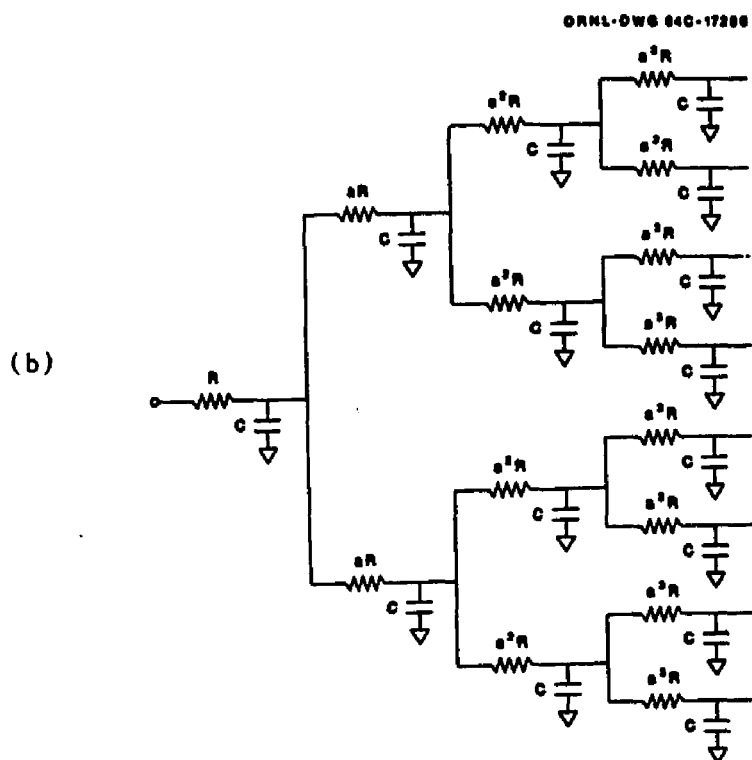
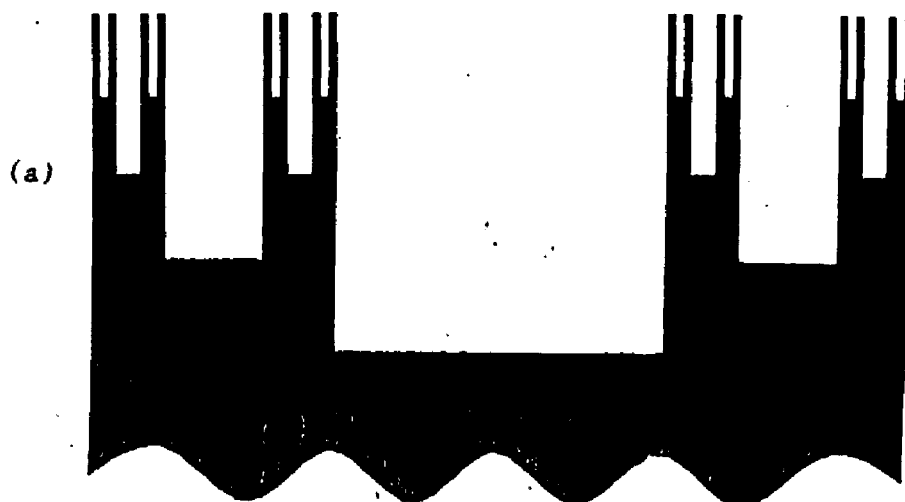


Figure 2.6 -

(a) Liu's cantor bar model of the electrode-electrolyte interface with a fractal surface. The electrolyte is the dark area.

(b) Equivalent circuit representation of the cantor bar geometry.

(From Ref. 47, reprinted by permission of the author.)

self-similarity ratio emerges directly, as in the Richardson curve of Figure 1.3.

More generally, Nyikos and Pajkossy<sup>49</sup> have shown how resistance and capacitance scale with length (for a self-similar distribution of R-C networks), so that when the total admittance is calculated from the combination of R and C it scales as  $\omega^\beta$ , where  $\beta$  is related to the fractal dimension by

$$\beta=1/(D-1) \quad . \quad (2.14)$$

This treatment builds on many of the assumptions in our Random Network Model except that no specific model structure is chosen; the authors simply argue that one can choose a fractal surface that may be represented by the self-similar R-C network, and calculate from there. We will consider one specific possible implementation of this idea in chapter IV.

The work of LeMehaute<sup>50</sup> is also noteworthy; he assumes that most real life electrochemical systems have a fractal morphology on the interface, and derives the contributions of a fractal interface using a highly mathematical TEISI model. In this treatment he computes fractal time derivatives of kinetic phenomenon and illustrates scaling relationships.

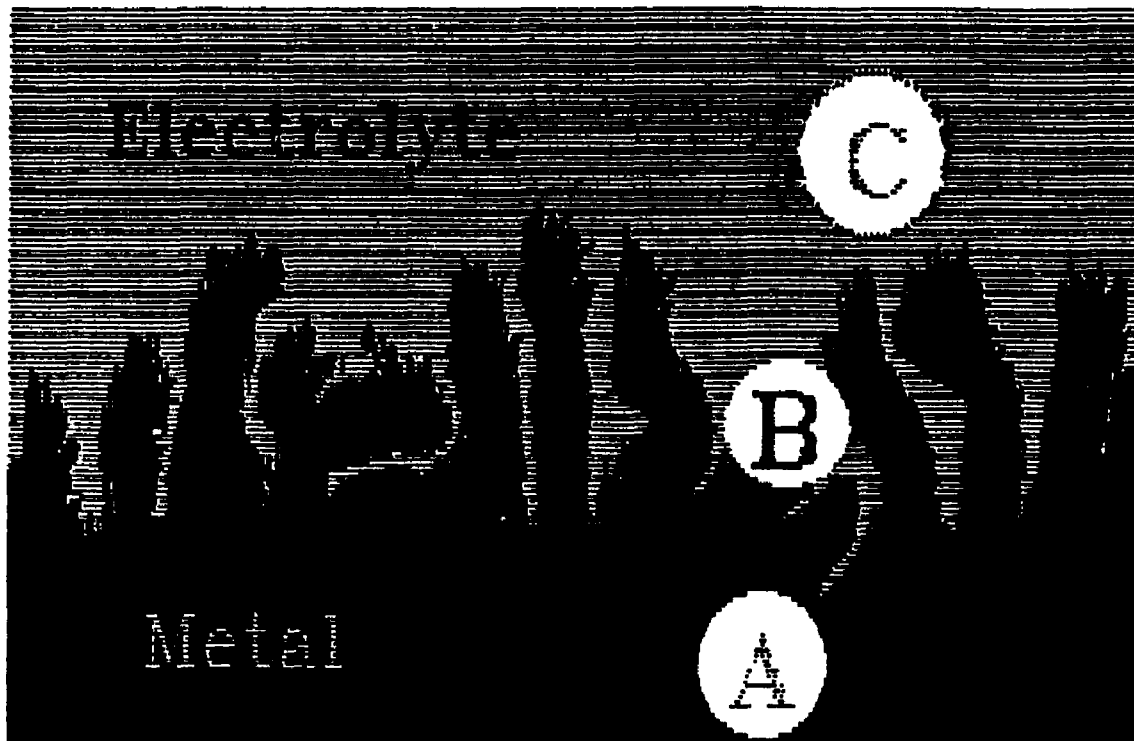
The DeLevie model is a convenient one dimensional description of trends in the a.c. impedance behavior of porous electrodes. What the model lacks is the morphological sophistication of real-life systems. Similarly, the treatment of Liu has built in to it a realistic model of a rough interface, but it is still two dimensional and not connected with real structures. In the next section, we present our Random Network Model, a theoretical treatment of porous electrodes that mimics the three dimensional heterogeneous distribution of metal and electrolyte, while still providing a means of exhaustive calculations of its a.c. impedance behavior.

### III. Porous Electrode Simulation

#### A. The Random Network Model

The Random Network model is designed to merge the techniques of handling conduction in inhomogeneous media (chapter I) with the theory of the porous electrode-electrolyte system (chapter II). The result is effectively a four phase inhomogeneous system composed of air, electrolyte, metal, and semiconductor. (In this chapter we refer to the phase that is present on the interface as a semiconductor, although following the theory presented in Chapter II the same electrical properties of the interface would arise from the existence of the metal-electrolyte interface.) The electrode problem may actually be divided up into two subcategories: a) The rough electrode is a description of the entire system consisting of the bulk electrolyte and the bulk metal of the electrode, and everything in between (the whole of Figure 3.1). b) The porous electrode is a description of the system from the point of view of a simple heterogeneous distribution of metal and electrolyte, (area (B) in Figure 3.1), and is the focus of this study.

Following the methodology of Equations 1.3-1.5, we assume that our electrode system is composed of a heterogeneous mixture of metal, electrolyte and semiconductor microscopic components. These components are assumed to be large enough to be represented by an average resistivity or complex dielectric constant, the same values as for bulk quantities of the materials. We use the equivalent circuit representations introduced in Chapter II to represent the electronic



**Figure 3.1**

Schematic of a rough electrode surface. Regions A and C are comprised of bulk metal and electrolyte respectively, while region B may be viewed as a composite inhomogeneous system.

properties of these phases. Our objective is to obtain the total macroscopic impedance of the system. With a detailed knowledge of the system's composition (since after all, we built it ourselves), we set out to correlate the impedance information with the system's microscopic characteristics. Due to the charge storage behavior of the insulating phase (semiconductor) on the metal-electrolyte interface, the system's impedance is frequency dependent, and much useful information is hypothesized to lie in the impedance vs. frequency relation. This theoretical study mimics the a.c. impedance characterization of electrochemical systems that is widely utilized experimentally<sup>32</sup>. It is important to note that this simulation is not meant to model the d.c. operation of the electrode and to follow in detail the mechanisms of Equations 2.8-2.10, but merely to simulate the electronic behavior of the porous electrode system under small signal a.c. excitations at any given in moment in time.

### B. The Algorithm

We define a three dimensional cubic lattice,  $A(i,j,k)$ , where  $i, j$  and  $k$  range from 1 to  $N$ ,  $N$  being the total number of nodes.  $N$  is chosen so that it is sufficiently large to approximate an infinite system. Construction of the electrode is simulated using a random number generator to place "metal" at various points in the lattice, until the desired porosity is reached (plane  $k=N$  is all metal as in the back electrode contact). A check is made of the resulting electrode to insure that it is continuous and that no pieces of

metal are "hanging" in mid-air. Any disjointed pieces of metal are removed, and are randomly replaced on the remaining available lattice points. This process is repeated until a continuous electrode of the desired porosity is obtained. Lattice plane  $k=1$  is defined as electrolyte, to represent the solution; the electrode itself begins at lattice plane  $k=2$ . The electrolyte is allowed to "seep into" the pores in the electrode until all accessible pores are filled, by searching for continuous electrolyte paths from the  $k=1$  plane throughout the electrode. Any point on the lattice which has been left empty (i.e. no metal was placed there, and electrolyte was precluded from penetrating to that site) is considered to be "air".

The geometric assumptions in this model are that the various components consist of volume filling cubes centered on the lattice points  $A(i,j,k)$ . Two adjoining cubes of metal (or even two on the diagonal with each other) prevent the electrolyte from seeping past them. The result of this constraint is that the fluid flow through the system shows classic percolation behavior. If the electrolyte is introduced at  $k=1$ , then at low porosities it flows only a small part of the way through to  $k=N$ . At porosities above the percolation threshold the electrolyte does indeed flow throughout the system. There is, in fact, a whole body of literature that includes this complication of pore connectivity in an analysis of electrode structure.<sup>54</sup> We have minimized these effects by limiting our consideration of the system to be above the fluid flow percolation threshold. An alternative to this approach would be to consider the

system components as closely packed spheres that permit fluid flow in the interstices, which effectively eliminates all "air" in the system (electrolyte flows everywhere that there is an absence of metal). This introduces discontinuities in the electrolyte connectivity as far as the cubic lattice is concerned, and great pains must be taken to insure that the computer will recognize electrical connectivity between islands of electrolyte. This can be done using a more laborious algorithm for calculating the electrical conductances, but we have not done so in this phase of the work.

Each lattice point represents a node in a three dimensional circuit network (See Figure 3.2), and each pair of nodes defines a circuit branch in the network. The impedance of each branch is determined by the characteristics of the two surrounding nodes. Thus, for example, if  $A(1,1,1)$  is electrolyte and  $A(1,2,1)$  is also electrolyte, the circuit branch defined by those nodes will consist of two series resistors of resistance  $R_e$  (the resistance of a microscopic section of the electrolyte). If  $A(1,1,2)$  is metal and  $A(1,2,2)$  is metal, then the circuit branch defined by those two nodes will be two series resistors of value  $R_m$  (the resistance of a microscopic section of the metal). Finally, the branch defined by  $A(1,1,1)$  - electrolyte, and  $A(1,1,2)$  - metal, is represented by an  $R_m$  and an  $R_e$  resistor in series, with the addition of a parallel R-C element in series with them to represent the semiconductor-electrolyte interface that results (see Figure 3.2). We refer to "semiconductor" on the interface, although the interface could more generally have an insulating layer or a simple Gouy-Chapman layer on

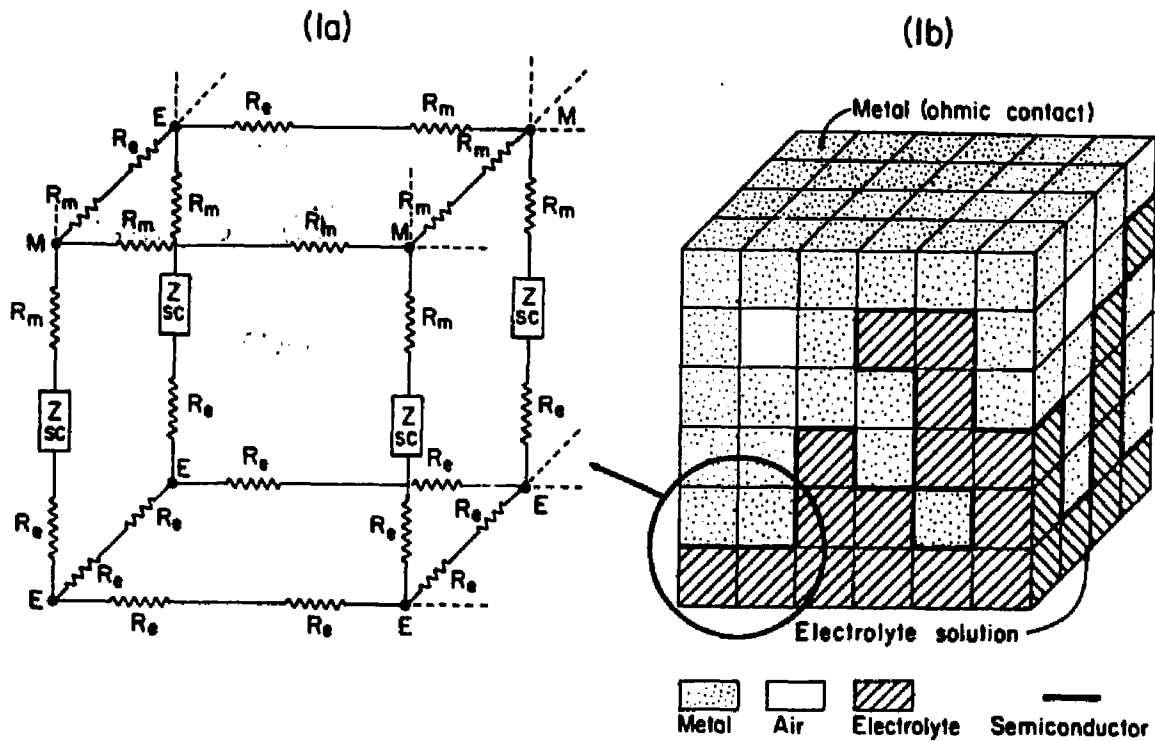


Figure 3.2 - The Random Network Model of a porous electrode. Figure 3.2b shows our idealized picture of the electrode's structure (the metal was distributed using a random number generator), and 3.2a shows a portion of this structure represented by a network of circuit elements.

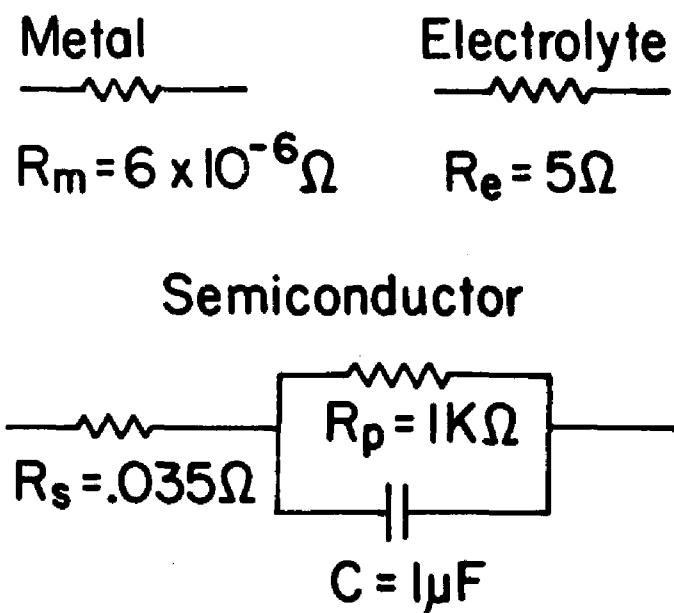


Figure 3.3 - Equivalent circuit representation of the three different phases in our electrode system, and typical values chosen to represent physical processes in realistic systems. Note that the interface is described as semiconducting, and appropriate circuit element values were chosen to reflect this. To represent an insulating phase on the interface these values may be suitably altered.

it. Any circuit branches leading into nodes that are defined as "air" are taken to be of infinite resistance.

A typical set of values that were chosen as unit impedances for the individual components of metal, electrolyte, and semiconductor are presented in Figure 3.3. These values were obtained by taking the magnitudes of bulk impedance for  $1 \text{ cm}^3$  sized samples, (we chose numbers characteristic of Zn and ZnO). In the interest of generating physically meaningful numbers, we defined a unit pore of arbitrary diameter  $10 \mu\text{m}$  and scaled the impedance values to represent the dimensions of the unit pore. In effect we consider each of the microscopic components (metal or electrolyte) as being comprised of a cube whose side is of length  $10 \mu\text{m} = 10^{-3} \text{ cm}$ . When we normalize the  $1 \text{ cm}^3$  values given in Figure 3.3 to this shrunken size, we have to multiply the impedance of the  $1 \text{ cm}^3$  "resistor chunks" by a factor of  $10^3$ . This is due to the fact that resistors have a dimensional dependence of  $\rho/A$  where  $\rho$  is the longitudinal length and  $A$  is the cross-sectional area. The determination of the scaling factors for the input parameters that represents the semiconductor is not so straightforward. As far as the geometry of the model goes, the semiconductor-electrolyte interface is "pure" surface area with no thickness (i.e. we do not devote any lattice coordinates to the interface- the computer just determines where it is and calculates an enhanced impedance corresponding to the semiconductor on the interface at that point). The thickness of the semiconductor deposit is implicit in the values that were chosen for  $R_p$ ,  $R_s$ , and  $C$ . (The numbers chosen in Figure 3.3 approximate a 100 Angstrom

layer of moderately doped semiconductor of area  $1 \text{ cm}^2$ ). Thus, when we normalize the components from  $1 \text{ cm}$  to  $10^{-3} \text{ cm}$ , we multiply  $R_s$  and  $R_p$  by  $10^6$  and divide the capacitance  $C$  by  $10^6$  because of the reduced surface area. These components are then assembled into a system, as described above (see Figure 3.2) and the total impedance of the system is calculated. The total impedance is then rescaled by an appropriate factor that normalizes the whole system to a  $1 \text{ cm}^3$  size. Specifically, for an,  $11 \times 11 \times 11$  system there is  $10 \times 10^{-3} \text{ cm}$  per side. Thus, we scale the total calculated impedance by  $10^4$ . This final normalization assures that no matter what size we choose for our lattice size, the numbers will be comparable.

Once the electrical network has been defined, the computer sets up the Kirchoff's law equations,

$$G \cdot V = I \quad (3.1)$$

where  $G$  is the conductance matrix (see Equation (1.3); here:  $G_{ii} = \sum_j g_{ij}$  and  $G_{ij} = -g_{ij}$ ),  $V$  is a vector containing the voltages on each node, and  $I$  is a similar vector containing the net current inputs to each node. Due to the fact that our model includes reactive circuit elements, the quantities  $G$ ,  $V$ , and  $I$  in Equation 3.1 are complex, and may be represented as  $2N-1$  equations of the form:

$$\begin{pmatrix} G_r & -G_i \\ G_i & G_r \end{pmatrix} \begin{pmatrix} V_r \\ V_i \end{pmatrix} = \begin{pmatrix} I_r \\ I_i \end{pmatrix} \quad (3.2)$$

where the subscripts  $r$  and  $i$  stand for the real and imaginary

components respectively.

A known current is sent uniformly into all the bottom nodes, which is designed to insure an equipotential surface on the bottom plane. We have verified that the resulting voltage fluctuations on the bottom plane are in fact very small. The impedance of the system is calculated between one node on the electrolyte plane (1,1,1) and one node of the top electrode plane (N,N,N) by solving for the V's on all the nodes. For a system of  $N=15$  (a lattice of size  $15 \times 15 \times 15$  with 3375 nodes) there are up to 6748 equations to be solved. This is a formidable computer problem. Indeed, the task would be prohibitive without taking advantage of the fact that the conductance matrix,  $G$ , is a sparse-symmetric matrix.

A number of techniques exist for the solution of this class of problems. A widely used method for solving the simultaneous equations in the Random Resistor lattice is the Gauss Seidel iteration procedure with over-relaxation<sup>6</sup>. This is a very economical method, in both execution time and storage space. A serious drawback of this technique is the relatively strict requirements that it places on the coefficient matrix in order for convergence to be achieved, namely that the matrix be either diagonally dominant, or at least positive definite<sup>51</sup>. For Kirchoff's law problems with purely resistive components, diagonal dominance is assured, since the diagonal elements of the conductance matrix are simply the sum of the off diagonal elements. In our problem, with complex impedances, we find that at frequencies for which the real conductance

approaches the same order of magnitude as the imaginary conductance, the Gauss-Seidel procedure does not converge. This is due to the form of the Equations 3.2 where there are many additional off-diagonal elements due to the  $G_i$  values. When these imaginary components are large, the matrix is no longer diagonally dominant, and becomes indefinite. For most of our computations, we have resorted to the use of a Gaussian elimination routine which takes advantage of both the sparseness and the symmetry of the G coefficient matrix<sup>13</sup>. Only the nonzero elements in the upper half triangle of the matrix are stored, and an efficient pivoting strategy is chosen to minimize non-zero matrix fill during the pivoting and to minimize the number of multiplications required in the solution. In practice, the technique is approximately an order of magnitude more expensive to use than Gauss-Seidel in both speed and storage requirements. The advantage of using the Gaussian elimination technique is that a solution is guaranteed for all but the most ill-conditioned problem. The solutions obtained from the Gauss-Seidel procedure (in the range in which convergence is reached) are in complete agreement with the results obtained using the modified Gaussian elimination technique.

The final step in "fine tuning" the model is to choose a minimum lattice size that provides some degree of statistical significance but is still within reasonable limits of available computer time. If the random network model is a true statistical model then the calculation should not be sensitive to the lattice size of the system. Thus the minimum lattice size was chosen by solving a

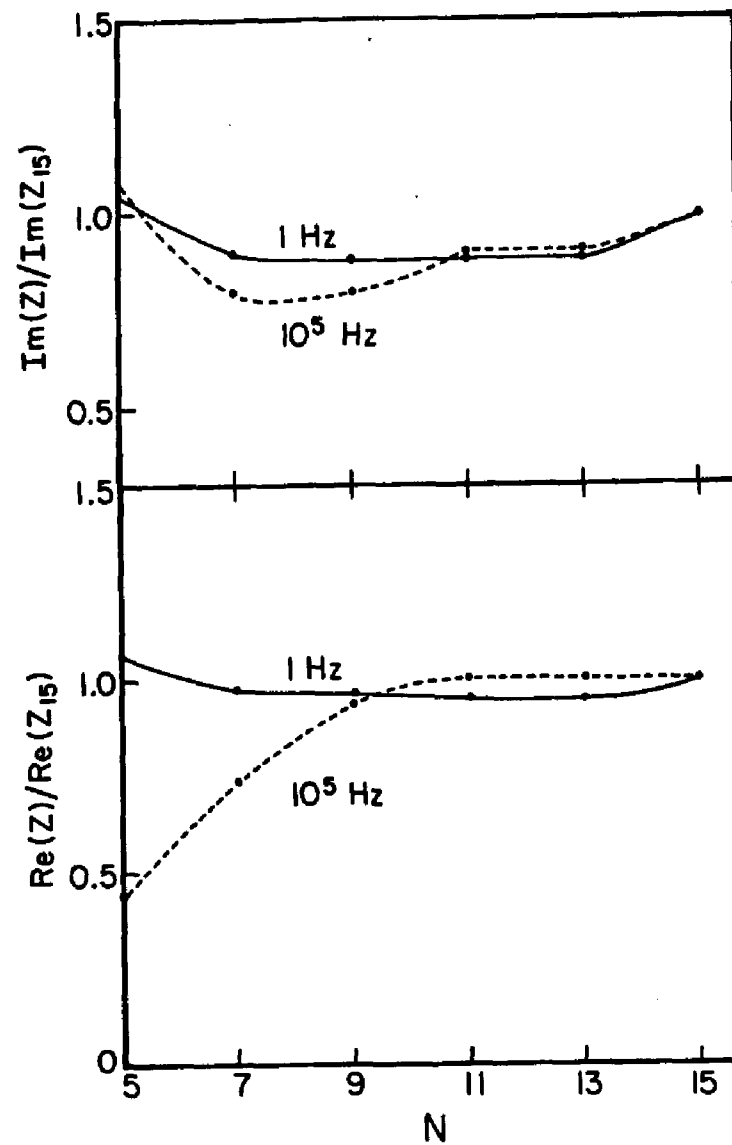


Figure 3.4 - The impedance of the system in the RNM as a function of lattice size. Although the solution is converging, numerical limitations prevented us from solving for larger systems, and some statistical deviation is assumed to result.

single problem on increasingly large lattice sizes until the answer converges to a fixed value. The results of this study is presented in Figure 3.4. There are two significant difficulties that we encountered in trying to solve for the impedance of our system. Firstly, we were not able to eliminate all "dead end" conductive paths as Kirkpatrick did in his composite media simulations. He was able to do this because all he was concerned about was the system's degree of connectivity. Thus , he was able to choose only the cluster's "backbone" and measure the resistance of that to decide on the conductivity of his system. In our system, the capacitance of all surfaces contributes to the impedance, and it is not clear which current paths may be discarded, if any. Secondly, as already mentioned, the complex impedance doubled the size of the network, and turned the problem from a well-behaved calculation into an ill-conditioned one. For most of the results in this study a system of size  $11 \times 11 \times 11$  was used. Computational limitations forced us to choose this minimum size, although continuous boundary conditions were employed to minimize finite-size effects.

### C. Homogeneities & Systematic Correlations

From the foregoing description, it is obvious that our model differs in a number of important ways from the purely random conductor-insulator distributions of Chapter I. For one thing, although the distribution of metal is governed by a random number generator, we have modified the random statistics to require a connectivity in the metal for a realistic representation of an electrode (although some metal powder - electrolyte electrode

systems have been experimentally explored). One effect that this has on the electronic properties of the system is that metal connectivity is assured across the system - it always lies above the percolation threshold. Another constraint introduced is the fact that the three phases electrolyte, semiconductor and metal are not mixed homogeneously, but are distinctly separated by an orderly juxtaposition to each other (see Figure (3.3) for a schematic representation of this). Even in the pores where the paths are contorted and the surface rough and complicated, there is always a continuous distribution of electrolyte and metal respectively on either side of an interface. It is thus impossible to formulate an effective medium theory for this arrangement. Any treatment of local inhomogeneity in an average medium such as in chapter I cannot be made to preserve this structured distribution around an interface.

The RNM simulation is a valid approximation of conduction in a porous electrode system, and superior to previous treatments of electrodes precisely because it preserves the stochastic nature of the system to the extent that it is present. Thus far, that aspect has not been properly treated in the literature.

It may be more precise to say that, unlike the conductor-insulator problem which presents a bulk inhomogeneity in two or three dimensions, the porous electrode system is a problem of a quasi two dimensional inhomogeneity of the interface. Although the system's bulk component composition will also exhibit bulk properties and its

behavior will be governed by these bulk-related phenomena, it is the properties due to the random morphology of the rough surface that is of greatest interest to us, and we will attempt to show when these interfacial contributions dominates the system's properties. We define the surface (or interface) in the model as being those metal sites that are adjoined by one or more electrolyte sites.

#### D. The Model Parameters

The input parameters of the model consist of the porosity,  $p$ , the electrolyte resistance,  $R_e$ , and the metal electrode resistance,  $R_m$  (all bulk impedance values are for a typical  $1 \text{ cm}^3$ ). By varying the system input parameters, we can simulate various physical conditions in our electrode system. Variations in  $R_e$  correspond to different electrolyte concentrations.  $Z_{sc}$ , the impedance of the semiconductor is represented by a resistor,  $R_p$ , in parallel with a capacitor,  $C_p$ , which are both in series with a resistor,  $R_s$  (Figure 3.3). Varying these parameters correspond to variations in the state of charge of the electrode (i.e. the thickness of the semiconductor buildup), and the doping level of the semiconductor buildup (determined by the reaction mechanisms at the electrode surface during electrode discharge). The resulting (frequency dependant) impedance of the interface is:

$$Z_{sc} = R_s + \frac{R_p}{1 + (\omega\tau_p)^2} - j \frac{\omega R_p^2 C_p}{1 + (\omega\tau_p)^2} \quad (3.3)$$

where  $\tau_p = R_p C_p$ .  $R_s$  corresponds to the resistivity of the bulk semiconductor,  $R_p$  to the faradaic current leakage through the

interface (a space charge region if it is a semiconductor), and  $C_p$  to the capacitance of the interfacial charge storage region. We obtained the  $C_p$  value by assuming a semiconductor buildup of approximate thickness  $l'=100 \text{ \AA}$ , an area of  $a=1 \text{ cm}^2$  and the relationship

$$C = \frac{\epsilon_0 \epsilon a}{l'} , \quad (3.4)$$

Where  $\epsilon_0$  is the permittivity of free space and  $\epsilon$  is the dielectric constant of the material on the interface (we chose  $\epsilon=10$  representative of a ZnO deposit). As previously mentioned, the primitive size of our lattice was taken to be  $10\mu$ , such that for a lattice of size  $11 \times 11 \times 11$  we have a cube of side  $100\mu$ .

#### E. Model Output- Bulk Contributions

Figure 3.5 is a typical result from the electrode RNM at a given  $p=.4$ , using the input values of Figure 3.3. We may loosely categorize the imaginary impedance vs. frequency behavior as being composed of three regimes: At low frequency ( $f < 100 \text{ Hz}$ ) we have a slope 1 behavior on the log-log plot, at high frequency ( $f > 10^5 \text{ Hz}$ ) we see a slope (-1) dependence, and in the middle frequency regime is some sort of transition region or regions. The low and high frequency behavior is the same as would be observed from measuring the impedance of an R-C circuit, however, when an attempt is made to calculate the capacitance, it is observed that the capacitance has different values at low and high frequency. The measure of capacitance is usually taken as an indication of a measure of some active interfacial area, and thus it would appear that there are two

# IMPEDANCE VS. FREQUENCY-P192

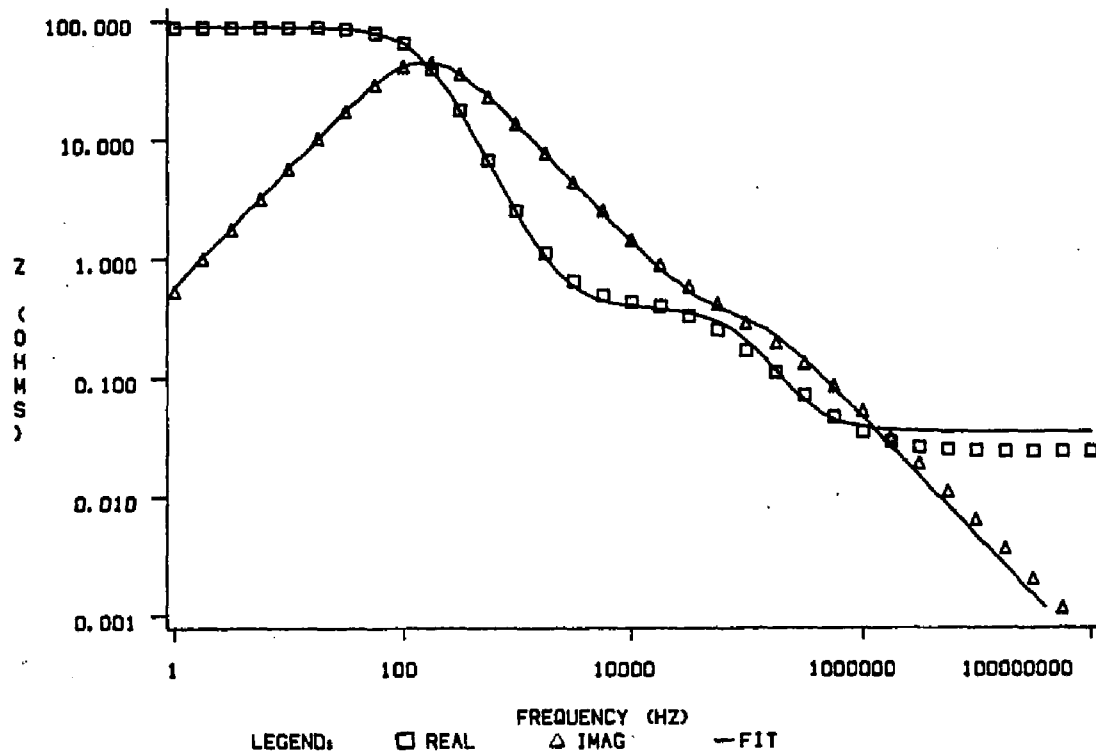


Figure 3.5 - A typical data set from the RNM computation. The data points are from the simulation, and the solid line drawn through the points is a fit to the data (see section III.G).

characteristic areas being measured in this system; one at high frequencies and another at low frequencies.

DeLevie<sup>36</sup> had suggested that for cylindrical pore electrodes the penetration depth of the ac signal decreases with increasing frequency (see Chapter II). If we accept this as applicable to our electrode model, this would account for the lower capacitance being measured at higher frequencies- the current is not penetrating far into the pores and hence the current is scanning less area. It is easy to verify that in fact that is what is occurring in our RNM electrode. Figure 3.6 depicts the current distribution as it flows through the semiconductor (labeled S), electrolyte (E) and metal (M). These values were obtained by actually calculating the current flow throughout the nodes in our model. At low frequency (Figure 3.6a) all the semiconductor interfaces are passing an appreciable amount of current; all of the electrolyte paths are contributing to the current flow as well (in order for the current to reach all the interfaces it must go through all the electrolyte). In contrast to this, at high frequency (Figure 3.6b) we note that only a small minority of all the semiconductor interfaces are passing any current at all.

Figure 3.1 schematically depicts the process. The current flows through the bulk electrolyte (labeled C) and tries to get through to (A), the metal of the electrode. Before it can do that, it must pass through the interface (labeled B). How (and where) it passes through the interface is the determining factor in the ac

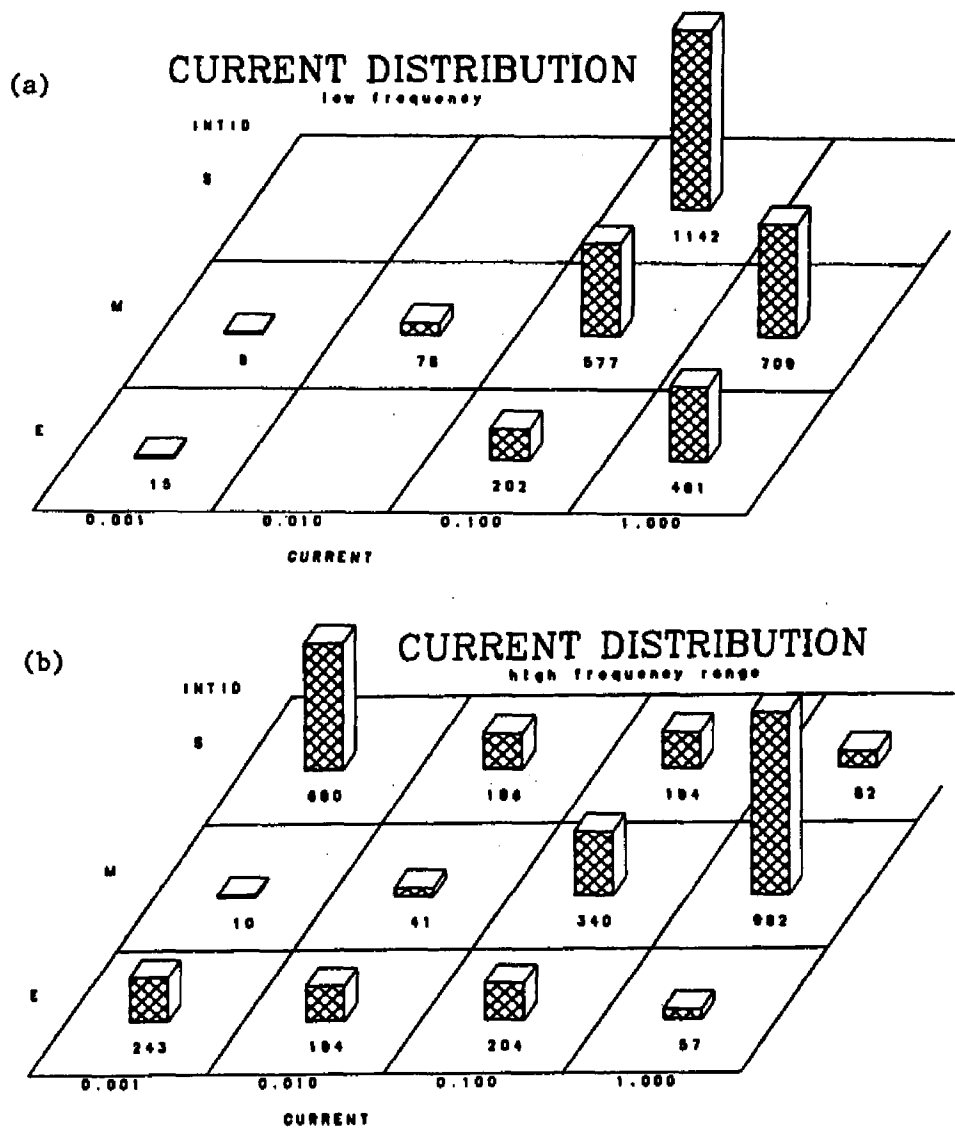


Figure 3.6

The bar charts depict the current distribution among the three internal phases in the electrode. The left axis corresponds to the three phases electrolyte, metal, and semiconductor. The bottom axis is the current magnitude (in % of input current), and the Z axis corresponds to an enumeration of how many of the microscopic components are carrying the stated amount of current. Thus, at low frequency (3.6a) all of the semiconductor interfaces are carrying a significant percentage (.1%) of the current because there is a large current flow deep into the electrolyte. At high frequency (3.6b), most interfaces and electrolyte are carrying almost no current, since the current flow shorts into the metal before traveling deeply into the pores through many electrolyte components.

capacitance. If the interface is an easier path for the current to go through than continuing through the electrolyte in the pores (since the pores may be narrow the resistance could be high), then the current will "short" right through (B) into (A) at it's earliest opportunity (near  $k=1$  in our model). If on the other hand, the journey through  $R_e$  is the least-resistive path, the current will travel through the electrolyte deep into the pores before being forced to enter the interface into (A).

The low frequency capacitance value would then be a measure of the total surface area of the electrode (at least as far as the current got), and the low frequency impedance data may be taken to be a measurement of the electrode surface area. Further verification of this may be obtained by increasing the electrolyte resistivity and watching if the value of  $C_{1f}$  changes as the electrolyte becomes more and more unsuited for performing this area measurement. Sure enough, Figure 3.7 shows how we can make the value of  $C_{1f}$  approach  $C_{hf}$  simply by raising the  $R_e$ . Conversely, we can raise the value of  $C_{hf}$  simply by lowering  $R_e$  which improves the penetrability of the current even at high frequency. Figure 3.7 shows this as well, but we were prevented from making  $C_{hf} = C_{1f}$  due to numerical difficulties.

To further test the current's area scanning capability, we varied the porosity and measured the corresponding  $C_{1f}$ . Figure 3.7 shows how closely the low frequency capacitance is able to follow trends in the roughness factor. (The roughness factor,  $\rho$ , is a measure of

## Capacitance vs. Electrolyte Resistance

(Porosity = .40)

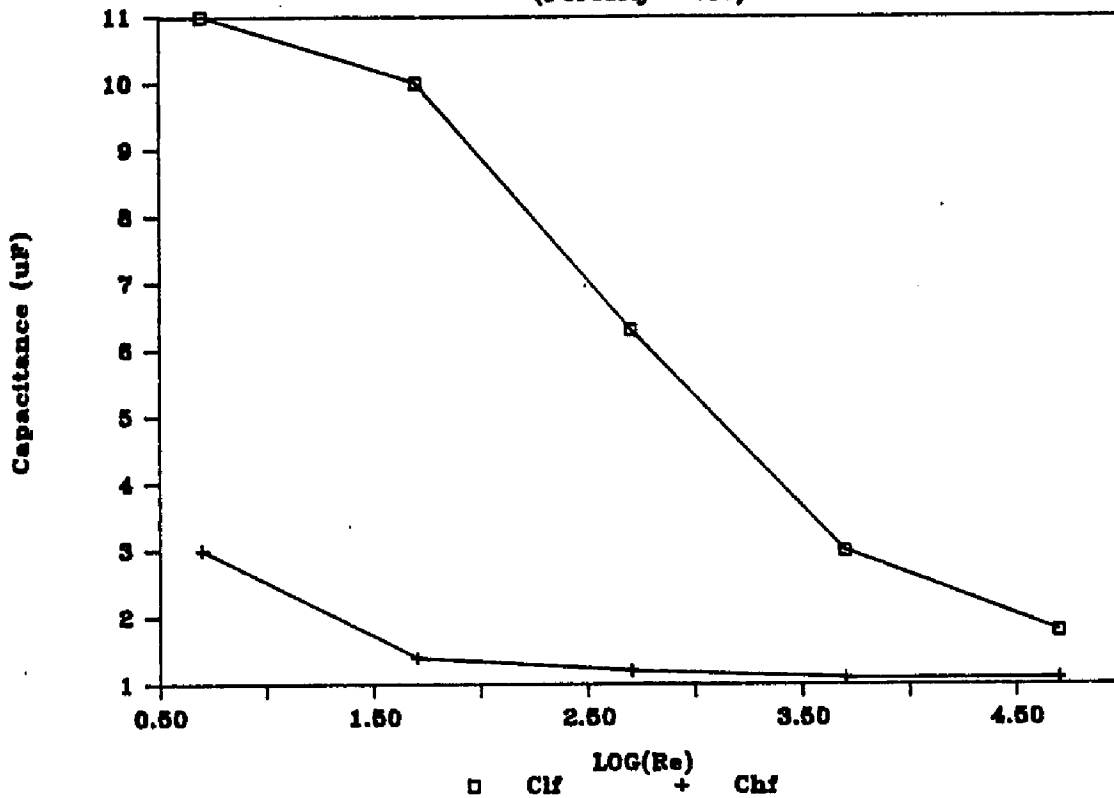


Figure 3.7 - The change in the measured low frequency and high frequency capacitance as  $R_e$  is varied. At high  $R_e$ , even the low frequency capacitance fails to measure the apparent surface area of the pores.

total interfacial area of a rough electrode to the geometric surface area- the area of a flat  $1 \text{ cm}^3$  electrode. The capacitance in our system is also normalized to a  $1 \text{ cm}^3$  value by our simulation so we may compare  $\rho$  and  $C_{lf}$  directly.) The value of  $\rho$  in Figure 3.8 was computed by simply counting up all the metal-electrolyte unit interfaces in the computer model for each porosity. Figure 3.8 is a conclusive demonstration of the utility of the value of the low frequency capacitance from the impedance measurement. At the same time it also demonstrates that  $C_{hf}$  is a measurement corresponding to the geometric surface area. Another interesting feature of the  $\rho$  vs. porosity plot is that  $\rho$  is not a unique measure of porosity. A roughness factor of .9 may be obtained by a porosity of 0.36 or 0.62 . The bell shaped curve is due to a competition of two opposing factors for the increased surface area. On the one hand, as the volume fraction of metal increases from zero, the more available metal surfaces there are for metal-electrolyte interfaces. On the other hand, as the volume fraction of metal increases beyond .5 or so, the probability of having a metal site bordered by another metal (and hence no metal-electrolyte interface) also increases!

To quantify the penetrability of the current with decreasing frequency, DeLevie had introduced the penetration depth,  $\lambda = (|Z|/R)^{\frac{1}{2}}$  (see chapter II; this R is related to our parameter  $R_e$ ). The concept should be applicable here as well, except that a single-valued penetration depth is of little meaning in this 3-D system, as opposed to DeLevie's one-dimensional pore. The current may penetrate deeply into wide necks of the pores, but get choked off in a

## Roughness and Low Freq. Capacitance as a Function of Porosity ( $R_e=50$ ohm)

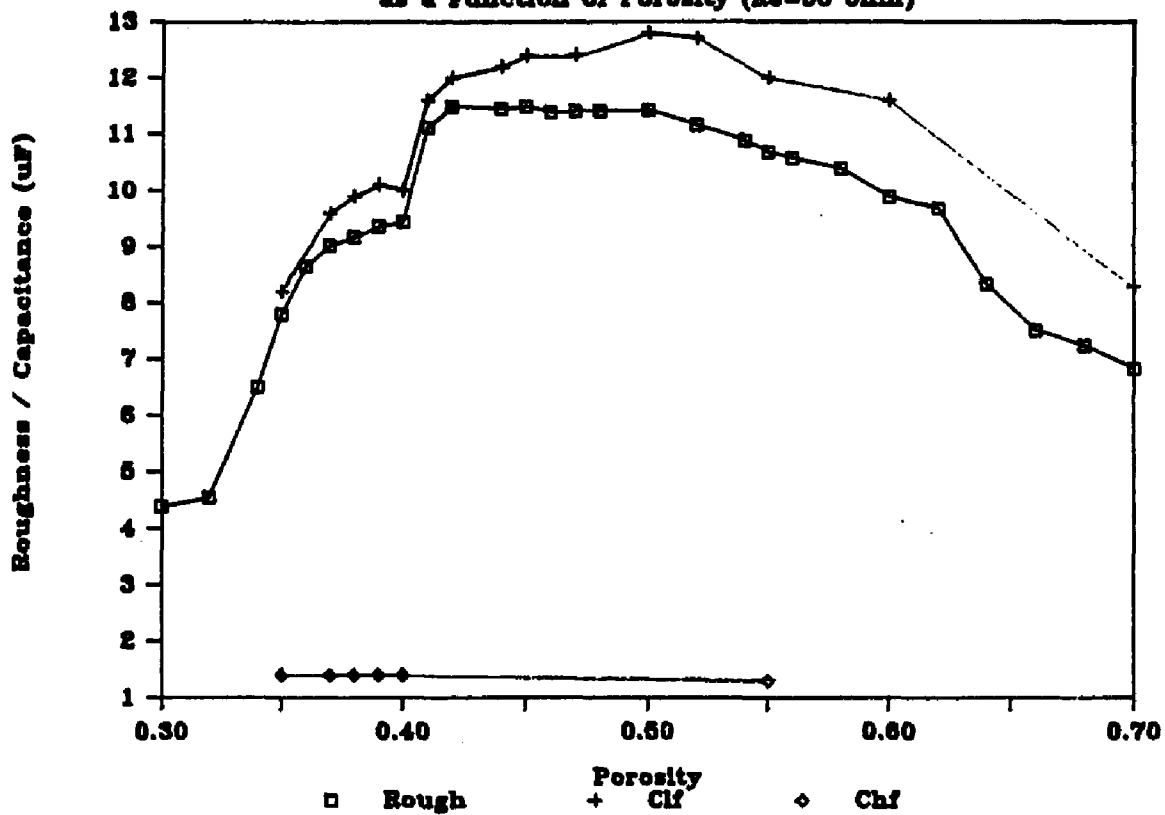


Figure 3.8 - A comparison of the actual surface roughness and the measured low frequency capacitance illustrates that the low frequency capacitance may be used to measure the apparent surface area of the pores. For comparison, the high frequency capacitance is shown, and it is not sensitive to changes in electrode surface.

narrow horizontal cross link. Nevertheless, the value of  $\lambda$  is useful if thought of as a measure of the current "penetrability". In Figure 3.9,  $\sqrt{Z_{sc}}$  is plotted as a function of frequency. We may draw a horizontal line across the figure corresponding to any value of  $\sqrt{R_e}$  that we choose to introduce into our system. At the frequencies that the  $R_e$  line falls below  $Z_{sc}$ , then the current will flow through the electrolyte. At frequencies for which  $Z_{sc}$  falls below  $R_e$ , then the current will short through the interface.

#### F. Duplicating the DeLevie results as a Special Case of RNM:

To demonstrate that our RNM is a more general case of the DeLevie model, we can numerically simulate DeLevie's cylindrical pore electrode by constructing uniform pores with our random resistors. The DeLevie parameters  $R$  and  $Z$  differ from our  $Z_{sc}$  and  $R_e$  due to the fact that the former quantities are normalized to the unit pore length. We may consider a resistance and impedance per unit length as (see Figure 3.10):

$$R = \frac{\rho}{A} \quad \text{and} \quad Z = \frac{\rho' \ell'}{4\sqrt{A}} \quad (3.5)$$

where  $A$  is the geometric area of the pore,  $\ell'$  is the thickness of the semiconductor layer, and  $\rho'$  is an effective (complex) resistivity of the semiconductor. Consider an ideally polarized interface where

$$|Z_{sc}| = 1/(\omega C)$$

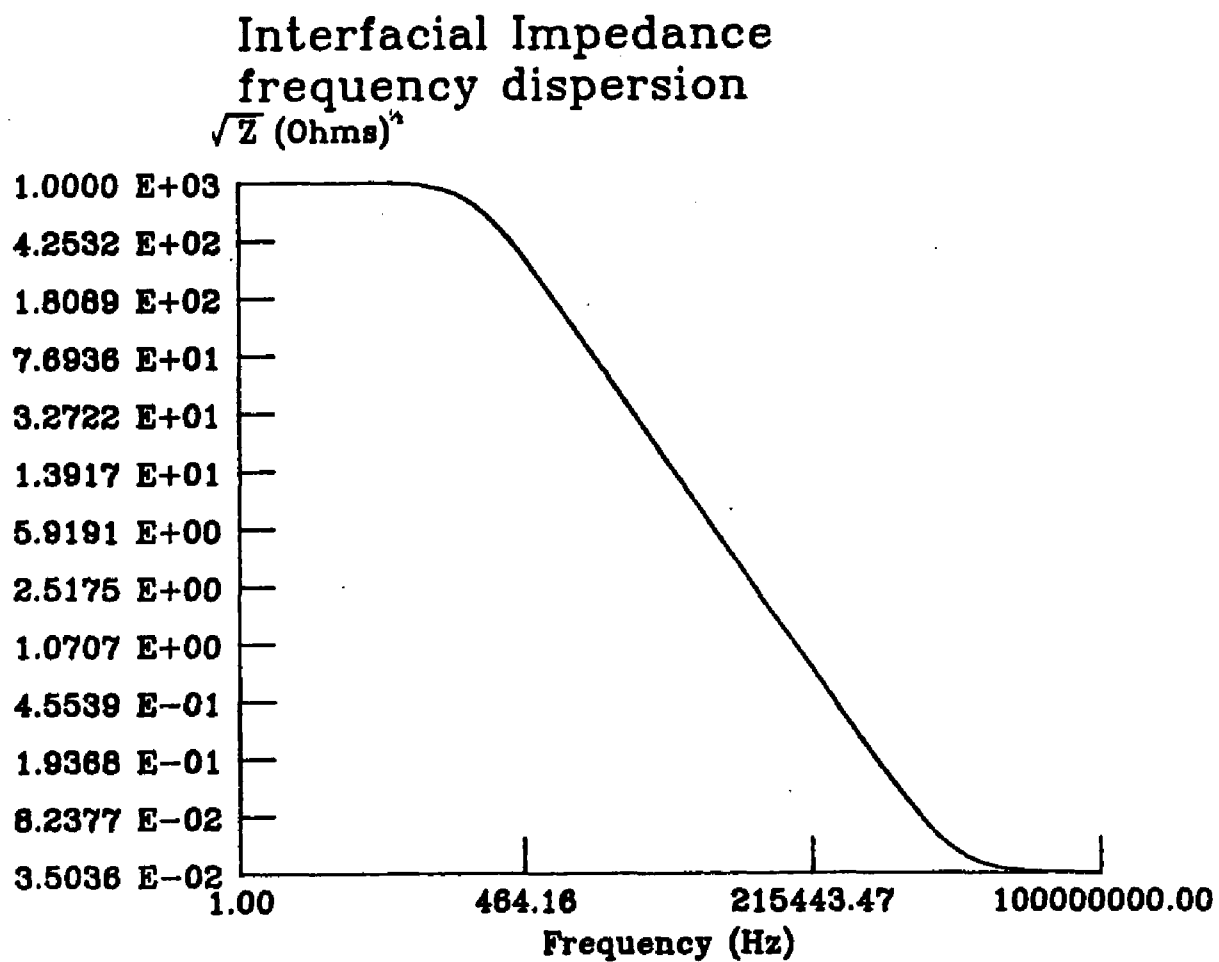
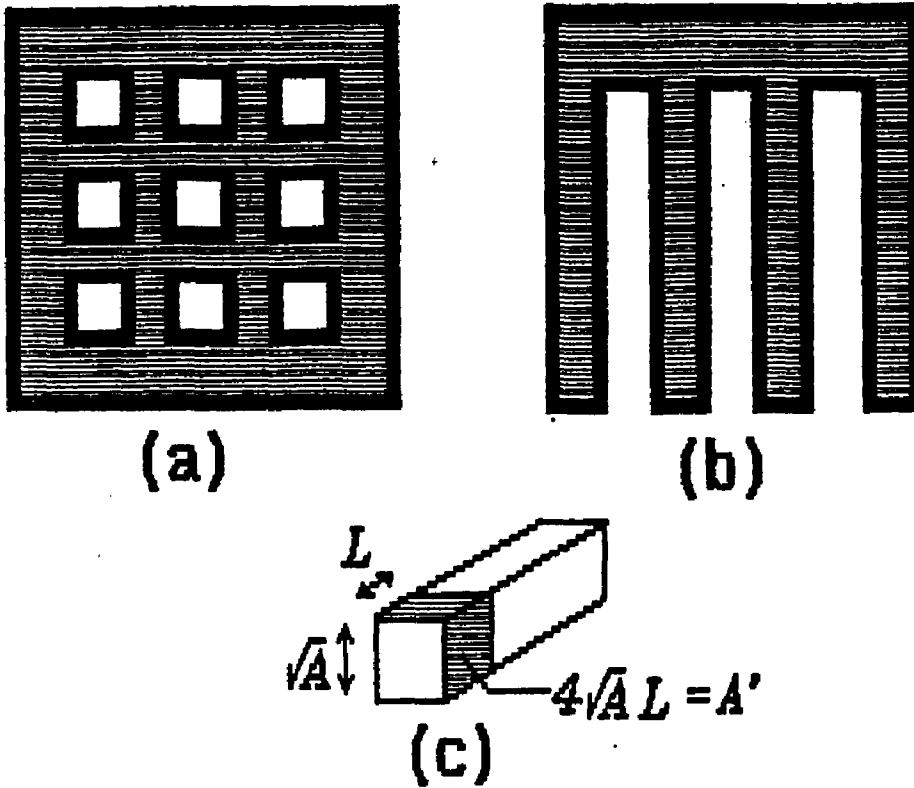


Figure 3.9- The impedance of the interface alone as a function of frequency.



## Figure 3.10

Rectangular pores set up to explore simple pore behavior. (a) is a view of the 9 pores in a "top view" cross section, and (b) is a "side view" cross section. (c) illustrates the dimensions of each pore.

and a dielectric solid, for which we may define

$$C = \frac{\epsilon_0 \epsilon a}{d}$$

where  $\epsilon$  is the static dielectric constant,  $\epsilon_0$  is the permittivity of free space. From Figure 3.9  $a=4\sqrt{A}\ell$ ,  $d=\ell'$  so that

$$Z_{sc} = \frac{\ell'}{\omega \epsilon_0 \epsilon 4\sqrt{A}\omega} = \frac{\rho' \ell'}{4\sqrt{A}} \quad (3.6)$$

Therefore the impedance per unit length is  $Z_{sc}/\ell$  and

$$\lambda = \left( \frac{Z}{R} \right)^{\frac{1}{2}} = \left( \frac{\ell' \sqrt{A}}{4\epsilon_0 \epsilon \rho \omega} \right)^{\frac{1}{2}} \quad (3.7)$$

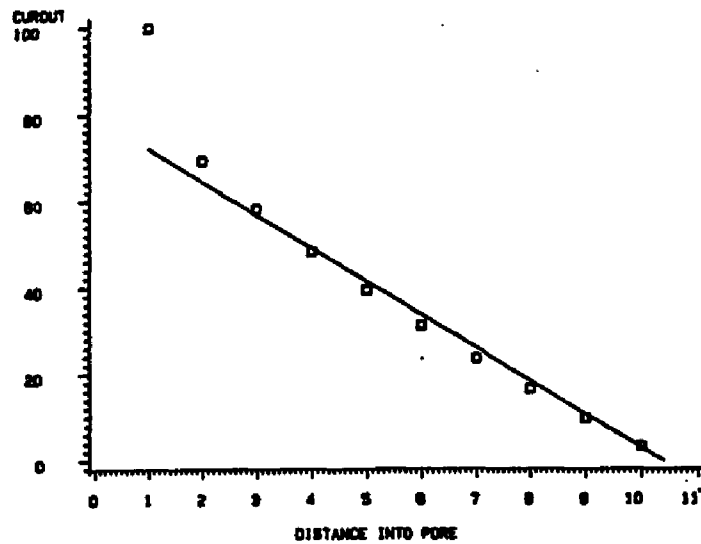
taking the values (as before),  $\ell'=100A=10^{-8}$  m,  $A=10^{-10}$  m, and  $\epsilon=10$ , we get a penetration depth of

$$\lambda = \begin{array}{ll} 2.8 \times 10^{-3} & \text{at 1 Hz} \\ 9.0 \times 10^{-5} \text{ m} & \text{at 1 KHz} \\ 2.8 \times 10^{-6} & \text{at 1 MHz.} \end{array}$$

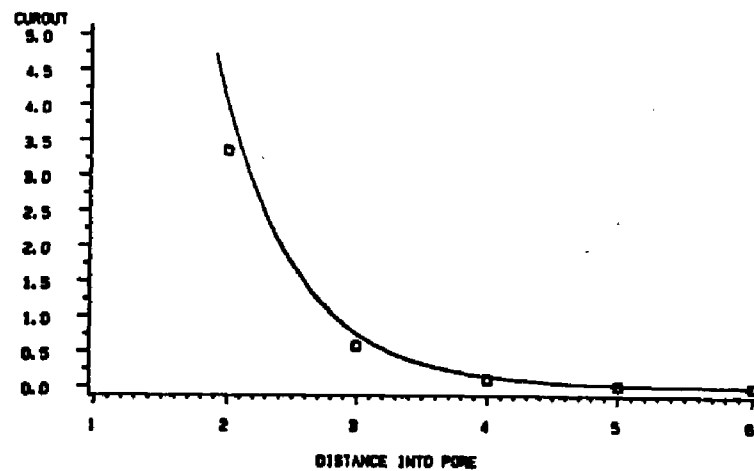
For our system of (as previously mentioned) size  $100\mu$  on a side, this corresponds to a current flow throughout the electrode at under 1 KHz, which is in agreement with the location of the low frequency regime in Figure 3.5.

To construct a simple cylindrical pore system with the RNM, we arrange our resistors so that they are distributed as in Figure 3.10, with the shaded area denoting the metal and the white area being the pores where the electrolyte flows. We solve for the current distribution in the system, and plot  $I$ , the current flow in

## CURRENT FLOW THROUGH PORE - LOW FREQ



## CURRENT FLOW THROUGH PORE - HI FREQ



LEGEND: □ DATA — THEORY

Figure 3.11- Magnitude of current flow into the pores as a function of distance, using the pore structure of Figure 3.10. The data points are the results of the calculation, and the line is a fit from the DeLevie model.

the Z direction, as a function of z, the distance into the electrolyte (Figure 3.11). Figure 3.11a is the result for high frequencies (1 MHz with an  $R_e$  of 1 K $\Omega$ ). The shape of the curve is exponential, and following DeLevie<sup>52</sup> we look for a result of the form

$$I = I_0 \exp^{-z/\lambda} \quad (3.8)$$

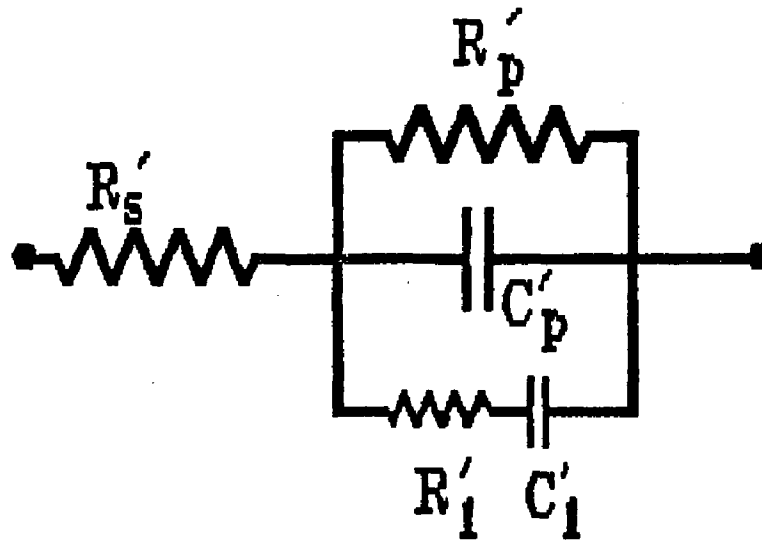
Upon fitting this function to the data of 3.11a, we arrive at  $\lambda=0.6$  which indicates that at high frequency the penetration depth is not even through to the first lattice point. Figure 3.11b shows the results at low frequency, and we may also try to fit that curve to Equation 3.8. However, if  $\lambda \gg z$  then the exponential may be expanded as

$$I_0 = I (1-z/\lambda) \quad (3.9)$$

We perform a linear fit of this function to the data in Figure 3.11b and we obtain  $\lambda = 10.4$ . This verifies the frequency behavior is as DeLevie postulated, and the exponential behavior of the current in the pores.

#### G. Other Features of the Impedance Curve

Returning to the impedance curve of Figure 3.4, we identify more features that may be correlated with the input parameters. As an alternative to the high frequency and low frequency capacitance that we have been using thus far to model the frequency response of the system, we can construct an equivalent circuit model for the system



**Figure 3.12**

Equivalent circuit model of the total electrode impedance.

that is valid at all frequencies. Figure 3.12 introduces some more circuit elements, but this time they correspond to the output parameters of the system (the total impedance calculated) and hence are denoted by the ' (prime) notation. The curve drawn through the data in Figure 3.5 is a theoretical fit to the impedance dispersion curve, using the circuit of Figure 3.12. By fitting the theoretical impedance curve, we evaluate the output parameters (the circuit element magnitudes).

We have varied the input parameters, and correlated that with changes in the equivalent circuit output parameters.  $R'_s$  is determined from the values of the real impedance at high frequency. Its value is usually governed (dominated) by the value of the input electrolyte resistivity,  $R_e$ .  $R'_p$  is determined by examining the system's real impedance at low frequency. The space charge resistance, input parameter  $R_p$ , of the semiconductor on the interface usually decides the value of  $R'_p$ . The peak in the imaginary component of the impedance occurs at  $\omega\tau_p=1$ , where  $\tau_p=R_p C_p$ . These dependencies can be altered. For example, if the value of  $R_e$  is made very small, then even at high frequencies it is the  $R_s$  of the interface, the bulk impedance of the insulator, that will dominate the high frequency  $R'_s$ . If  $R_e$  is made very large, then at low frequencies it is  $R_e$  that will dominate the real impedance, and not the interface's  $R_p$ .

#### H. Summary - Bulk Properties of the System

We have shown how the system's bulk properties may be related to

it's microscopic composition. The prominence of the electronic properties of the interface is largely due to the semiconductor on the interface being the sole contributor to the charge storage in the system. There is an interplay between the deliverance of the current to the interface by the electrolyte and the interface's ability to pass current (and we have shown that this interplay is highlighted by the frequency dependence of the impedance). The penetrability of the current is solely determined by the complex impedance of the interface (Figure 3.8) and the electrolyte's concentration. The increased electrode surface area can be determined from the low frequency impedance data, whereas the high frequency impedance typifies flat electrode behavior of the area outside the pores. Thus far, however, we have not focused on the ramifications of the porous electrode's surface inhomogeneities and irregularities other than as a means of increasing the total surface area available for the electrochemical reaction. This information is contained in the middle frequency impedance data (see section II.E), and it remains for us to attempt to extract some unique interpretation from it.

#### I. Model Output-Surface Contributions

In an attempt to assess the surface roughness contributions to the impedance, we address two questions:

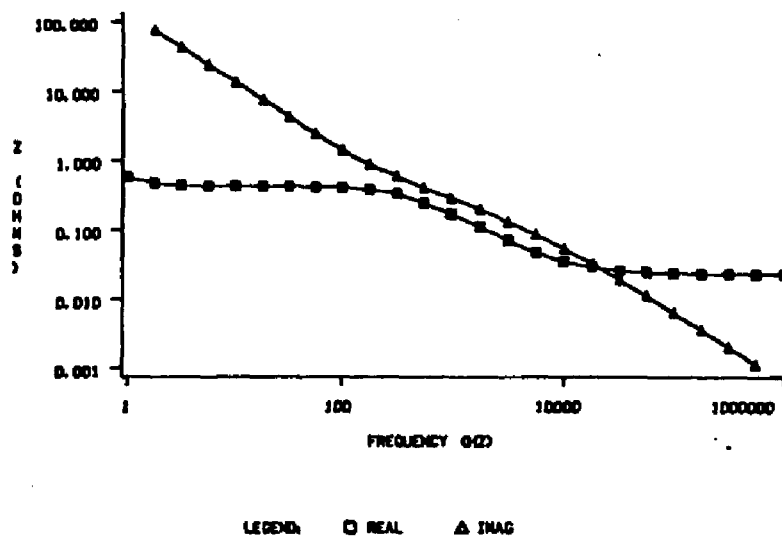
- 1) Can a characteristic topology and morphology of the RNM porous electrode surface be identified? Can the features of the surface be quantified?

- 2) Can a characteristic morphology so identified be correlated with the electrode's impedance characterization?

To answer the second question first, recall that in section I.E we cited the work of Deutscher et. al. in showing a fractal impedance behavior of metal film fractal structures, and in section II.E we cited the work of Liu who used a similar approach in calculating the impedance behavior of a fractal electrode interface. These studies both utilized systems whose impedance was linearly dependent on area, so that a self-similar area-length relationship translated into a self-similar impedance-length relationship. Thus, Liu chose an ideally polarized electrode interface so that the interfacial impedance was that of a pure capacitor, to obtain the simple area dependent impedance. Following suit, we change the RNM interfacial impedance from the general non-IPE treatment of Figure 3.3 to that of an IPE by eliminating  $R_p$ . It follows that if we identify the electrode surface as fractal, we can directly correlate it with a CPA appearing in the impedance spectrum.

Solving the RNM for the ideally polarized interface presents some additional numerical and computational difficulties. This is due to the fact that at low frequencies the impedance of the interface is so high. This prevents us from examining the results of all input parameters, including the configuration of figures 3.3 and 3.5. By reducing the capacitance from  $10^{-6}$  to  $10^{-4}$  Farads, we successfully obtain the results of Figure 3.13, which indeed shows a CPA behavior in the intermediate frequency range. From Figure 3.13b we evaluate

## IMPEDANCE - IPE - P220



## MID FREQUENCY CPA - P220

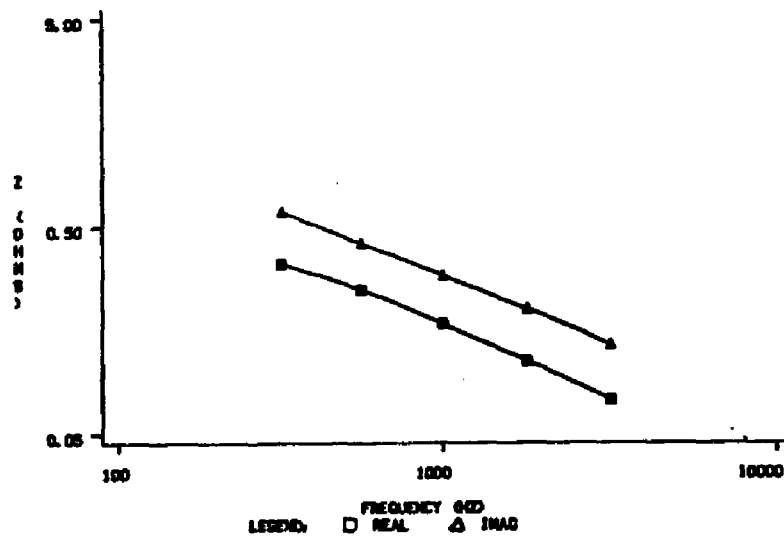


Figure 3.13- CPA behavior of the impedance in the RNM when we assume an ideally polarized electrode.

the slope of the log-log plot as  $-.67$ . We find, however, that the value of the CPA slope may be altered simply by changing the impedance values of the constituents of the system. Changing the values of  $C_p$  or of  $R_m$  (the metal resistance) varies the CPA slope from  $.6$  to  $.8$ . This strongly supports the notion that the appearance of a CPA dependence is not uniquely determined by the purely structural morphology of the electrode.

To further explore the possible fractal nature of our surface, we utilize Equation 1.14, to calculate the density-density correlation function for the surface metal sites. In our calculation we averaged the two particle correlations over the six nearest neighbor directions, and we did not employ continuous boundary conditions. The correlation function as a function of radius does not show a linear relationship on a log-log plot (Figure 3.14). In fact, the behavior is not unlike that of a Richardson curve for a circle (Figure 1.3). We verified that this was not due to finite-size effects by showing that the same behavior was obtained irrespective of system size. The same non-fractal correlation is obtained irrespective of porosity (Figure 3.15) which suggests that our RNM porous electrode surface is not fractal. The fact that our electrode does not exhibit fractal properties further undermines the Liu hypothesis which attempts to uniquely tie CPA behavior to fractal morphologies.

2 Particle Correlation Func.  
por=.4 lattice sizes 11,20,30  
C(r)

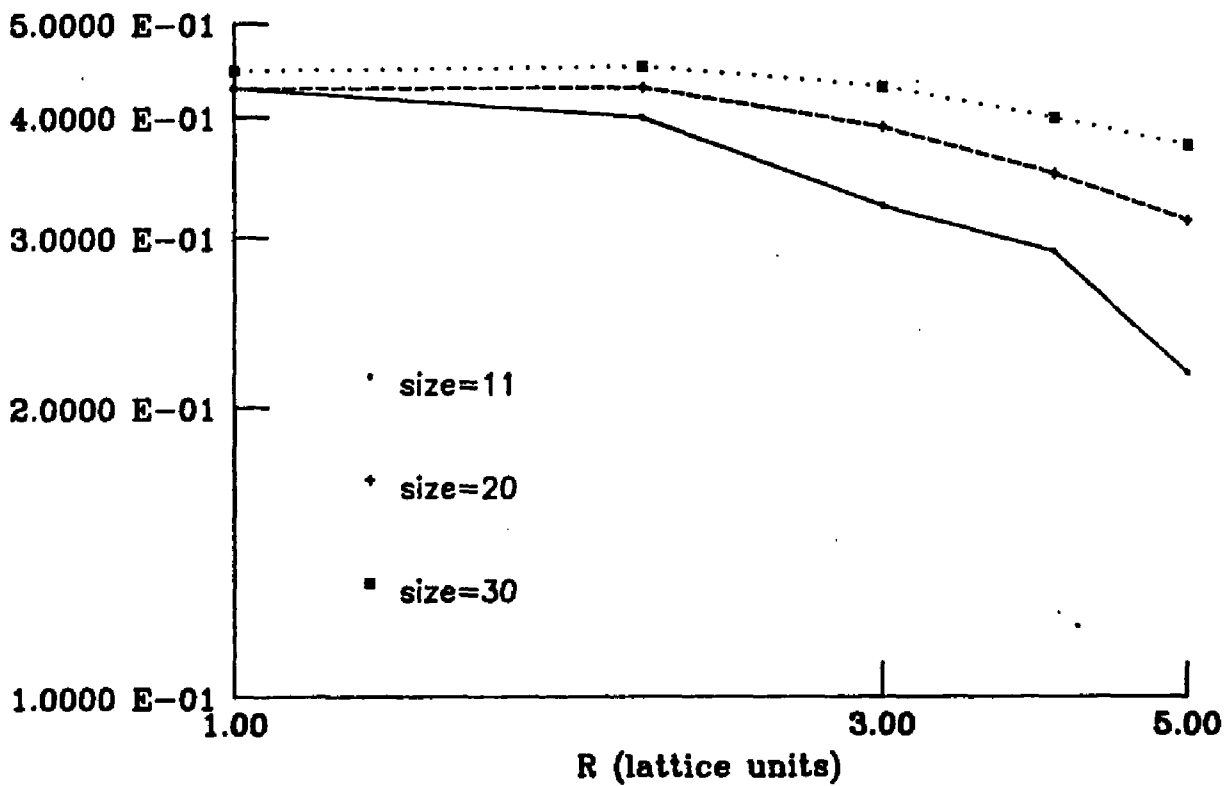


Figure 3.14- Correlation function of our RNM lattice.

## 2 Particle Correlation Func. for various porosities $C(r)$

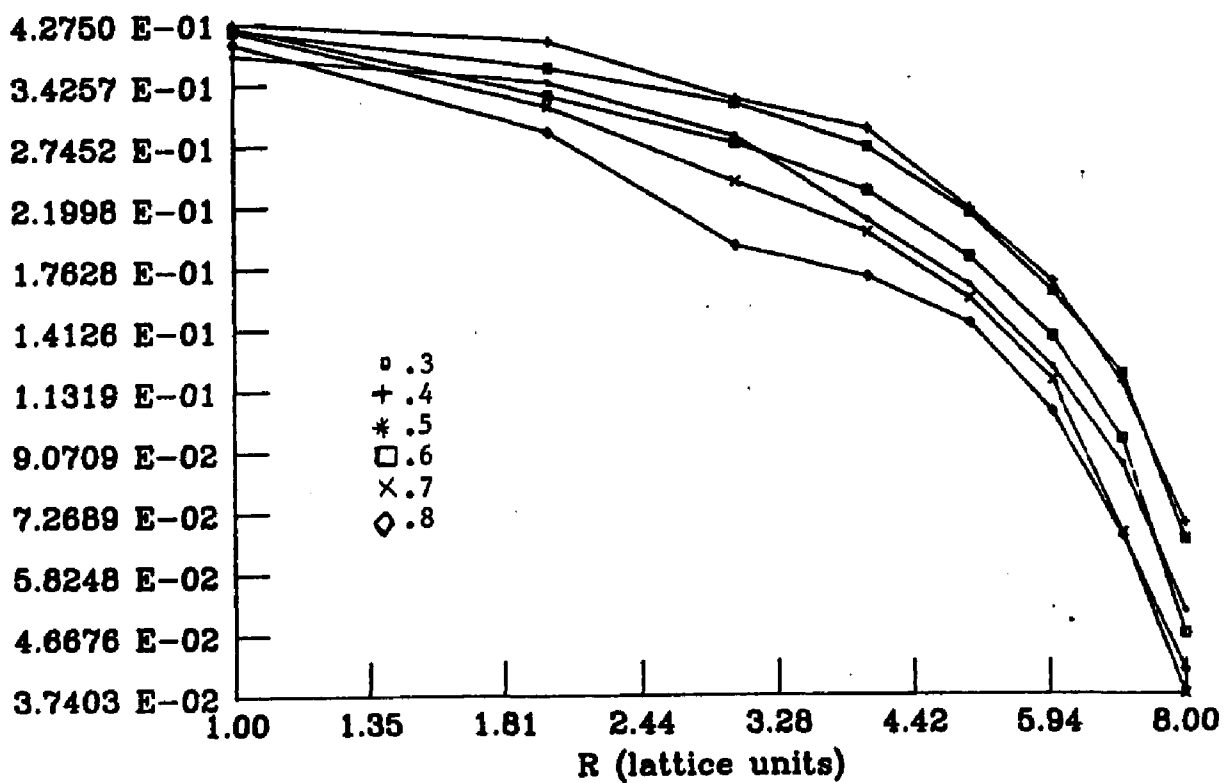


Figure 3.15- All porosities are measured at lattice size 11.

#### IV. Dissolution-Precipitation Reactions

Many electrode processes involve the formation of deposits on an electrode from solution and the dissolution of material back into solution. Corrosion, deposition, dendritic growth are all complex processes which involve a mass transfer between the solution and the electrode surface and back<sup>58</sup>. The reactions involved are diverse and complex, and we present an idealized computer simulation of the process in order to explore the physical structures formed on the electrode and to present methods for characterizing this surface. The simulation does not concern itself with the specifics of the chemical mechanisms involved, but rather treats the dissolution and precipitation as random processes on a large number of particles which are governed by average reaction and diffusion rates.

The use of computer simulation to study this class of phenomena is not new. Witten and Sander<sup>19</sup> were the first to apply this approach to the study of diffusion-limited aggregation, in which particles were allowed to form in spherical clusters growing from one initial seed site. They measured a correlation exponent (equation 1.14) of .34 on a square lattice. Meakin<sup>55</sup> has extended the original study by considering varying sticking probabilities, and clusters in three and four dimensional space. In two dimensions, Meakin lists the correlation exponent as .29 for a sticking probability of 0.25, and .26 for a sticking probability of 0.1. There has also been numerous experimental verification of these ideas, that indeed aggregation phenomena build fractal structures<sup>58-62</sup>. More recently, Voss and

Tomkiewicz<sup>56,57</sup> have applied the same methodology to the study of dendritic growth in electrodeposition, and were able to correlate their theoretical calculations of dendritic growth rates with experiment. Thus far, however, the simulations have considered only the aggregation of particles onto a substrate. The more general situation in which particles are allowed to dissociate back into solution has not yet been treated. It is precisely dissolution precipitation that has been identified<sup>63</sup> as a reaction mode in porous electrodes (as in the anodic formation of ZnO on Zn electrodes in KOH), and in order to achieve a better understanding of the type of surface structures that result from this system, we have extended the previous models. This simulation is an alternative approach to the simple random metal distribution that we utilized in the previous chapter for constructing our electrode, and it also serves as a more realistic picture of the surface as the electrode's deposit of insulating material builds during discharge.

#### A. The Simulation

A two dimensional  $N \times N$  lattice is defined on which two types of particles reside- solid particles are fixed (=solid phase), and dissolved particles are free to move through the lattice (=liquid phase) under Brownian motion. We define a dissolution probability,  $p_d$ , and a precipitation probability,  $p_p$ , (corresponding to the rate constants of those respective reactions) and a moving probability,  $p_m$  (corresponding to the diffusion of dissolved particles). Dissolved particles precipitate only if they are part of a cluster of

minimum size  $N_m$ . Each time step in the simulation every particle in the system is examined in a random order, and the following occurs:

- 1) If the particle is in the bulk of the solid then it remains stationary.
- 2) If the particle is part of the "solid", and it resides on the surface, it is given a chance to dissolve into solution with probability  $p_d$ .
- 3) If the particle is dissolved, it moves one step in a random direction with probability  $p_m$ . If the lattice site that it tries to move to is occupied, the particle remains in place.
- 4) If the particles is dissolved in solution but adjoins the solid surface, and is part of a cluster bigger than  $N_m$ , it is given a chance to precipitate with probability  $p_p$ . When the particle precipitates, the adjoining cluster also precipitates.

These checks are made sequentially, so that a particle may be dissolved in step (2) and reattached in step (4). The simulation is begun with rows 1 to  $N/2$  populated with a flat-surfaced solid and with the top half of the lattice empty.

The simulation proceeds for  $t$  time steps, and a schematic representation of what the system might look like after time  $t$  is shown in Figure 4.1. The particles in solution are represented by the stars, the particles of the bulk are represented by squares, and the particles on the interface are highlighted by the thick continuous line. As illustrated in Figure 4.1, quantitative measures of possible interest for the precipitated particles are the maximum,

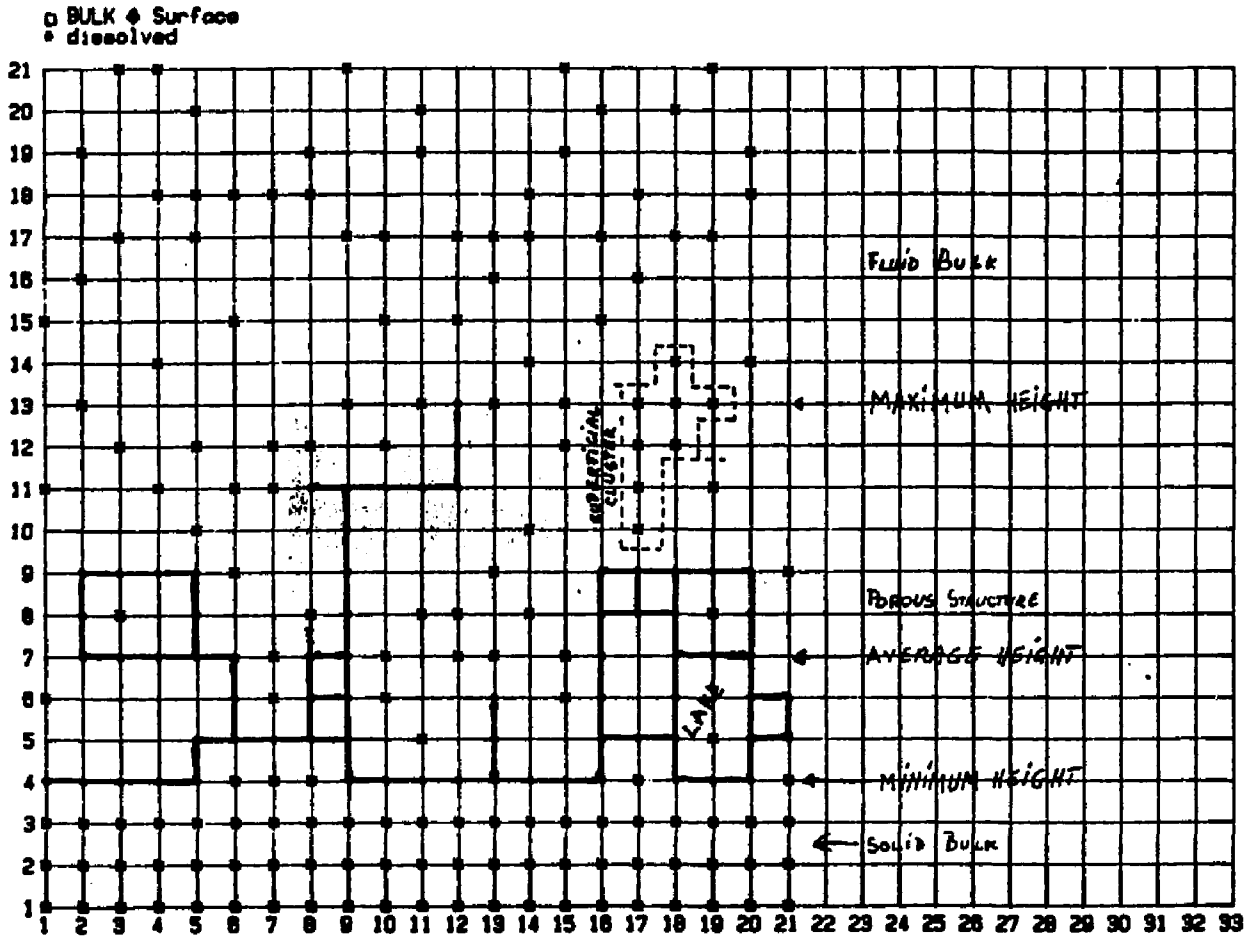


Figure 4.1- Schematic of the DP model at a given time  $t$ . Some quantities of possible theoretical interest are labeled. The surface of the solid is indicated by the heavy line.

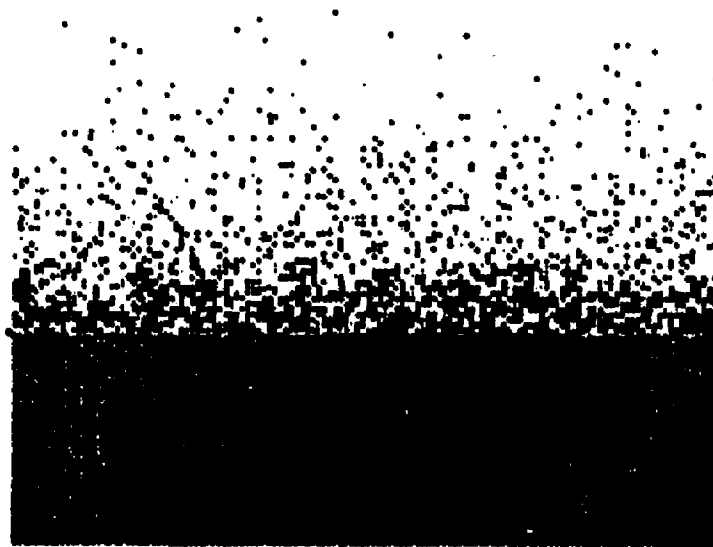


Figure 4.2- A typical system configuration of the DP model. This result is for a dissolution probability of .5, a precipitation probability of .5 after 1000 time steps.

minimum and average growth heights, as well as the surface roughness and the possible fractal dimension of the interface. The solution may be characterized by the diffusion behavior of the dissolved particles. Figure 4.2 shows an actual simulation result at  $t=1000$  for  $p_d=p_p=.5$ , and for  $p_m=1.0$ . It remains for us to attempt to quantify the system and ultimately to correlate the solute behavior and the grown structures with the input parameters of the simulation.

#### B. Solution characterization-Diffusion Behavior

It is to be expected that if the simulation is realistic, the particles in solution should follow the diffusion equation:

$$\frac{\partial C(x,t)}{\partial t} = D \frac{\partial^2 C(x,t)}{\partial x^2} \quad (4.1)$$

with the following boundary conditions:

$$\begin{aligned} c(x,0) &= c^* & , x > 0 \\ c(\infty,t) &= c \\ D \left( \frac{\partial C}{\partial x} \right)_{x=0} &= k_1 C(0,t) - k \end{aligned} \quad (4.2)$$

where  $D$  is the diffusion coefficient,  $C(x,t)$  is the time and position dependent concentration,  $c^*$  is the bulk solution concentration,  $k$  is the dissolution constant and  $k_1$  is the precipitation constant. The third boundary condition establishes a fictitious flat plane at  $x=0$  at which all the dissolution-precipitation process is said to occur at all times,  $t$ . As the simulation progresses and the surface gets to be rough, this is no longer true. With this approximation, we assume that one can always find a plane at  $x=x_0$  where the dissolution precipitation process effectively occurs. The

value of  $x_0$  is a parameter to be determined in fitting our data to these calculations.

The differential equation 4.1 may then be solved to yield:

$$C(x,t) = c^* + \left( \frac{k-k_1c^*}{k_1} \right) \left\{ \operatorname{erfc} \left( \frac{x}{2(Dt)^{1/2}} \right) - \exp \left( \frac{k_1(x+k_1t)}{D} \right) \operatorname{erfc} \left( \frac{x}{2(Dt)^{1/2}} + k_1 \sqrt{\frac{t}{D}} \right) \right\} \quad (4.3)$$

Solving for the total number of particles in solution, from Fick's first law,

$$\frac{\partial N}{\partial t} = -D \frac{\partial C}{\partial x} \quad (4.4)$$

$$\frac{\partial N}{\partial t} \Big|_{x=0} = k - k_1 C(0,t) \quad (4.5)$$

These diffusion equation have been successfully utilized to predict the behavior of experimental dissolution-precipitation systems<sup>64</sup>.

In our simulation  $c^*=0$ , and equation 4.4 may be integrated to give the total number of particles in solution at time  $t$  as:

$$N(t) = \frac{kD}{k_1^2} \left\{ \exp \left( \frac{k_1^2 t}{D} \right) \operatorname{erfc} \left( k_1 \sqrt{\frac{t}{D}} \right) + 2k_1 \left( \frac{t}{\pi D} \right)^{1/2} - 1 \right\} \quad (4.6)$$

Figure 4.3 is a plot of concentration as a function of position from the surface into the the solution at  $t=1000$  time steps. The dotted line separates the  $x$  axis into two regions. In region I is the

S5517 1000

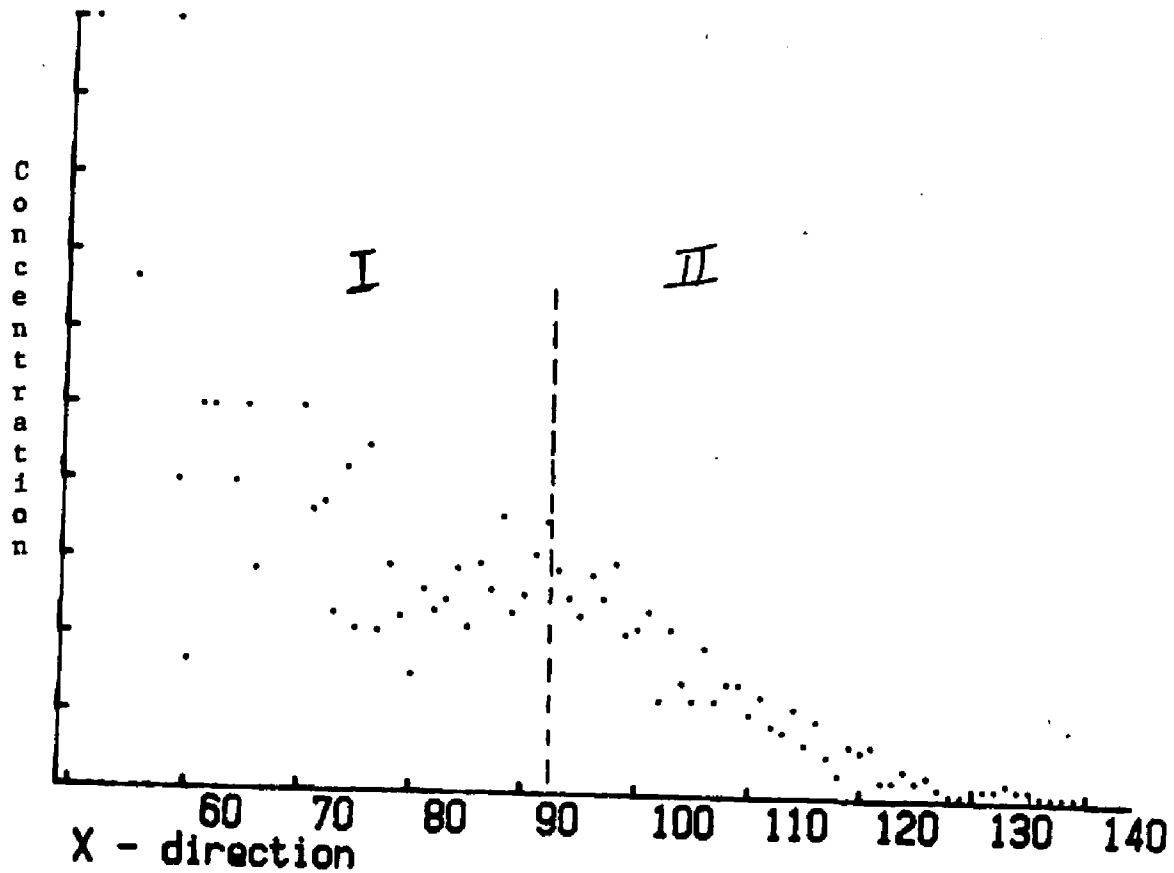


Figure 4.3- The concentration in solution as a function of the distance away from the surface. Region I is still within the heterogeneous mixture of solid and solute. Region II is above the height of the tallest solid peak.

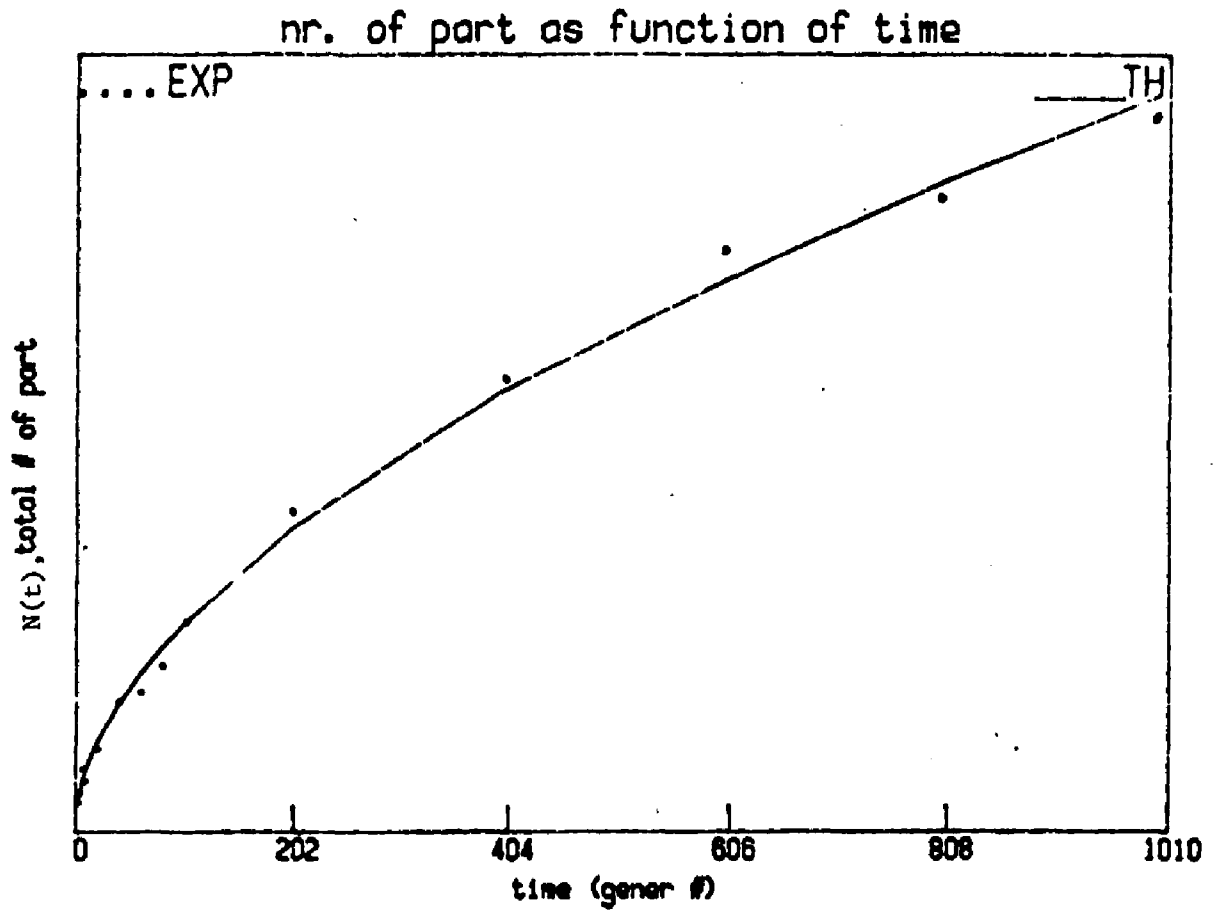


Figure 4.4- The total number of dissolved particles in solution as a function of time. The straight line is a fit of the data to equation 4.6 .

rough interface where there is an inhomogeneous mixture of solid and dissolved particles, which leads to large fluctuations in the dissolved particle concentration. In region II is the bulk of the solution, and we see the particle concentration begin to behave similar to what we would expect from equation 4.3. The total number of particles in solution as a function of time is shown in figure 4.4, and the solid line is a fit from equation 4.6. This is a confirmation of the validity of the simulation, and from the fit parameters we obtain the theoretical  $k$ ,  $k_1$ , and  $D$ . Using this methodology, it is possible to correlate the simulation input parameters  $p_p$ ,  $p_d$ ,  $p_m$  with the more realistic diffusion equation parameters.

### C. Solid Characterization - Rough Surface

We can also explore how surface topology is altered by variations in the previously mentioned input parameters. The surface properties can be quantified by calculating the density-density correlation function as in chapter III, or the more straightforward Richardson curve for the surface length under different measuring scales. (The Richardson curve measurement on a discrete lattice is easily implemented on a two dimensional surface as opposed to the 3D Random Network Model of chapter III.) Figure 4.5 is the result of such a calculation for the system of figure 4.2. The calculation is done by considering pair correlations only for the surface particles. First, the computer identifies which particles are on the interface (the solid line in Figure 4.1) and then discards all the other points. The pair correlation for each radius  $r$  is computed, and

this is averaged over the four nearest-neighbor directions. Figure 4.5 shows a power law behavior in the relationship between  $C(r)$  and  $r$ , with a correlation exponent of 0.22 .  $C(r)$  is shown at  $t=600,800,1000,2000$  time steps, and the power law behavior is stable for  $r < 9.0$  . Finite size effects seem to dominate for  $r > 9.0$  . From equation 1.14 the fractal dimension of this two-dimensional system is  $D=1.78$  . This value is considerably higher than that calculated by Witten, Sander and Meakin, but it is worthwhile to note that the geometry of our system is very different from the spherical cluster aggregation that they considered. It is hoped that a further understanding of the significance of the values in this system may be realized by examining the fractal dimension under different dissolution and precipitation probabilities, as well as at longer times.

The surface constructed using the dissolution-precipitation model is an alternative starting point for our Random Network Model. Instead of taking the electrode configuration as constructed using a simple random number generator, we can utilize the dissolution-precipitation lattice as a template for assembling our system of resistors and capacitors. This is a more realistic system than the Cantor Bar model, and is an example of exactly the type of self-similar R-C network that Nyikos and Pajkosky proposed to construct for the examination of fractal interfaces (see section II.E).

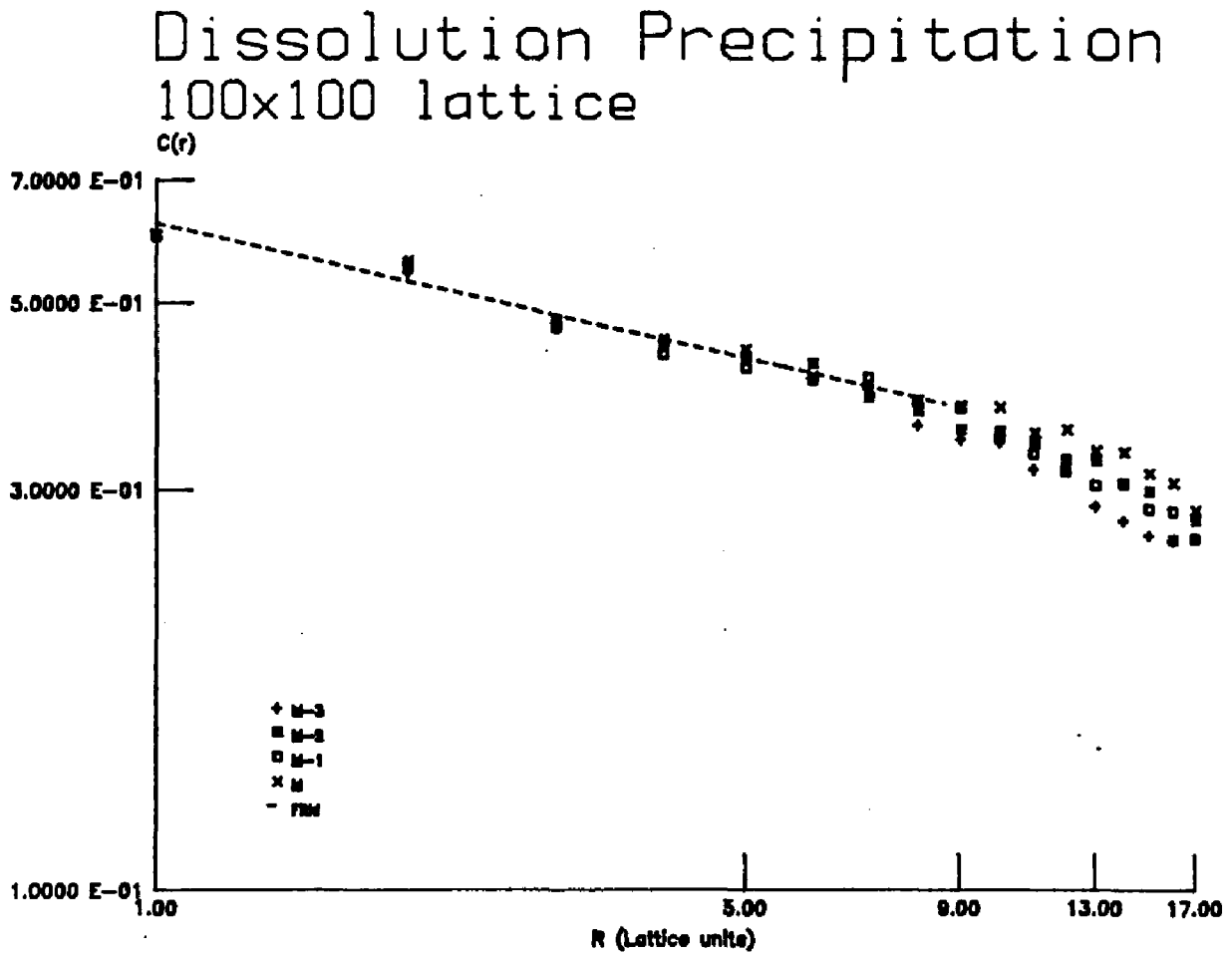


Figure 4.5- Correlation function calculation for the surface of the DP model for the system shown in Figure 4.2 .

## V. Summary and Conclusions

Recent developments in the field of composite media and disordered systems have availed us with powerful techniques for characterizing stochastic multi-phase systems. Analytic methods such as Effective Medium Theory offer a simplicity of concept and execution, but are more limited in their ability to model some of the salient features of real life systems. Computer simulations offer more flexibility in this regard, and provide a powerful means of exploring the behavior of systems with detailed constraints and/or complicated geometries. The drawback to numerical simulations is that one is limited by a finite accuracy that may not be sufficient in some cases (the "round off error"), and by the fact that as fast as today's computer's are, sophisticated models and large systems require huge amounts of computer time.

The simulations presented in this work are physical simulations involving the construction of a model structure to mimic the topological features of a system. The process is really a two step one:

Firstly, the system must be constructed in the computer using a realistic set of rules and input parameters. We have presented two examples of this - the random network model (RNM) and the dissolution-precipitation (DP) model. These representations are very different from each other and complementary; the RNM model is an attempt to depict a typical porous metal structure in an electrolyte with the major topological contribution arising from the

metal's porosity, while the DP model is an attempt to depict the topology of an electrode surface as it is roughened by chemical deposition and dendritic growth. An "ideal" examination of porous electrodes might utilize both of these configurations simultaneously. To be sure, both of these models introduce simplifying assumptions, and additional variations can be considered to explore how different pictures of the geometry and different constraints would affect the results.

Secondly, once the system has been constructed, the computer may be utilized to analyze and perhaps quantify the structure of the model. In addition, other properties of the system may be calculated and an attempt can be made to correlate these with the system's topology. In this work we examined the impedance behavior of the system and its dependence on morphology. The electrical impedance is very well suited for this type of exploration, due to the fact that impedance in addition to being dependent on the internal structure of the material is highly dependent on its macroscopic shapes and lengths. The frequency dependence of the impedance is especially useful in this respect as well, for it allows us a consistent way of weighting various surface and bulk effects with respect to each other and seeing the result. It is also a measurement that is well open to experimental exploration and verification.

In addition to providing sample implementations of the above methodology, we have provided some new insight into the porous electrode system. We have demonstrated the utility of the low frequency

capacitance values in measuring the apparent area of the rough electrode, and provided a caveat concerning "what is called low frequency?"- showing that it is dependent on the relative magnitudes of the interfacial impedance and the electrolyte resistivity in each electrode system. In this regard, for all its simplicity, the DeLevie model is an excellent representation of the current flow in a porous structure. We have found that in an ideally polarized rough electrode there is a fractional power law relationship between  $Z$  and  $\omega$  with an identical scaling exponent for the real and the imaginary components (the CPA behavior), but we have shown that it is not necessarily related to fractal structures, as suggested by Liu and others. All in all, the RNM seems to be a useful addition to an electrochemist's toolbox. The DP model of Chapter IV is an attempt to explore the morphology of the electrode which results from the electrochemical reactions on the interface. We have shown that the model is valid by verifying the predicted diffusion behavior of the dissolved particles. The DP model also produces a fractal surface which is fertile ground for additional research.

#### Future Work

We have treated a single geometry in the RNM, and an extension of that to alternate geometries is warranted. One example is allowing the fluid to flow through the cracks between adjoining metal sites. The DP model is as yet largely unexplored - we have not yet examined how the fractal geometry of the system changes under varying sticking and dissolution probabilities. An important further consideration is to attempt to construct the system in different geometries

(for example on a triangular lattice) to verify that the results are independent of the numerical implementation of the model. The merging of the RNM and the DP model is an important further step to be explored. We can thus construct a realistic picture of the electrode under operating conditions, and solve for the impedance of such a structure. New techniques are being utilized for the calculation of network impedances utilizing a transfer matrix algorithm<sup>68,69</sup>. The advantage of this method is greater accuracy as well as greater efficiency in utilizing computer storage. This method has already been applied to 2-D complex impedances<sup>70</sup>, and extension to three dimensions should be trivial. Any further work in network simulations would benefit by utilizing these transfer matrix techniques instead of the simultaneous equation solutions.

APPENDIX

PROGRAM LISTINGS



```

* * * * * LIST OF CALLED SUBROUTINES: ( -- DENOTES A S.R. NOT LISTED
* * * * * HERE)
* * * * *
* * * * * --CALL ERASET (208,0.5,1)
* * * * * --CALL DATIM (EVCPU,ETIME,ETCPU)
* * * * * CALL NODES(A,LBIA,POR,DSEED,MODEL)
* * * * * CALL DISPLA(A,ISHOW,MODEL)
* * * * * CALL CORLAT (A,PORVOL,VOLNOR,1)
* * * * * CALL CHOP (A)
* * * * * CALL CONDUCT(A,G,JA,IA,B,N,NTCALC,MODEL)
* * * * * --CALL MOOO(N,NTCALC,IBUF1,IA,JA,IPM,IER,IB,IU,JU)
* * * * * --CALL MSSO(N,IBUF1,IBUF2,IA,JA,IPM,IU,JK,IER,IUP,IAT,JAT,IAP,
* * * * * --CALL MNSO (N,ISM,IA,JA,G,IAT,JAT,IAP,IPM,IU,DU,DI,IER,IUP,X)
* * * * * --CALL MBSO(N,ISL,IPM,IU,DU,DI,B,G,X)
* * * * * CALL CURCAL (A,G,MODEL)
* * * * * CALL CURPRF (A,LBIA,G)
* * * * * CALL LATSET(A,POR,MODEL)
* * * * * CALL PLANE (A,1,5)
* * * * * CALL RNDM(A,MB,DSEED,KMIN,KMAX,1)
* * * * * CALL CONECT(A,K,5,1,IFND,KFND)
* * * * * CALL RANPT(A,DSEED,KMIN,KMAX,IT,JT,KT)
* * * * * CALL NEARN(A,IT,JT,KT,0,1,IFND,KFND)
* * * * * CALL FILABL(A,LBIA)
* * * * * CALL GCUBS(DSEED,3,R)
* * * * * CALL DISPLA (A,ISHOW,MODEL)
* * * * * CALL NOD146(A,1,IAT,NEIGH,IGND,Z1,Z2)
* * * * * CALL NOD235(A,2,IAT,NEIGH,IGND,Z1,Z2)
* * * * * *****
C SIZE OF A IS (IM,JM,KM)
C PARAMETER (MAXSIZ = 11, INODES = MAXSIZ**3)
C PARAMETER (ISMAX = 2*INODES, ISMIN = ISMAX-2)
C PARAMETER (NT = 10 * (4*INODES - 3*MAXSIZ*MAXSIZ + 3))
C PARAMETER (NT1 = 15*NT, IBOT = 46*NT, NFD = NT - ISMIN)
C PARAMETER (IBOT = 2*NFD)
C
C INTEGER*4 A(MAXSIZ,MAXSIZ,MAXSIZ),LBIA(MAXSIZ,MAXSIZ,MAXSIZ)
C REAL *8 DSEED,DHOLD,X
C LOGICAL*1 IPOLAR
C
C COMMON/DIMS/IDIM1, IDIM2, IDIM3, IDIM4
C COMMON/NUMB/PI
C COMMON/LATICE/ IM,JM,KM,KMSTOP
C COMMON/ELEC/Z(6,2),CUR(2),NCUR,NGND,REAL1,RMAG1
C COMMON/GTEMP/ NG1,NG2,NG3,NG4,NG5,ICOL(6),DUMHY(12)
C DIMENSION IDIM(7),CURNOR(2)
C ***** VARIABLE SIZE PROBLEM *****
C
C THE BASIC SYSTEM DIMENSIONS ARE IM,JM,KM
C ISM = IM*JM*KM-1

```

```

C      N = # OF SIMUL EQU (N.LE.2*S)
C      1) ISMIN=N-2*(IM*JM*KM-1)
C      -----
C      2) DIMENSION OF ISMAX IS ISMIN+2
C      DIMENSION IA(ISMAX),IB(ISMAX),IU(ISMAX),IPH(ISMAX),
C      11PH(ISMAX),DI(ISMAX),IX(ISMAX),B(ISMAX),X(ISMAX),
C      11UP(ISMAX),IAT(ISMAX)
C      -----
C      3) NT IS # OF PACKED ELEMENTS IN MATRIX G--THE SPARSE
C      STORAGE COEFF. MATRIX
C      NT=4*(4*S-3*IM*JM+3)
C      DIMENSION JA(NT),G(NT)
C      -----
C      4) NT1 IS BETWEEN 2*NT+N AND 3*NT AND HERE WE SET IT TO 14*NT
C      NT1=14*IDIM(3)
C      DIMENSION U(NT1)
C      -----
C      5) IBOT SHOULD BE BETWEEN 2*NT+N AND 3*NT BUT HERE WE USE 44*NT
C      IBOT=44*IDIM(3)
C      DIMENSION JU(IBOT)
C      -----
C      6) NRD = NT - N
C      DIMENSION JAT(NRD),IAP(NRD)
C      -----
C      7) IBOT SHOULD BE 2*NRD
C      DIMENSION JUA (IBOT)
C      -----
C      DIMENSION IER(7),ERIND(4)
C      DATA ERIND/'MDOO','MSSO','MNSO','MBSO'/
C      *****
C      IDIM1=ISMIN
C      IDIM2=ISMAX
C      IDIM3=NT
    
```

POR01030  
 POR01040  
 POR01050  
 POR01060  
 POR01070  
 POR01080  
 POR01090  
 POR01100  
 POR01110  
 POR01120  
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 POR01180  
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 POR01200  
 POR01210  
 POR01220  
 POR01230  
 POR01240  
 POR01250  
 POR01260  
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 POR01280  
 POR01290  
 POR01300  
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 POR01370  
 POR01380  
 POR01390  
 POR01400  
 POR01410  
 POR01420  
 POR01430  
 POR01440  
 POR01450  
 POR01460  
 POR01470  
 POR01480  
 POR01490  
 POR01500  
 POR01510



```

5 CALL DATTIM (EVCPU,ETIME,ETCPU)
C WE'RE ONLY INTERESTED IN TOTAL CPU TIME
C INITIALIZE TIME COUNTERS AND CONVERT TO MINUTES
  STIME = ETCPU/60.0
  CONLAS = ETIME/60.0
  TIMLAS = STIME
C *****
C NRPT=0
C FIRST SET UP LATTICE PARAMETERS
  READ (5,10,END=900) IM, JM, KM, POR, POREND, PORINC
10 FORMAT (3I2,3F3.2)
  READ (5,19) MODEL
19 FORMAT (I3)
  IF (MODEL.LT.0) THEN
    ISOLVE = 0
    MODEL = IABS(MODEL)
  ELSE
    ISOLVE = 1
  ENDIF
  IF (IM.GT.MAXSIZ) THEN
    WRITE (7,11) IM, MAXSIZ
    GOTO 910
  ENDIF
11 FORMAT (' ERROR-- IM= ',I3,' IS GT MAXSIZ= ',I3)
  IF (PORINC.EQ.0) PORINC = .1
C----- EXTRA PARAMETER DEFINES HOW HIGH UP IN THE
C          LATTICE THE ELECTROLYTE IS EXPECTED TO FLOW
C
  READ (5,20) KMSTOP
20 FORMAT (I2)
  IF (KMSTOP.GT.KM) KMSTOP = KM
C
  IS = IM*JM*KM
  ISM = IS - 1
C NOW READ IN ELECTRICAL PARAMETERS
  READ (5,50) RELIN,RMETIN,RSIN,RFIN,CPIN
50 FORMAT (5E9.1)
C *****NORMALIZE THE ELEMENTS TO THE LATTICE SIZE
C
C PORSIZE OF 10 MICRON= 1E-3 CM; WE USE UNIT IN CM
C
  PORSIZ = 1.0E-3
  AREA = (IM-1) * (JM-1)
C          SYSIZE IS THE SIZE OF SYSTEM IN CM**2=PORSIZ**2 * AREA
C          1 / SYSIZE IS THE SCALING FACTOR TO SCALE
C          THE SYSTEM UP TO THE 1 CM**2 STANDARD VAL.

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POR02020
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POR02100
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POR02120
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POR02500

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        SYSIZE = PORSIZ**2 * AREA
        SCALE = 1.0/SYSIZE
C   CAPACITORS ARE TO BE MULT BY SCALE; RESISTORS DIVIDED
C
        WRITE (6,60) PORSIZ,AREA,SYSIZE,SCALE
60  FORMAT (1 PORE SIZE-IN CM-FOR THE UNIT PORE ',1P,E9.3,
1' LATTICE AREA = (IM-1)*(JM-1) = ',0PF6.0,/,
1' SIZE OF WHOLE SYSTEM= ',1P,E12.3,/,
1' AREA MUST BE SCALED BY ',0PF10.0,' TO APROXIMATE A LARGER',
1' 1 CM**2 SIZE SYSTEM')
*   NOW NORMALIZE THE CIRCUIT ELEMENTS FROM 1 CM**2 TO PORSIZE**2
*
        ENORM = 1.0 / (PORSIZ**2)
*
*   THE ELECTROLYTE & METAL RESISTOR IS NORMALIZED BY L/A = 1/L ONLY
*
        RNORM = 1.0 / PORSIZ

        REL = RELIN * RNORM
        RMET = RMETIN * RNORM
        RS = RSIN * ENORM
        RP = RPIN * ENORM
        CP = CPIN / ENORM
        READ (5,100) CUR(1),CUR(2)
100 FORMAT (2E7.0)
        READ (5,110) FRBEG,FREND,IPPD
110 FORMAT (2E7.0,12)
C   HOW MANY TIMES DO U REPEAT WITH A DIFFERENT RNDM # ?
C   IRPT=0 MEANS THAT ONLY ONE RNDM SEED SHOULD BE USED
*   IRPT LT 0 MEANS JUST CONSTRUCT LATTICE -- DONT SET UP MATRICES
*   AND DONT SOLVE EQUATIONS
        READ (5,20) IRPT
*   THERE ARE TWO NESTED LOOPS HERE: 1-CHANGE POR 2- CHANGE FREQ
***=====>>>> START LOOP FOR DATA <<<<=====***
120 FREQ=FRBEG
    IF (FREQ.LT.0.0) FREQ=1.0
    NCUR = 1
C
C   SET FLAG TO INDICATE DISTRIBUTION OF CURRENT EQUALLY
C   THROUGH BOTTOM (ELECTROLYTE) PLANE
C
        IDIST = 1
C
        NGND = IS
122 FRCNT = 0.0
C
C   FRCNT IS FREQ LOOP COUNTER- OUTERMOST LOOP
C

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POR02990
POR03000

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```

C NOW SET UP AND DISPLAY PHYSICAL SYSTEM
C
  WRITE(6,124) IM,JM,KM,POR
  WRITE(7,124) IM,JM,KM,POR
124 FORMAT ('LATTICE SIZE = ',3I4,' POROSITY = ',F3.2)
  READ (5,125) DSEED
125 FORMAT(D13.0)
C INITIALIZE 'NEW LATTICE' FLAG
C
  IWENT = 0
C
C
C STORE INITIAL DSEED VALUE FOR THIS LATTICE
  DHOLD=DSEED
C ----- WRITE SUMMARY TO DATABASE FILE -----
  WRITE (8,141) IM,JM,KM,MODEL
141 FORMAT ( ' 01 ',3I2,14 )
  WRITE (8,142) POR,KMSTOP,NRPT
142 FORMAT ( ' 02 ',F3.2,2I3)
  WRITE (8,143) RELIN,RMETIN,RSIN,RPIN,CPIN
143 FORMAT ( ' 03 ',1P,5E9.1)
  WRITE (8,144) FRBEG,FREND,IPPD
144 FORMAT ( ' 04 ',1P,2E7.0,13)
  WRITE (8,121) DHOLD
121 FORMAT ( ' 05 ',F13.0)
C ----- END OF PRELIMINARY DATABASE ENTRY -----
  CALL NODES(A,LBLA,POR,DSEED,MODEL)
  IF (IDUMP.EQ.1) THEN
    WRITE (9,133) ((A(I,JJ,KK),I=1,IM),JJ=1,JM),KK=1,KM)
133 FORMAT ( ' ',33I2)
  ENDIF
  ISHOW = 1
  CALL DISPLA(A,ISHOW,MODEL)
* LOOK FOR LATTICE CORRELATIONS TO TRY AND FIGURE OUT GEOMETRY
  WRITE (6,*) 'METAL CORRELATIONS:'
  WRITE (7,*) 'METAL CORRELATIONS:'
  CALL CORLAT (A,PORVOL,VOLNOR,1)
  WRITE (6,*) 'ELECTROLYTE CORRELATIONS:'
  WRITE (7,*) 'ELECTROLYTE CORRELATIONS:'
  CALL CORLAT (A,PORVOL,VOLNOR,2)
C
C ----- STORE CURRENT VALUE OF KM
  KHOLD = KM
  IF (MODEL.EQ.3) GOTO 130
  IF (KMSTOP.EQ.KM) GOTO 130
  CALL CHOP (A)
C
C
C
*****===== >>>> NEXT FREQUENCY <<<<=====*****

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POR03010
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POR03500
POR03510

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130 IF (IDUMP.EQ.1) THEN
* ONLY LIST EVERY MULTIPLE OF 10
  IF (MOD(INT(FRCNT),1PPD).EQ.0) WRITE (9,131) FREQ
131  FORMAT (' FREQ',1P,E10.3)
  ENDIF
  WRITE (6,135)
135  FORMAT (' ',///,'#####')
  WRITE (6,505) FREQ
C NOW SET UP IMPEDANCES OF DIFFERENT CIRCUIT ELEMENTS AT THIS FREQ
  OMEGA = 2 * PI * FREQ
  DENOM = 1 + (OMEGA * CP * RP)**2
*HERE WE HAVE THE OPTION OF AN IDEALLY POLARIZED ELECTRODE
* IF !POLAR = TRUE THEN THE ELECTRODE IS IDEALLY POLARIZED
  !POLAR = .TRUE.
  ZREAL = RS + RP/DENOM
  ZIMAG = -OMEGA * CP * RP * RP /DENOM
*.....
  IF (!POLAR) THEN
    ZREAL = RS
    ZIMAG = - 1.0 / (OMEGA*CP)
  ENDIF
*****
C
C Z(1,N) IS METAL RESISTANCE; Z(2,N) IS ELECTROLYTE RESIST
C Z(3,N) IS METAL+S.C. RESIST; Z(3,N) IS ELEC +S.C. RES
C *** N REFERS TO: 1-REAL 2-IMAG *****
C
  Z(1,1)=RMET
  Z(1,2)=0.0
  Z(2,1)=REL
  Z(2,2)=0.0
  Z(3,1)=ZREAL+RMET
  Z(3,2)=ZIMAG
  Z(4,1)=ZREAL+REL
  Z(4,2)=ZIMAG
C FLAG #6 DENOTES METAL ON BOTTOM WHICH IS NOT TO HAVE SC INTFC
C ABOVE IT--- IT'S PURPOSE IS JUST TO DISTRIBUTE THE CURRENT
C INTO THE PLANE ABOVE IT
  Z(6,1) = RMET
  Z(6,2) = 0.0
  WRITE(6,132)REL,RMET,RS,RP,CP
132 FORMAT (' REL,RMET,RS,RP,CP= ',1P,5E13.3)
  WRITE(6,138) CUR(1),CUR(2)
138 FORMAT(' INPUT CURRENT= ',1P,E12.2,' +(J*) ',E12.2)
  WRITE (6,150)
150 FORMAT (' IMPEDANCES , REAL & IMAG. :')
  WRITE (6,200){Z(1,J) ,I=1,4},J=1,2)
200 FORMAT (' Z= ',4E13.4)
  IF (IDUMP.EQ.1) THEN

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POR03520
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IF (MOD(INT(FRONT), IPPD).EQ.0) WRITE (9,200)((Z(I,J),I=1,4),J=1,2)
ENDIF
C
C NOW CONSTRUCT CONDUCTIVITY MATRIX
CALL CONDUCT(A,G,JA,IA,B,M,NTCALC,MODEL)
C
IF (ISOLVE.EQ.0) THEN
  WRITE (7,301)
  FORMAT (' LATTICE CONSTRUCTED -- NO SOLVE OPTION IN EFFECT')
  GOTO 760
ENDIF
C
C IF THIS IS THE SAME LATTICE AS BEFORE, GO RIGHT TO SR MNSO
C SINCE IPM AND SYMBOLIC FACTORIZATION IS UNCHANGED!
C
IF (IMENT.NE.0) GOTO 380
C
  IBUF1 = IBOT
  IERIND=1
  CALL M000(N,NTCALC,IBUF1,IA,JA,IPM,IER,IB,IU,JU)
  IF (IER(1).NE.0) WRITE (6,350) ERIND(ERIND),(IER(1),I=1,7)
  350 FORMAT (' ERROR IN ',A4,4X,' IER=',7I10)
  IF (IER(1).EQ.0) WRITE (6,351) ERIND(ERIND),(IER(1),I=1,7)
  351 FORMAT (' NORMAL TERMINATION CODES FROM ',A4,4X,7I8)
  IBUF1 = IBOT
  IBUF2 = IBOT
  IERIND=2
  CALL MNSO(N,IBUF1,IBUF2,IA,JA,IPM,IU,JU,IK,IER,IUP,IAT,JAT,IAP,
  1IPM,IX,IB,JUA)
  IF (IER(1).EQ.0) GOTO 360
  -----ERROR DOCUMENTATION
  WRITE (6,350) ERIND(ERIND),(IER(1),I=1,7)
  353 FORMAT (' FIRST AND LAST ELEMENT IN ROW I OF G= ',2I11)
  WRITE (6,354) N,IK
  354 FORMAT (' NUMBER OF ROWS PROCESSED = ',I8,' TOTAL NO. OF ',
  C' OPERATIONS = ',I20)
  -----
  360 WRITE (6,352) IBUF1,IBUF2
  352 FORMAT (' SR MNSO: NUMBER OF BUFFER LOCATIONS USED IN JU',
  1' AND JUA = ',2I20)
  WRITE (6,370) IK
  370 FORMAT (' NUMBER OF MULTIPLICATIONS REQUIRED = ',I20)
  IBUF3 = IU(N+1) - 1
  WRITE (6,371) IBUF3
  371 FORMAT (' MINIMUM BUFFER SPACE FOR JU AND U IN MNSO = ',I20)
C
C ABNORMAL TERMINATION
C IF (IER(1).NE.0) GOTO 820
C

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POR04010  
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 POR04500

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C SET FLAG TO INDICATE THAT THIS LATTICE HAS MOOD AND MSSO SOLVED      POR04510
C                                                                           POR04520
  IWENT = 1                                                                POR04530
  ISW = IB(1)                                                                POR04540
380 IERIND = 3                                                                POR04550
  CALL MNSO (N, ISW, IA, JA, G, IAT, JAT, IAP, IPM, IU, JU, U, DI, IER, IUP, X) POR04560
  IF (IER(1).NE.0) WRITE (6,350) ERIND(IERIND),(IER(1),I=1,7)          POR04570
  ISL=0                                                                      POR04580
C IF FLAG 'IDIST' IS SET, THEN CURRENT IS TO BE UNIFORMLY DISTRIBUTED  POR04590
C OVER ALL OF BOTTOM PLANE (ELECTROLYTE).                                POR04600
C                                                                           POR04610
  IF (IDIST.NE.1) GOTO 410                                                  POR04620
C                                                                           POR04630
C                                                                           POR04640
  WAREA = IM * JM                                                            POR04650
  WRITE (6,395) WAREA                                                         POR04660
395 FORMAT(' CURRENT DISTRIBUTED EVENLY THROUGH BOTTOM',F4.0,' NODES')  POR04670
  CURNOR(1) = CUR(1) / WAREA                                                 POR04680
  CURNOR(2) = CUR(2) / WAREA                                                 POR04690
  IEND = 2 * IM * JM                                                         POR04700
  DO 400 I1=1,IEND,2                                                         POR04710
  B(I1) = CURNOR(1)                                                         POR04720
  B(I1+1) = - CURNOR(2)                                                     POR04730
400 CONTINUE                                                                POR04740
410 CALL MBSO(N, ISL, IPM, IU, JU, U, DI, B, G, X)                          POR04750
  DIV1=CUR(1)**2 + CUR(2)**2                                                 POR04760
  ZREAL = (G(1)*CUR(1) + G(2)*CUR(2))/DIV1                                  POR04770
  ZIMAG = (G(2)*CUR(1) - G(1)*CUR(2))/DIV1                                  POR04780
  WRITE (6,503) DHOLD                                                         POR04790
  WRITE(6,172) ZREAL,ZIMAG                                                   POR04800
172 FORMAT(' COMPL IMPED.= ',1P,E12.4,' (J*) ',E12.4)                    POR04810
  IF (IDUMP.EQ.1) THEN                                                       POR04820
*   DUMP V OF ALL NODES                                                       POR04830
  IF (MOD(INT(FRCNT),IPPD).EQ.0) WRITE (9,182) (G(I1),I1=1,N)            POR04840
182 FORMAT (1P,6E13.4)                                                       POR04850
  ENDIF                                                                       POR04860
  CALL CURCAL (A,G,MODEL)                                                    POR04870
  WRITE (6,420)REAL1,RMAG1                                                    POR04880
420 FORMAT (' CALCULATED CURRENT = ',1P,E12.2,' (J*) ',E12.2)            POR04890
C NORMALIZE IMPEDANCE TO GIVE 1 CM**2 READING:                              POR04900
C                                                                           POR04910
  ZRLNOR = ZREAL / SCALE                                                     POR04920
  ZIMNOR = ZIMAG / SCALE                                                     POR04930
  WRITE (6,190) SCALE                                                         POR04940
190 FORMAT (' NORMALIZED 1 SQ CM IMPEDANCE BY ',F8.0,' NORM FACTOR:')  POR04950
  WRITE (6,172) ZRLNOR,ZIMNOR                                               POR04960
C                                                                           POR04970
C                                                                           POR04980
C                                                                           POR04990
C DOCUMENTATION PAGE                                                         POR05000
C                                                                           POR05010

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      II=1
      WRITE(7,500)
500  FORMAT(' ',/, ' SUMMARY :')
      WRITE(7,124) IM,JM,KHOLD,POR
      IF (KMSTOP.NE.KHOLD) WRITE (6,502) KMSTOP
502  FORMAT (' ',/, ' PROBLEM TRUNCATED AT PLANE # ',12,/)
      WRITE(7,505) FREQ
503  FORMAT (' DSEED = ',F13.0,'D0')
505  FORMAT(' FREQ = ',1P,E12.3)
      WRITE (7,420)REALI,RMAGI
C ----- CHECK FOR NUMERICAL ERROR -----
      TOL = .03
      DEVRL = (ABS(CUR(1)-REALI)) / CUR(1)
      DEVIM = (ABS(CUR(2)-RMAGI)) / CUR(2)
C IS THE DEVIATION BETWEEN CURR IN AND CURR OUT LT ALLOWED TOLERANCE?
      IF (DEVRL.LT.TOL.AND.DEVIM.LT.TOL) GOTO 610
      WRITE (6,602) DEVRL,DEVIM
      WRITE (7,602) DEVRL,DEVIM
602  FORMAT (' ***** DEVIATION OF CURRENT - REAL + IMAG = ',
1     2P,2F6.1,' PERCENT')
      IBOMB = 1
C ----- END OF ERROR CHECK -----
610  WRITE (7,190) SCALE
      WRITE (7,172) ZRLNOR,ZIMNOR
C ---- MAINTAIN DATABASE -----
      WRITE (8,650) FREQ,ZRLNOR,ZIMNOR
650  FORMAT (' 10',1P,E12.3,2E12.4)
C ----- END DATABASE ENTRY -----
C
C      CALL CURPRF (A,LBLA,G)
700  CONTINUE
C
C      FINISHED A GROUP OF TRIALS AT A GIVEN POROSITY AND A GIVEN FREQ.
C
C      CALCULATE TIMES:
C
C      CALL DATTIM (EVCPU,ETIME,ETCPU)
C
C      CONNOW = ETIME/60.0
      TIMNOW = ETCPU/60.0
      CONTIM = CONNOW - CONLAS
      RUNTIM = TIMNOW - TIMLAS
C
      IF (CONTIM.GT.0) GOTO 720
      WRITE (7,710)
710  FORMAT (' ACCOUNTING BOUNDARY CROSSSED-- INVALID TIME DATA')
      STIME = 0.0
      GOTO 750

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POR05360
POR05370
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POR05420
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POR05450
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POR05470
POR05480
POR05490
POR05500

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720 WRITE (7,725) CONTIM,RUNTIM
725 FORMAT (' ELAPSED TIME = ',F7.2,' CPU TIME = ',F7.2,' MINUTES')
C
C
C   NOW STORE THESE VALUES FOR NEXT TIME AROUND
750 CONLAS = CONNOW
    TIMLAS = TIMNOW
C
*****
*   USER REQUESTED ABORT?? -MAY BE DONE BY SETTING LOC X"464 NONZERO
760 IF (ITYBIT(0).NE.0) THEN
    WRITE (7,751)
    WRITE (6,751)
751   FORMAT (' USER INTERRUPT IN HEXLOC X"464. PROG TERMINATING')
    GOTO 900
    ENDIF
*****
C   NEXT POR & NEXT FREQ
    FRCNT=FRCNT + 1
    FREQ = FRBEG * 10 ** (FRCNT/IPPD)
    IF (FREQ.LE.FREND) GOTO 130
    WRITE(6,799)
799  FORMAT('NEXT PROBLEM:')
820  NRPT = NRPT+1
C   RESET KM FOR NEXT PROBLEM
C
    KM = KHOLD
    IF (NRPT.LE.IRPT) GOTO 120
    POR = POR + PORINC
    IF (POR.LE.POREND) GOTO 120
C   END OF THIS DATA SET
    TT = TIMNOW - STIME
    WRITE (7,850) TT
850  FORMAT (' TOTAL CPU TIME FOR THIS PROBLEM WAS: ',F7.2,' MINUTES')
C
C   PROGRAM IS TERMINATED BY EOF CONDITION ON INPUT DATASET
C   AT EOF IN THE NEXT READ, CONTROL IS TRANSFERRED TO 900
    GOTO 5
900  IF (IBOMB.EQ.0) GOTO 910
    WRITE (6,905)
    WRITE (7,905)
905  FORMAT (' CHECK INPUT DATA... CALCULATION INCORRECT!!!')
910  CONTINUE
    STOP
    END
*-----
*           04/02/85 -- IMPORTANT BUG FIX:
*   RANDOM PTS I,J,K ARE CORRECTLY CHOSEN IN RANPT NOW.
*

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POR05690
POR05700
POR05710
POR05720
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POR05750
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POR05800
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POR05990
POR06000

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C      SUBROUTINE NODES (A,LBLA,POR,DSEED,MMODEL)
C      SIZE OF A IS (IM,JM,KM)
C      REVISED 2/1/83
C
C      --THIS SUBROUTINE SETS UP THE PHYSICAL MODEL
C      THERE ARE A FEW CHOSABLE OPTIONS AS TO WHAT MODEL IS DESIRED:
C      MODEL 1 ==> STANDARD MODEL. MAKE SURE THAT
C                  ELECTRODE IS CONTINUOUS AND THAT ELECTROLYTE
C                  FLOW IS CONTINUOUS ("CUBES" MODEL)
C
C      MODEL 2 ==> METAL ELECTRODE IS CONSTRUCTED CONTINUOUS
C                  BUT ELECTROLYTE PLACED EVERYWHERE ELSE
C                  ("SPHERES MODEL")
C
C      MODEL 3 ==> ALTERNATE MODEL, CONSTRUCTED IN SUBROUTINE
C                  "LATSET".
C
C      MODEL 4 ==> METAL ELECTRODE NOT CONTINUOUS.
C                  ELECTROLYTE PLACED IN ALL NONMETAL SITES
C                  (SPHERES MODEL AGAIN).
C                  (FOR USE WITH RESISTORS AND CAPACITORS)
C
C      ---- IF PERIODIC BOUND CONDITIONS ARE REQUESTED, THEN 50 HAS BEEN
C            ADDED TO THE VALUE OF THE FLAG 'MODEL'
C
C      INTEGER*4 A(IM,JM,KM),LBLA(IM,JM,KM)
C      REAL *8   DSEED
C      COMMON/LATICE/IM,JM,KM,KMSTOP
C
C      MODEL = MMODEL
C      IF (MODEL.GT.50) MODEL = MMODEL-50
C
C      KMAX = KM-1
C      KM/N = 2
C      DO 5 I= 1,IM
C      DO 5 J= 1,JM
C      DO 5 K= 1,KM
C      A(I,J,K)=0
C 5  CONTINUE
C      ISP=IM*JM*(KM-2)
C      MB=(1-POR)*ISP
C      WRITE (6,10) MMODEL
C      WRITE (7,10) MMODEL
C 10  FORMAT (' USING MODEL NUMBER ',12)
C      WHICH MODEL ARE WE USING?
C      GOTO (20,20,15,17      ),MODEL
C      =====
C*  MODEL 3- USE ALTERNATE LATTICE SETUP ROUTINE (NON RANDOM)

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POR06080
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POR06100
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POR06400
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POR06470
POR06480
POR06490

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15 CALL LATSET(A, POR, MODEL)
   GOTO 505
C =====
C MODEL 4 - NO CONTIN. METAL
17 KMIN = 2
   KMAX = KM - 1
   CALL PLANE (A, 1, 5)
   CALL PLANE (A, KM, 5)
   CALL RNDM(A, MB, DSEED, KMIN, KMAX, 1)
   GOTO 505
C =====
C MODELS 1 & 2 -- OLD FASHIONED WAY
20 CALL RNDM(A, MB, DSEED, KMIN, KMAX, 5)
C SET BOTTOM PLANE = ELECTROLYTE ; TOP PLANE = METAL
   CALL PLANE(A, 1, 2)
   CALL PLANE (A, KM, 1)
C *****
C NOW CHECK FOR CONTINUATION OF METAL --NO ISLANDS ALLOWED
C THE (KM-1) PLANE IS ALL CONNECTED TO THE TOP METAL
   DO 50 I=1, IM
   DO 50 J=1, JM
   IF (A(I, J, KM-1).EQ.5) A(I, J, KM-1)=1
50 CONTINUE
C
60 KFND=0
C THIS LOOP LOOPS DOWN FROM K=(KM-2) TO K=2
   DO 400 KT=4, KM
   K=KM-KT+2
C
C FIRST CONNECT FROM ABOVE
   DO 75 I=1, IM
   DO 75 J=1, JM
   IF (A(I, J, K).NE.5) GOTO 75
   IF (A(I, J, K+1).EQ.1) A(I, J, K)=1
75 CONTINUE
C NOW GO CONNECT METAL TOGETHER ON THE REST OF KTH PLANE
100 CALL CONECT(A, K, 5, 1, IFND, KFND)
C FINISHED PLANE YET ?
   IF (IFND.NE.0) GOTO 100
C IF FINISHED, GO TO NEXT PLANE
400 CONTINUE
C
C FINISHED GOING THRU ALL PLANES ...GO AAIN?
   IF (KFND.NE.0) GOTO 60
C
C FINISHED CONNECTIN ALL METAL; NOW DEAL W/ ISOLATED PTS
C ICTR COUNTS # OF POINTS TO BE REPLACED

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POR06500
POR06510
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POR06590
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POR06680
POR06690
POR06700
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POR06800
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POR06870
POR06880
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POR06900
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POR06920
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POR06940
POR06950
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POR06970
POR06980
POR06990
POR07000

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      ICTR=0
      KK=KM-2
      DO 500 K=2, KK
      DO 500 I=1, IM
      DO 500 J=1, JM
      IF (A(I,J,K).NE.5) GOTO 500
C     THIS IS AN ISLAND OF METAL
      A(I,J,K)=0
450  CALL RANPT(A,DSEED,KMIN,KMAX,IT,JT,KT)
C     IS THIS REPLACEMENT PT OK?
      IFND=0
      CALL NEARN(A,IT,JT,KT,0,1,IFND,KFND)
      IF (IFND.NE.1) GOTO 450
      ICTR=ICTR+1
      WRITE (6,460) I,J,K,IT,JT,KT
460  FORMAT (' PT # ',313,' REPLACED BY ',313)
500  CONTINUE
      WRITE (6,501) ICTR
501  FORMAT (' # OF PTS REPLACED= ',16)
C
C     =====
C     NOW FIND ELECTROLYTE CONNECTIONS
505  CONTINUE
C     WE HAVE TWO METHODS--- 1) FILL IN ONLY CONNECTING ELEC PATHS
C                             2) FILL IN ELEC WHEREVER THERE IS NO METAL
      GOTO (540,515,540,515),MODEL
C     =====
C     WE HAVE CHOSEN MODEL #2 OR #4 -- FILL IN ALL NON-METAL
C     SPACE WITH ELECTROLYTE
515  DO 520 K = KMIN,KM
520  CALL PLANE (A,K,2)
      CALL FILABL(A,LBLA)
      RETURN
C     =====
C     MODEL #1 - WATCH FOR ELECTROLYTE FLOW
540  CALL PLANE (A,2,2)
C     NOW GO THROUGH ALL PLANES BOTTOM TO TOP
550  KFND=0
      KT=KM-1
      DO 700 K=3,KT
C     FIRST MAKE CONNECTIONS FROM BELOW
      DO 600 I= 1, IM
      DO 600 J= 1, JM
      IF (A(I,J,K).NE.0) GOTO 600
      IF (A(I,J,K-1).NE.2) GOTO 600
      A(I,J,K) = 2
600  CONTINUE
C
C     NOW DEAL WITH NN ON PLANE

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POR07010
POR07020
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POR07320
POR07330
POR07340
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POR07420
POR07430
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POR07460
POR07470
POR07480
POR07490

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610	CALL CONECT(A,K,0,2,IFND,KFND)	POR07500
	IF (IFND.NE.0) GOTO 610	POR07510
C	NOW GO TO NEXT PLANE	POR07520
700	CONTINUE	POR07530
C	FINISHED GOING THROUGH ALL PLANES. DO IT AGAIN?	POR07540
	IF (KFND.NE.0) GOTO 550	POR07550
	CALL FILABL(A,LBLA)	POR07560
	RETURN	POR07570
	END	POR07580
	-----	POR07590
C		POR07600
	SUBROUTINE CONECT(A,K,ISTAY,ILOOK,IFND,KFND)	POR07610
C	SIZE OF A IS (IM,JM,KM)	POR07620
C	SIZE OF G IS (ISM,7,2) WHERE ISM=IM*JM*KM-1	POR07630
C		POR07640
C	SR TO SCAN A PLANE AT K FROM THE 4 DIFFERENT CORNERS OF THE PLANE	POR07650
	INTEGER*4 A(IM,JM,KM)	POR07660
	REAL *8 DSEED	POR07670
	COMMON/LATICE/IM,JM,KM,KMSTOP	POR07680
C	IF A PREVIOUS CALL TO NEARN FOUND NO NEW VALUES THEN WE'RE FINISHED	POR07690
C	IFND IS THE FLAG TO INDICATE WHETHER A NEW VALUE HAS BEEN FOUND	POR07700
	IFND=0	POR07710
	DO 325 I= 1,IM	POR07720
	DO 325 J= 1,JM	POR07730
	CALL NEARN(A,I,J,K,ISTAY,ILOOK,IFND,KFND)	POR07740
325	CONTINUE	POR07750
	IF (IFND.NE.1) GOTO 400	POR07760
	IFND=0	POR07770
	DO 335 J= 1,JM	POR07780
	DO 335 I= 1,IM	POR07790
	IT=IM-I+1	POR07800
	JT=JM-J+1	POR07810
	CALL NEARN(A,IT,JT,K,ISTAY,ILOOK,IFND,KFND)	POR07820
335	CONTINUE	POR07830
	IF (IFND.NE.1) GOTO 400	POR07840
	IFND=0	POR07850
	DO 345 I= 1,IM	POR07860
	DO 345 J= 1,JM	POR07870
	JT=JM-J+1	POR07880
	CALL NEARN(A,I,JT,K,ISTAY,ILOOK,IFND,KFND)	POR07890
345	CONTINUE	POR07900
	IF (IFND.NE.1) GOTO 400	POR07910
	IFND=0	POR07920
	DO 355 J= 1,JM	POR07930
	DO 355 I= 1,IM	POR07940
	IT=IM-I+1	POR07950
	CALL NEARN(A,IT,J,K,ISTAY,ILOOK,IFND,KFND)	POR07960
355	CONTINUE	POR07970
400	CONTINUE	POR07980

```

      RETURN
      END
*-----
C
      SUBROUTINE RNDM(A,MB,DSEED,KMIN,KMAX,IVAL)
C
C      SUBROUTINE TO GENERATE RANDOM LATTICE
C
      REAL *8 DSEED
      INTEGER*4 A(IM,JM,KM)
      COMMON/LATICE/IM,JM,KM,KMSTOP
C
C
      DO 100 N=1,MB
10  CALL RANPT(A,DSEED,KMIN,KMAX,I,J,K)
      IF (A(I,J,K).NE.0) GOTO 10
C  PLACE METAL ON NODE (PLACE THE VALUE IVAL ON A(I,J,K))
      A(I,J,K) = IVAL
100 CONTINUE
      RETURN
      END
*-----
C
      SUBROUTINE PLANE(A,IZ,ICONST)
C  S.R. TO SET A WHOLE PLANE AT SOME VALUE K=IZ TO A CONSTANT
C
      INTEGER*4 A(IM,JM,KM)
      COMMON/LATICE/IM,JM,KM,KMSTOP
C
      DO 50 I =1,IM
      DO 50 J =1,JM
      IF (A(I,J,IZ).NE.0) GOTO 50
      A(I,J,IZ) = ICONST
50  CONTINUE
      RETURN
      END
*-----
C
      SUBROUTINE NEARN(A,I,J,K,ISTAY,ILOOK,IFND,KFND)
C
C  S.R. TO LOOK AT NEAREST NEIGHBORS FOR VALUE 'ILOOK'
C  IF 'ILOOK' IS FOUND IN ONE OF NN THEN A(I,J,K)=ILOOK TOO
      INTEGER*4 A(IM,JM,KM)
      COMMON/LATICE/IM,JM,KM,KMSTOP
C
      IF (A(I,J,K).NE.ISTAY) GOTO 400
      IF (I-1.LT.1) GOTO 20
      IF (A(I-1,J,K).EQ.ILOOK) GOTO 300
20  IF (I+1.GT.IM) GOTO 30

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POR07990
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POR08470

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      IF (A(I+1,J,K).EQ.ILOOK) GOTO 300
30  IF (J-1.LT.1) GOTO 40
      IF (A(I,J-1,K).EQ.ILOOK) GOTO 300
40  IF (J+1.GT.JM) GOTO 50
      IF (A(I,J+1,K).EQ.ILOOK) GOTO 300
50  IF (K-1.LT.1) GOTO 60
      IF (A(I,J,K-1).EQ.ILOOK) GOTO 300
60  IF (K+1.GT.KM) GOTO 100
      IF (A(I,J,K+1).EQ.ILOOK) GOTO 300
C NO NN VALUE OF ILOOK FOUND:
100 GOTO 400
C ONE OF ADJACENT NODES = ILOOK
300 A(I,J,K) = ILOOK
      IFND=1
      KFND=1
400 RETURN
      END
-----
C
      SUBROUTINE RANPT(A,DSEED,KMIN,KMAX,I,J,K)
C
C SUBROUTINE TO GENERATE RANDOM LATTICE PT (I,J,K)
* NUMBER BETWEEN [KMIN,KMAX] = INT(R(1)*{KMAX-KMIN+1}+KMIN)
      REAL *8 DSEED
      INTEGER*4 A(IM,JM,KM)
      DIMENSION R(3)
      COMMON/LATICE/IM,JM,KM,KMSTOP
C
      CALL GGUBS(DSEED,3,R)
      I=INT(R(1)*IM + 1.0)
      J=INT(R(2)*JM + 1.0)
      K=INT(R(3)*{KMAX-KMIN+1} + KMIN)
      RETURN
      END
-----
C
      SUBROUTINE CHOP (A)
C
C SUBROUTINE TO EFFECTIVELY CHOP OFF THE UPPER PLANES
C AND ELIMINATE THEM FROM OUR PROBLEM
C IF THERE IS LITTLE ELECTROLYTE PENETRATION THERE.
C
      INTEGER*4 A(IM,JM,KM)
C
      COMMON/LATICE/IM,JM,KM,KMSTOP
      COMMON/ELEC/Z(6,2),CUR(2),NCUR,NGND,REALI,RMAGI
C
C CHANGE KM TO KMSTOP
      KM = KMSTOP
C SET GND NODE TO NEW LOCATION
C IN LOWER RT HAND CORNER OF PLANE # KMSTOP

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POR08480
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POR08670
POR08680
POR08690
POR08700
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POR08800
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POR08890
POR08900
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POR08960
POR08970
POR08980

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      NGND = IM*JM*KM
C
C CHECK TO SEE THAT CORNER NODE IS CONNECTED TO METAL
C
      IF (A(IM,JM,KM).EQ.1) GOTO 100
      IF (A(IM,JM,KM).EQ.2) WRITE (6,20)
20  FORMAT (' ELECTROLYTE AT CORNER TOP PLANE DISPLACED FOR GND')
      DO 50 I=1,IM
      DO 50 J=1,JM
      II = IM-I+1
      JJ = JM-J+1
      A(II,JJ,KM) = 1
      IFND = 0
      CALL NEARN(A,II,JJ,KM,1,1,IFND,KFND)
C IFND IS SET (=1) IF THIS PT IS CONNECTED TO
C ANOTHER METAL.
      IF (IFND.NE.0) GOTO 100
50  CONTINUE
C
100 WRITE (6,175) KMSTOP
175 FORMAT (' ,//,' LATTICE TRUNCATED ABOVE PLANE # ',13)
C COMPUTE NEW STATISTICS; NO NEED TO RE-DISPLAY LATTICE
C
      ISHOW = 0
C
      CALL DISPLA (A,ISHOW,MODEL)
      RETURN
      END
*-----
C
C
      SUBROUTINE DISPLA (A,ISHOW,MODEL)
      INTEGER*4 A(IM,JM,KM)
      DIMENSION LBL(7),LAB1(20)
      COMMON/LATTICE/IM,JM,KM,KMSTOP
      DATA LBL/' ','M','E','X','X','X','B'/
C
C ISHOW = 1 IS A FLAG THAT PRINTS OUT A PICTURE OF
C THE LATTICE
C ISHOW = 0 IS A FLAG THAT JUST CAUSES PRINT OUT OF STATISTICS
      WRITE (6,4)
4  FORMAT(' KEY : " "-AIR M-METAL E-ELECTROLYTE')
C INITIALIZE COUNTERS FOR DISTRIBUTION TOTALS
      NMET = 0
      NELEC = 0
      NAIR = 0
      NSC = 0
C
C NOW GO THROUGH EACH PLANE :

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POR08990
POR09000
POR09010
POR09020
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POR09040
POR09050
POR09060
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POR09080
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POR09120
POR09130
POR09140
POR09150
POR09160
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POR09340
POR09350
POR09360
POR09370
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POR09390
POR09400
POR09410
POR09420
POR09430
POR09440
POR09450
POR09460
POR09470
POR09480
POR09490

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C      DO 600 K=KM,1,-1
C      *****
C      NOW COMPUTE THE DISTRIBUTIONS ON THIS PLANE :
C
      ICONT = 0
      IF (MODEL.GT.50) ICONT = 1
      KMET=0
      KELEC=0
      KSC=0
      DO 250 I=1,IM
      DO 250 J=1,JM
      IAT = A(I,J,K)
      IF (IAT.EQ.0) GOTO 250
      IF (IAT.EQ.1) KMET=KMET+1
      IF (IAT.EQ.2) KELEC=KELEC+1
C      NOW CHECK IN THE 3 FORWARD DIRECTIONS FOR SC INTFC
      IF (I+1.GT.IM) THEN
        IF (ICONT.EQ.1) THEN
          NEXT = A(I,J,K)
          IF (IAT+NEXT.EQ.3) KSC = KSC + 1
        ENDIF
      ELSE
        NEXT = A(I+1,J,K)
        IF (IAT+NEXT.EQ.3) KSC = KSC + 1
      ENDIF
      IF (J+1.GT.JM) THEN
        IF (ICONT.EQ.1) THEN
          NEXT = A(I,1,K)
          IF (IAT+NEXT.EQ.3) KSC = KSC + 1
        ENDIF
      ELSE
        NEXT = A(I,J+1,K)
        IF (IAT+NEXT.EQ.3) KSC = KSC + 1
      ENDIF
140 IF (K+1.GT.KM) GOTO 250
      IF (IAT+A(I,J,K+1).EQ.3) KSC=KSC+1
C
C      250 CONTINUE
      KAIR = (IM*JM) - KMET - KELEC
C
C      NOW ACCUMULATE TALLY FOR WHOLE LATTICE
      NMET = NMET + KMET
      NELEC = NELEC + KELEC
      NAIR = NAIR + KAIR
      NSC = NSC + KSC
C      END PLANE DISTRIBUTION CALC
C      *****
C      ---- IF ISHOW IS SET THEN DISPLAY LATTICE----
C

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POR09500
POR09510
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POR09580
POR09590
POR09600
POR09610
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POR09630
POR09640
POR09650
POR09660
POR09670
POR09680
POR09690
POR09700
POR09710
POR09720
POR09730
POR09740
POR09750
POR09760
POR09770
POR09780
POR09790
POR09800
POR09810
POR09820
POR09830
POR09840
POR09850
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POR09980
POR09990

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        GOTO 600
22 WRITE(6,52) (LAB1(J), J=1,JM)
52 FORMAT (T28,20A2)
        GOTO 600
23 WRITE(6,53) (LAB1(J), J=1,JM)
53 FORMAT (T27,20A2)
        GOTO 600
24 WRITE(6,54) (LAB1(J), J=1,JM)
54 FORMAT (T26,20A2)
        GOTO 600
25 WRITE(6,55) (LAB1(J), J=1,JM)
55 FORMAT (T25,20A2)
        GOTO 600
26 WRITE (6,56) (LAB1(J),J=1,JM)
56 FORMAT (T24,20A2)
        GOTO 600
27 WRITE (6,57) (LAB1(J),J=1,JM)
57 FORMAT (T23,20A2)
        GOTO 600
28 WRITE (6,58) (LAB1(J),J=1,JM)
58 FORMAT (T22,20A2)
        GOTO 600
29 WRITE (6,59) (LAB1(J),J=1,JM)
59 FORMAT (T21,20A2)
600 CONTINUE
C SUMMARY
C
      IS=IM*JM*KM
C COMPUTE TOTAL # INTFC SITES POSSIBLE
      INTOT = (KM-2)*(IM*(JM-1) + JM*(IM-1)) + IM*JM*(KM-1)
      PAREA = IM*JM
      ROUGH = NSC/PAREA
      WRITE (6,700) NELEC,NAIR,NMET,NSC, ROUGH
      WRITE (7,700) NELEC,NAIR,NMET,NSC ,ROUGH
700 FORMAT ('TOTALS: (E/A/M)=',I4,'/',I4,'/',I4,10X,'# SC INTFC=',
116,' ROUGH. = ',F5.2)
      WRITE(6,710) IS,INTOT
      WRITE(7,710) IS,INTOT
710 FORMAT(' TOTAL LATTICE PTS - ',I6,5X,'# INTERFACE SITES- ',I6)
      RETURN
      END
-----
C
C
      SUBROUTINE FILABL(A,LBLA)
      INTEGER*4 A(IM,JM,KM),LBLA(IM,JM,KM)
      COMMON/LATICE/IM,JM,KM,KMSTOP
*
* INITIALIZE ARRAY LBLA TO REFLECT LATTICE NODE #'S
*

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POR10490
POR10500
POR10510
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POR10530
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POR10600
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POR10620
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POR10690
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POR10980

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*
* ICNTR COUNTS THE NUMBER OF NONZERO NODES IN THE LATTICE
  ICNTR = 0
  DO 100 K=1, KM
  DO 100 J=1, JM
  DO 100 I=1, IM
*   -- IF A(I,J,K) = 0 THEN LBLA IS ALSO 0
    LBLA(I,J,K) = A(I,J,K)
    IF (A(I,J,K).NE.0) THEN
      ICNTR = ICNTR + 1
      LBLA(I,J,K) = ICNTR
    ENDIF
C
C 100 CONTINUE
  RETURN
  END
*-----*
*
*
  SUBROUTINE CONDOC(A,G,JA,IA,B,N,NT,MODEL)
C
C S.R. TO CONSTRUCT G MATRIX   REV 5/28/84
C
  INTEGER*4 A(IM,JM,KM)
  COMMON/DIMS/IDIM1, IDIM2, IDIM3, IDIM4
  COMMON/LATICE/IM, JM, KM, KMSTOP
  COMMON/ELEC/Z(6,2), CUR(2), NCUR, NGND, REAL1, RMAG1
  COMMON/GTEMP/ NG1, NG2, NG3, NG4, NG5, ICOL(6), GREAL(6), GIMAG(6)
  DIMENSION G(IDIM3), JA(IDIM3)
  DIMENSION IA(IDIM2), B(IDIM2)
C
C SEE SUBROUTINE NODES FOR THE SIGNIFICANCE OF MODEL
C
C ---> HERE-IF MODEL > 50 THEN PERIODIC BOUNDARY CONDITIONS
C ---> HAVE BEEN REQUESTED
C
  ICONT = 0
  IF (MODEL.GT.50) ICONT = 1
C
C G IS COEFFICIENT MATRIX STORED IN SPARSE-SYMMETRIC MODE
C JA IS COLUMN ADDRESSES OF EACH ELEMENT OF PACKED G MATRIX
C IA IS THE ROW POINTER TO THE BEGINNING OF EACH ROW--
C   IA(N) CONTAINS THE INDEX OF THE ELEMENT IN G THAT
C   STARTS THE N'TH ROW OF MATRIX G
C B IS THE RHS OF THE SIMULTANEOUS EQU.
C
  DO 50 I=1, IDIM3
  G(I)=0.0

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POR11370
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POR11390
POR11400
POR11410
POR11420
POR11430
POR11440
POR11450
POR11460
POR11470

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```

C ICOL(11) WILL CONTAIN COLUMN INFORMATION ABOUT THE ADJ. NODES
C IF ICOL(11)=0 THEN EITHER: 1-THERE IS NO NODE THERE (AIR)
C                                     OR: 2-IT BELONGS TO LOWER HALF TRIANGLE
C                                     OF THE MATRIX--LEAVE OUT
C                                     OR: 3-IT IS THE GROUND NODE
C
DO 160 I1=1,6
  ICOL(11) = 0
160 CONTINUE
  IZERO = 0
  ITEMP=0
C HOW MANY ZERO ELEMENTS ARE IN THE REST OF THIS ROW?
  IF (J.EQ.JM) GOTO 162
  DO 175 JJ=J,JM
  IF (A(I,JJ,K).EQ.0) ITEMP=ITEMP+1
175 CONTINUE
C
162 IF (J+1.GT.JM) GOTO 170
  IF (A(I,J+1,K).EQ.0) GOTO 170
  IF (NNA+1.EQ.NGND) NG2 = N
  IF (NNA+1.EQ.NGND) GOTO 170
  ICOL(2) = N + 2
170 IF (I+1.GT.IM) GOTO 190
  IF (NNA+JM.EQ.NGND) NG1 = N
  IF (NNA+JM.EQ.NGND) GOTO 190
  IF (A(I+1,J,K).EQ.0) GOTO 190
C HOW MANY ELEMENTS BTWN HERE & NODE IN FRONT OF THIS (NON-ZERO)
  IZERO=ITEMP
  I1 = I + 1
  DO 180 JJ=1,J
  IF (A(I1,JJ,K).EQ.0) IZERO=IZERO+1
180 CONTINUE
  ICOL(3) = N + 2*(JM-IZERO)
190 IF (K+1.GT.KM) GOTO 220
  IF ((NNA+IM*JM).EQ.NGND) NG3 = N
  IF ((NNA+IM*JM).EQ.NGND) GOTO 220
  IF (A(I,J,K+1).EQ.0) GOTO 220
C HOW MANY ZEROS BTWN HERE & NODE ON TOP?
C FIRST COUNT ZEROS ON REST OF CURRENT PLANE
  IZERO=ITEMP
C IF (IZERO.GT.0) WRITE (6,997) N, IZERO
C 997 FORMAT (' LINE # ',13, ' CKPT#1----IZERO=',13)
  IF (I+1.GT.IM) GOTO 202
  ISTART=I+1
  DO 200 I1=ISTART,IM
  DO 200 JJ=1,JM
  IF (A(I1,JJ,K).EQ.0) IZERO=IZERO+1
200 CONTINUE
C IF (IZERO.GT.0) WRITE (6,998) N, IZERO
C 998 FORMAT (' LINE # ',13, ' CKPT#2----IZERO=',13)
C NEXT PLANE UP

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POR12970
POR12980
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POR13110
POR13120
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POR13140
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POR13160
POR13170
POR13180
POR13190
POR13200
POR13210
POR13220
POR13230
POR13240
POR13250
POR13260
POR13270
POR13280
POR13290
POR13300
POR13310
POR13320
POR13330
POR13340
POR13350
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POR13370
POR13380
POR13390
POR13400
POR13410
POR13420
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POR13440
POR13450
POR13460
POR13470

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202 KK=K+1
IEND=I-1
IF (IEND.LT.1) GOTO 207
DO 205 I1=1, IEND
DO 205 JJ=1, JM
IF (A(I1, JJ, KK).EQ.0) IZERO=IZERO+1
205 CONTINUE
C IF (IZERO.GT.0) WRITE (6,999) N, IZERO
C 999 FORMAT (' LINE #', I3, CKPT#3---IZE:0=', I3)
207 DO 210 JJ=1, J
IF (A(I1, JJ, KK).EQ.0) IZERO=IZERO+1
210 CONTINUE
C IF (IZERO.GT.0) WRITE (6,996) N, IZERO
C 996 FORMAT (' LINE #', I3, CKPT#4---IZE:0=', I3)
C ICOL(5) = N + 2*(IM*JM-IZERO)
C *****
C PERIODIC BOUNDARY CONDITIONS??
C 220 IF (ICONT.NE.1) GOTO 250
* CONTINUOUS BOUNDARY CONDITIONS -
* IN THIS CASE, NODES IN RELATIVE POS #1 AND #4 ARE IN UPR RH TRIANGLE
* OF THE MATRIX.
* E.G. FOR SIZE = 11 NODE #1 HAS AN ADJACENT NODE TO THE LEFT WITH
* MATRIX ADDRESS (1,11).
* IF (J-1.LT.1) THEN
* WE ARE ON LH EDGE
* IF (A(I, JM, K).NE.0) THEN
* IF (NNA+JM-1.EQ.NGND) THEN
* NG4 = N
* ELSE IZERO = ITEMP
* ICOL(4) = N + 2 * (JM-IZERO-1)
* END IF
* END IF
* IF (I-1.LT.1) THEN
* IF (A(I-1, J, K).NE.0) THEN
* IF (NNA+JM*(IM-1).EQ.NGND) THEN
* NG5 = N
* ELSE
* IZERO = ITEMP
* DO 225 I1 = I+1, IM
* DO 225 JJ = 1, JM

```

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POR13680
POR13690
POR13700
POR13710
POR13720
POR13730
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POR13780
POR13790
POR13800
POR13810
POR13820
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POR13900
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POR13940
POR13950
POR13960

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                IF (I1.EQ.IM.AND.JJ.GE.J) GOTO 225
                IF (A(I1,JJ,K).EQ.0) IZERO = IZERO + 1
                CONTINUE
                ICOL(1) = N. + 2 * (JM*(IM-1)-IZERO)
            END IF
        END IF
    END IF
C
* BECAUSE OF PERIODIC B.C. COLUMN INDICES MAY BE OUT OF ORDER
* SORT SO THAT THEY ARE IN ASCENDING ORDER
*
    DO 240 LL = 1,5
        DO 240 IN = LL+1,6
            IF (ICOL(LL).GT.ICOL(IN)) THEN
                SWAP
                    IHCOL = ICOL(LL)
                    TEMPGR = GREAL(LL)
                    TEMPGI = GIMAG(LL)
                    ICOL(LL) = ICOL(IN)
                    GREAL(LL) = GREAL(IN)
                    GIMAG(LL) = GIMAG(IN)
                    ICOL(IN) = IHCOL
                    GREAL(IN) = TEMPGR
                    GIMAG(IN) = TEMPGI
                END IF
            240 CONTINUE
C NOW START THE 1ST ROW OF REAL-IMAG PAIR
    250 NT=NT+1
C POINTER TO FIRST ELEMENT IN ROW
    IA(N) = NT
C COLUMN INDEX
    JA(NT) = N
C NOW MATRIX ITSELF
    G(NT) = GREAL
    IF (GIMAGO.EQ.0.0) GOTO 380
C INCREMENT POSITION IN G MATRIX
    NT = NT + 1
    JA(NT) = N + 1
    G(NT) = -GIMAGO
C
    380 DO 390 LL=1,6
        IF (ICOL(LL).EQ.0) GOTO 390
        NT = NT + 1
        JA(NT) = ICOL(LL)
        G(NT) = GREAL(LL)
        IF (GIMAG(LL).EQ.0.0) GOTO 390
        NT = NT + 1
        JA(NT) = ICOL(LL)+1

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POR13970
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POR14110
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POR14130
POR14140
POR14150
POR14160
POR14170
POR14180
POR14190
POR14200
POR14210
POR14220
POR14230
POR14240
POR14250
POR14260
POR14270
POR14280
POR14290
POR14300
POR14310
POR14320
POR14330
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POR14370
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POR14390
POR14400
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POR14420
POR14430
POR14440
POR14450

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      G(NT) = -GIMAG(LL)
390 CONTINUE
C
C DO NEXT LINE (IMAG LINE)
C
      N = N + 1
      NT = NT + 1
      IA(N) = NT
      JA(NT) = N
      G(NT) = -GREALO
C
      DO 400 LL=1,6
      IF (ICOL(LL).EQ.0) GOTO 400
      IF (GIMAG(LL).EQ.0.0) GOTO 395
      NT = NT + 1
      JA(NT) = ICOL(LL)
      G(NT) = -GIMAG(LL)
395 NT = NT + 1
      JA(NT) = ICOL(LL) + 1
      G(NT) = -GREAL(LL)
400 CONTINUE
500 CONTINUE
C MARK LAST ELEMENT
      IA(N+1) = NT + 1
* WRITE (7,499) NG1,NG2,NG3,NG4,NG5
499 FORMAT (' ADJACENT GND NODE MATRIX ADDRESSES: ',3I6)
      WRITE (6,505) NNA,NN,N,NT
505 FORMAT (' TOTAL NODES= ',I5,' NONZERO NODES= ',I5,
1' # OF EQU-S= ',I8,' PACKED ELEMENTS= ',I8)
***** BEGIN CONDITIONAL SECTION *****
C NOW LIST OFF MATRIX
* L=1
* WRITE (6,510) L
* 510 FORMAT (' *****',//,' ROW ',I4)
* DO 580 I=1,NT
* WRITE (6,560) JA(I),G(I)
* 560 FORMAT (' ELEMENT #',I4,' = ',1PE13.4)
* IF (I+1.LT.IA(L+1)) GOTO 580
* L = L + 1
* WRITE (6,510) L
* 580 CONTINUE
***** END CONDITIONAL SECTION *****
C
C SET UP RHS = INPUT CURRENTS
      B(NCUR) = CUR(1)
      B(NCUR+1) = -CUR(2)
      RETURN
      END
-----
C

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POR14460
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POR14610
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POR14630
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POR14680
POR14690
POR14700
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POR14780
POR14790
POR14800
POR14810
POR14820
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POR14890
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POR14940
POR14950

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C          SUBROUTINE NOD146(A, IPOS, IAT, NEIGH, IGND, Z1, Z2)
C          SR TO EVALUATE CONDUCTANCE ELEMENTS AT NODES 1,4,6
C          INTEGER*4 A(IM, JM, KM)
C          COMMON/LATICE/IM, JM, KM, KMSTOP
C          COMMON/ELEC/Z(6,2), CUR(2), NCUR, NGND, REAL1, RMAG1
C          COMMON/GTEMP/ NG1, NG2, NG3, NG4, NG5, ICOL(6), GREAL(6), GIMAG(6)
C          IAGND=A(IM, JM, KM)
C          G1 = Z(IAT, 1)
C          G2 = Z(IAT, 2)
C          NOTHING THERE AT POS IPOS
C          IF (NEIGH.EQ.0) GOTO 100
C          GND INTERFACE?
C          IF (IGND.EQ.NGND) GOTO 10
C          SEMIC INTFC?
C          IF (IAT+NEIGH.EQ.3) GOTO 20
C          NORMAL INTFC
C          GOTO 30
C          GND NODE
10  Z1 = G1 + Z(IAGND, 1)
   Z2 = G2 + Z(IAGND, 2)
   IF (IAT+NEIGH.NE.3) GOTO 100
   Z1 = G1 + Z(NEIGH+2, 1)
   Z2 = G2 + Z(NEIGH+2, 2)
   GOTO 100
C          S.C. INTFC
20  A1 = Z(NEIGH+2, 1) + G1
   A2 = Z(NEIGH+2, 2) + G2
   GOTO 50
C          NORMAL INTFC
30  A1 = Z(NEIGH, 1) + G1
   A2 = Z(NEIGH, 2) + G2
50  GREAL(IPOS) = -A1/(A1**2+A2**2)
   GIMAG(IPOS) = A2/(A1**2+A2**2)
100 RETURN
   END
*-----
C
C          SUBROUTINE NOD235(A, IPOS, IAT, NEIGH, IGND, Z1, Z2)
C          SR TO EVALUATE G MATRIX AT NODES 2,3,5
C          INTEGER*4 A(IM, JM, KM)
C          COMMON/LATICE/IM, JM, KM, KMSTOP
C          COMMON/ELEC/Z(6,2), CUR(2), NCUR, NGND, REAL1, RMAG1
C          COMMON/GTEMP/ NG1, NG2, NG3, NG4, NG5, ICOL(6), GREAL(6), GIMAG(6)
C

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POR14960
POR14970
POR14980
POR14990
POR15000
POR15010
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POR15100
POR15110
POR15120
POR15130
POR15140
POR15150
POR15160
POR15170
POR15180
POR15190
POR15200
POR15210
POR15220
POR15230
POR15240
POR15250
POR15260
POR15270
POR15280
POR15290
POR15300
POR15310
POR15320
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POR15350
POR15360
POR15370
POR15380
POR15390
POR15400
POR15410
POR15420
POR15430
POR15440
POR15450

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```

IAGND=A(IM,JM,KM)
A1=0.0
A2=0.0
G1=Z(IAT,1)
G2=Z(IAT,2)
C NOTHING THERE
C IF (NEIGH.EQ.0) GOTO 100
C GND INTFC?
C IF (IGND.EQ.NGND) GOTO 10
C S.G. INTERFACE?
C IF (IAT+NEIGH.EQ.3) GOTO 20
C NORMAL
C GOTO 30
C
C GND NODE
10 Z1 = G1 + Z(IAGND,1)
   Z2 = G2 + Z(IAGND,2)
   IF (IAT+NEIGH.NE.3) GOTO 100
   Z1 = Z(IAT+2,1) + Z(IAGND,1)
   Z2 = Z(IAT+2,2) + Z(IAGND,2)
   GOTO 100
C SC INTFC
20 G1 = Z(IAT+2,1)
   G2 = Z(IAT+2,2)
30 A1 = Z(NEIGH,1) + G1
   A2 = Z(NEIGH,2) + G2
   GREAL(IPOS) = -A1/(A1**2+A2**2)
   GIMAG(IPOS) = A2/(A1**2+A2**2)
100 CONTINUE
   RETURN
   END
*-----
C SUBROUTINE CURCAL (A,G,MODEL)
C SR TO CALCULATE 'CURRENT OUT' VALUE
C TO CHECK RESULT
C
INTEGER*4 A(IM,JM,KM)
COMMON /DIMS//DIM1, IDIM2, IDIM3, IDIM4
COMMON/LATICE/ IM,JM,KM,KMSTOP
COMMON/ELEC/Z(6,2),CUR(2),NGUR,NGND,REAL1,RMAG1
COMMON/GTEMP/ NG1,NG2,NG3,NG4,NG5,ICOL(6),DUMMY(12)
DIMENSION G(IDIM3)
DEFINE NODE #'S:
1 1-AT (IM-1)
2 2-AT (JM-1)
3 3-AT (KM-1)
4 4-AT JM+1 (FOR CONT BOUND COND.)
5 5-AT IM+1 (FOR COND BOUND COND.)
*
* 2 ---GND--- 4
* / /
*
POR15460
POR15470
POR15480
POR15490
POR15500
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POR15530
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POR15950

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```

      DIV = ZR*ZR + ZI*ZI
      REALI = REALI + (V3R*ZR + V3I*ZI)/DIV
      RMAGI = RMAGI + (V3I*ZR - V3R*ZI)/DIV
150  IF (IA4.EQ.0) GOTO 200
      V4R = G(NG4)
      V4I = G(NG4+1)
      IF (IA4+IAGND.EQ.3) IA4 = IA4+2
      ZR = Z(IA4,1) + Z(IAGND,1)
      ZI = Z(IA4,2)
      DIV = ZR*ZR + ZI*ZI
      REALI = REALI + (V4R*ZR + V4I*ZI)/DIV
      RMAGI = RMAGI + (V4I*ZR - V4R*ZI)/DIV
200  IF (IA5.EQ.0) GOTO 250
      V5R = G(NG5)
      V5I = G(NG5+1)
      IF (IA5+IAGND.EQ.3) IA5 = IA5+2
      ZR = Z(IA5,1) + Z(IAGND,1)
      ZI = Z(IA5,2)
      DIV = ZR*ZR + ZI*ZI
      REALI = REALI + (V5R*ZR + V5I*ZI)/DIV
      RMAGI = RMAGI + (V5I*ZR - V5R*ZI)/DIV
250  CONTINUE
C    WRITE (7,110) V1R,V1I,V2R,V2I,V3R,V3I,V4R,V4I,V5R,V5I
C 110 FORMAT (' V ON NODES = ',//,
C    C5(' ',1P,E12.2,' +(J*) ',E12.2,/)
      RETURN
      END

```

```

*-----*
*****
*
*   STORED ON FILE: CURPRF  FORTRAN  12/5/84
*
*   GENERATES A CURRENT PROFILE OF THE LATTICE
*****
SUBROUTINE CURPRF (A,LBLA,G)

```

```

REAL*4 NRMCUR
CHARACTER*1 LBL(0:3),CHOLD1(0:20),CHOLD2(0:20)
COMPLEX*8 C1
INTEGER*4 A(IM,JM,KM),LBLA(IM,JM,KM)
DATA LBL /'A','M','E','X'/
COMMON /LATTICE/IM,JM,KM,KMSTOP
COMMON /DIMS/IDIM1,IDIM2,IDIM3,IDIM4
COMMON /ELEC/Z(6,2),CUR(2),NCUR,NGND,REALI,RMAGI
DIMENSION G(IDIM3),RTHOLD(20),FHOLD(20)

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POR16450
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POR16600
POR16610
POR16620
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POR16670
POR16680
POR16690
POR16700
POR16710
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POR16940

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POR17440
POR17450
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POR17910
POR17920

PCTCUR = 0.0
ENDIF
WRITE (7,50) NODE1, NODE2, CI
CHOLD1(0) = LBL(IAT1)
RTHOLD(JPNTR) = PCTCUR
CHOLD2(JPNTR) = LBL(IAT2)
WRITE (7,51) RTHOLD, CHOLD2(JPNTR)
FORMAT ( ' ', F7.3, A1)

ENDIF
IF (I.LT.IM) THEN
  NODE2 = LBLA(I+1, J, K)
  IAT2 = A(I+1, J, K)
  IF (IAT2.NE.0.AND. IAT1.NE.0) THEN
    CALL CURLNK(C, NODE1, NODE2, IAT1, IAT2, CI)
    PCTCUR = 100.0 * ABS(CI) / NRMCUR
    IF (IAT1+IAT2.EQ.3.AND. PCTCUR.GT. THRESH) THEN
      PCTCUR = PCTCUR + 1
    ELSE IF (IAT1+IAT2.EQ.3) THEN
      NOCNT=NOCNT+1
    ENDIF
  ELSE
    PCTCUR = 0.0
  ENDIF
ENDIF
WRITE (7,50) NODE1, NODE2, CI
FHOLD(JPNTR) = PCTCUR
WRITE (7,51) FHOLD

ENDIF

100 CONTINUE
WRITE(6,110) CHOLD1(0), (RTHOLD(IX), CHOLD2(IX), IX=1, JM-1)
IF (I.LT.IM) WRITE (6,120) (FHOLD(IX), IX=1, JM)
FORMAT ( ' ', 4X, A1, 1X, 20(F7.3, 1X, A1, 1X))
FORMAT ( ' ', 20(F7.3, 3X))
CONTINUE
WRITE (6,310) K
FORMAT ( ' ', K PLANE = ' ', I2//)
310 FINISHED A WHOLE I-J PLANE, NOW PRINT THE VERTICAL LINKS
** BETWEEN THE CURRENT K PLANE AND K+1:
TOTPCT = 0.0
IF (K.LT.KM) THEN
  DO 350 I=1, IM
    JPNTR = 0
    DO 325 J=1, JM
      NODE1 = LBLA(I, J, K)
      IAT1 = A(I, J, K)
      JPNTR = JPNTR + 1
      NODE2 = LBLA(I, J, K+1)
      IAT2 = A(I, J, K+1)

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```

      IF (IAT2.NE.0.AND.IAT1.NE.0) THEN
      CALL CURLNK(G,NODE1,NODE2,IAT1,IAT2,C1)
      PCTCUR = 100.0 * ABS(C1) / NRMCUR
      IF (IAT1+IAT2.EQ.3.AND.PCTCUR.GT.THRESH) THEN
      INTCNT = INTCNT + 1
      ELSE IF (IAT1+IAT2.EQ.3) THEN
      NOCNT=NOCNT+1
      ENDIF
      ELSE
      PCTCUR = 0.0
      ENDIF
      TOTPCT = TOTPCT + PCTCUR
      CHOLD1(JPNTR) = LBL(IAT1)
      FHOLD(JPNTR) = PCTCUR
      CHOLD2(JPNTR) = LBL(IAT2)
      WRITE (6,50) NODE1,NODE2,C1
*
325  CONTINUE
      WRITE (6,326){CHOLD1(IX),IX=1,JM)
      WRITE (6,327){FHOLD(IX),IX=1,JM)
      WRITE (6,328){CHOLD2(IX),IX=1,JM)
326  FORMAT (' ',4X,20(A1,9X))
327  FORMAT (' ',20(F7.3,3X))
328  FORMAT (' ',20(A1,9X))
350  CONTINUE
      WRITE (6,360) TOTPCT
360  FORMAT (' TOTAL CURRENT (%) = ',F7.3//)
      ENDIF
400  CONTINUE
      WRITE (6,410) INTCNT,NOCNT,INTCNT+NOCNT
410  FORMAT (' ACTIVE INTERFACES = ',I6,' PASSIVE = ',I6,' TOTAL= ',I6)
      RETURN
      END
*-----*
*
*
* SUBROUTINE CURLNK(G,NODE1,NODE2,IAT1,IAT2,C1)
* SUBROUTINE TO CALCULATE THE CURRENT IN A GIVEN CIRCUIT BRANCH
*
*
* COMMON /LATICE/IM,JM,KM,KMSTOP
* COMMON /DIMS/IDIM1, IDIM2, IDIM3, IDIM4
* COMMON /ELEC/Z(6,2),CUR(2),NCUR,NGND,REAL1,RMAG1
* DIMENSION G(IDIM3)
*
* REAL*8 V1R,V2R,V1I,V2I,ZR,ZI
* COMPLEX*8 C1
* COMPLEX*16 CZ,CV1,CV2,CV0

```

```

POR17930
POR17940
POR17950
POR17960
POR17970
POR17980
POR17990
POR18000
POR18010
POR18020
POR18030
POR18040
POR18050
POR18060
POR18070
POR18080
POR18090
POR18100
POR18110
POR18120
POR18130
POR18140
POR18150
POR18160
POR18170
POR18180
POR18190
POR18200
POR18210
POR18220
POR18230
POR18240
POR18250
POR18260
POR18270
POR18280
POR18290
POR18300
POR18310
POR18320
POR18330
POR18340
POR18350
POR18360
POR18370
POR18380
POR18390
POR18400
POR18410

```

```

IATT1 = IAT1
INDX1 = 2*NODE1 - 1
INDX2 = 2*NODE2 - 1
V1R = G(INDX1)
V1I = G(INDX1+1)
V2R = G(INDX2)
V2I = G(INDX2+1)

CV1 = DCMLPX(V1R,V1I)
CV2 = DCMLPX(V2R,V2I)
CV0 = CV2 - CV1
IF (IATT1+IAT2.EQ.3) IATT1 = IATT1+2
ZR = Z(IATT1,1) + Z(IAT2,1)
ZI = Z(IATT1,2) + Z(IAT2,2)
CZ = DCMLPX(ZR,ZI)
*
*      I = V / Z IN COMPLEX ARITHMETIC
*      CI = CV0 / CZ
50  FORMAT (' ',14,' ',14,' CUR (%)= ',1P,E13.3,3X,E13.3,E13.3)
RETURN
END
*-----*
* SUBROUTINE TO CALCULATE LATTICE CORRELATIONS
* TO TRY AND QUANTIFY THE PROPERTIES OF
* THE GEOMETRICAL CHARACTERISTICS OF EACH POROUS ELECTRODE
* -- DENSITY-DENSITY CORRELATION CALCULATION ---
* -- OF THE SURFACE SHELL ---
* WRITTEN ON 10/30/85 IN VS FORTRAN77
* SUBROUTINE SHLCOR (A,LBLA,LPLANE)
*
* PARAMETER (MAXR=100)
* INTEGER A(IM,JM,KM),LBLA(IM,JM,KM)
* COMMON /LATTICE/ IM,JM,KM,KMSTOP
* DIMENSION COR(MAXR),R(MAXR)
*
* SET UP PARAMETERS FOR NEARN SEARCH
* ISTAY = 1
* ILOOK = 2
* NPART = 0
* N = 0
* DO 100 K=1,KM
* DO 100 J=1,JM
* DO 100 I=1,IM
* IFND=0
* CALL NEARN(A,I,J,K,ISTAY,ILOOK,IFND,KFND)
* IF (IFND.EQ.1 .AND. K.LT.KM) THEN
* WE WANT TO LEAVE A() THE WAY THAT IT WAS
* A(I,J,K) = 1
* LBLA(I,J,K) = 1

```

```

POR18420
POR18430
POR18440
POR18450
POR18460
POR18470
POR18480
POR18490
POR18500
POR18510
POR18520
POR18530
POR18540
POR18550
POR18560
POR18570
POR18580
POR18590
POR18600
POR18610
POR18620
POR18630
POR18640
POR18650
POR18660
POR18670
POR18680
POR18690
POR18700
POR18710
POR18720
POR18730
POR18740
POR18750
POR18760
POR18770
POR18780
POR18790
POR18800
POR18810
POR18820
POR18830
POR18840
POR18850
POR18860
POR18870
POR18880
POR18890
POR18900

```

```

                NPART = NPART + 1
            ELSE
                LBLA(I,J,K) = 0
            ENDIF
    100    CONTINUE
*
    ISHOW = 0
    CALL DISPLA(LBLA,ISHOW,MODEL)
    WRITE (6,*) 'SURFACE CORRELATIONS:'
    WRITE (7,*) 'SURFACE CORRELATIONS:'
    WRITE (6,*) NPART
    ROOT2 = SQRT(2.0)
    WRITE (7,*) NPART
    DO 500 IR=1,IM-3
*   CALCULATE C(R) FOR EACH R:
        N = N+1
        R(N) = IR
        COR(N) = 0.0
        DO 200 K=1,KM
            DO 200 J=1,JM
                DO 200 I=1,IM
*   AVERAGE C(R) OVER THE SIX NEAREST NEIGHBOR CUBIC DIRECTIONS
                    NLBLA = LBLA (I,J,K)
                    IF (I+IR.LT.IM)
1                       COR(N) = COR(N) + NLBLA * LBLA(I+IR,J,K)
                    IF (I-IR.GT.1)
1                       COR(N) = COR(N) + NLBLA * LBLA(I-IR,J,K)
                    IF (J+IR.LT.JM)
1                       COR(N) = COR(N) + NLBLA * LBLA(I,J+IR,K)
                    IF (J-IR.GT.1)
1                       COR(N) = COR(N) + NLBLA * LBLA(I,J-IR,K)
                    IF (K+IR.LT.KM)
1                       COR(N) = COR(N) + NLBLA * LBLA(I,J,K+IR)
*   K=1 PLANE ALWAYS CONTAINS ELECTROLYTE
                    IF (K-IR.GT.1)
1                       COR(N) = COR(N) + NLBLA * LBLA(I,J,K-IR)
200    CONTINUE
        COR(N) = COR(N) / (NPART*6.)
        WRITE (6,*) R(N),COR(N)
        WRITE (7,*) R(N),COR(N)
*   ----- NOW CALCULATE 2ND NEAREST NEIGHBOR PAIR CORRELATIONS
*   CALCULATE C(R) FOR EACH R:
        N = N+1
        R(N) = IR*ROOT2
        COR(N) = 0.0
        DO 300 K=1,KM
            DO 300 J=1,JM
                DO 300 I=1,IM
                    NLBLA = LBLA(I,J,K)
*   AVERAGE C(R) OVER THE 12 2ND NEAREST NEIGHBOR CUBIC DIRECTIONS

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POR18910
POR18920
POR18930
POR18940
POR18950
POR18960
POR18970
POR18980
POR18990
POR19000
POR19010
POR19020
POR19030
POR19040
POR19050
POR19060
POR19070
POR19080
POR19090
POR19100
POR19110
POR19120
POR19130
POR19140
POR19150
POR19160
POR19170
POR19180
POR19190
POR19200
POR19210
POR19220
POR19230
POR19240
POR19250
POR19260
POR19270
POR19280
POR19290
POR19300
POR19310
POR19320
POR19330
POR19340
POR19350
POR19360
POR19370
POR19380
POR19390
POR19400

```

```

      IF (J+IR.LT.JM) THEN
      IF (I+IR.LT.IM)
1      COR(N) = COR(N) + NLBLA * LBLA(I+IR,J+IR,K)
      IF (I-IR.GT.0)
1      COR(N) = COR(N) + NLBLA * LBLA(I-IR,J+IR,K)
      IF (K+IR.LT.KM)
1      COR(N) = COR(N) + NLBLA * LBLA(I,J+IR,K+IR)
      IF (K-IR.GT.0)
1      COR(N) = COR(N) + NLBLA * LBLA(I,J+IR,K-IR)
      ENDIF
      IF (J-IR.GT.0) THEN
      IF (I+IR.LT.IM)
1      COR(N) = COR(N) + NLBLA * LBLA(I+IR,J-IR,K)
      IF (I-IR.GT.0)
1      COR(N) = COR(N) + NLBLA * LBLA(I-IR,J-IR,K)
      IF (K+IR.LT.KM)
1      COR(N) = COR(N) + NLBLA * LBLA(I,J-IR,K+IR)
      IF (K-IR.GT.0)
1      COR(N) = COR(N) + NLBLA * LBLA(I,J-IR,K-IR)
      ENDIF
      IF (K-IR.GT.1) THEN
      IF (I+IR.LT.IM)
1      COR(N) = COR(N) + NLBLA * LBLA(I+IR,J,K-IR)
      IF (I-IR.GT.0)
1      COR(N) = COR(N) + NLBLA * LBLA(I-IR,J,K-IR)
      ENDIF
      IF (K+IR.LT.KM) THEN
      IF (I+IR.LT.IM)
1      COR(N) = COR(N) + NLBLA * LBLA(I+IR,J,K+IR)
      IF (I-IR.GT.0)
1      COR(N) = COR(N) + NLBLA * LBLA(I-IR,J,K+IR)
      ENDIF
300 CONTINUE
      COR(N) = COR(N) / (NPART*12.)
      WRITE (6,*) R(N),COR(N)
      WRITE (7,*) R(N),COR(N)
500 CONTINUE
      RETURN
      END
-----
* SUBROUTINE TO CALCULATE LATTICE CORRELATIONS
* TO TRY AND QUANTIFY THE PROPERTIES OF
* THE GEOMETRICAL CHARACTERISTICS OF EACH POROUS ELECTRODE
* -- DENSITY-DENSITY CORRELATION CALCULATION ---
* WRITTEN ON 6/16/85 IN VS FORTRAN77
* SUBROUTINE CORLAT (A, PORVOL, VOLNOR, ILOOK, COR, IMAX)
*
      PARAMETER (MAXR=25)
      INTEGER A(IM, JM, KM)

```

```

POR19410
POR19420
POR19430
POR19440
POR19450
POR19460
POR19470
POR19480
POR19490
POR19500
POR19510
POR19520
POR19530
POR19540
POR19550
POR19560
POR19570
POR19580
POR19590
POR19600
POR19610
POR19620
POR19630
POR19640
POR19650
POR19660
POR19670
POR19680
POR19690
POR19700
POR19710
POR19720
POR19730
POR19740
POR19750
POR19760
POR19770
POR19780
POR19790
POR19800
POR19810
POR19820
POR19830
POR19840
POR19850
POR19860
POR19870
POR19880
POR19890

```

```

COMMON /LATTICE/ IM,JM,KM,KMSTOP
DIMENSION ICOR(MAXR),COR(MAXR)
INTEGER*4 IR
*
  IMAX = MAXR
  IMIN = MIN (IM,JM,KM)
  IF (MAXR.GT.IMIN) THEN
    WRITE (*,*) 'ERROR - SPEC. MAX CORRELATION LENGTH TOO HIGH'
    WRITE (*,*) 'SETTING MAXR = ',IMIN
    IMAX = IMIN
  ENDIF
*
* FIRST VARY R TO GET THE CORRELATION FUNCTION, ICOR(IR)
*
DO 500 IR=1,IMAX
  N = 0
  ICOR(IR) = 0
*
* FOR EACH R - SUM OVER THE WHOLE LATTICE.
  DO 100 K=1,KM
  DO 100 J=1,JM
  DO 100 I=1,IM
*
    IAT = A(I,J,K)
    IF (IAT.EQ.ILOOK) THEN
      N = N + 1
*****
      UP
      IQ = K + IR
      IF (IQ.GT.KM) IQ = IQ - KM
      IF (IAT.EQ.A(I,J,IQ)) ICOR(IR) = ICOR(IR) + 1
*****
      DOWN
      IQ = K - IR
      IF (IQ.LT.1) IQ = IQ + KM
      IF (IAT.EQ.A(I,J,IQ)) ICOR(IR) = ICOR(IR) + 1
*****
      RIGHT
      IQ = J+IR
      IF (IQ.GT.JM) IQ = IQ - JM
      IF (IAT.EQ.A(I,IQ,K)) ICOR(IR) = ICOR(IR) + 1
*****
      LEFT
      IQ = J-IR
      IF (IQ.LT.1) IQ = IQ + JM
      IF (IAT.EQ.A(I,IQ,K)) ICOR(IR) = ICOR(IR) + 1
*****
      FRONT
      IQ = I + IR
      IF (IQ.LT.IM) IQ = IQ - IM
      IF (IAT.EQ.A(IQ,J,K)) ICOR(IR) = ICOR(IR) + 1
*****
      BACK
      IQ = I - IR
      IF (IQ.LT.1) IQ = IQ + IM

```

```

POR19900
POR19910
POR19920
POR19930
POR19940
POR19950
POR19960
POR19970
POR19980
POR19990
POR20000
POR20010
POR20020
POR20030
POR20040
POR20050
POR20060
POR20070
POR20080
POR20090
POR20100
POR20110
POR20120
POR20130
POR20140
POR20150
POR20160
POR20170
POR20180
POR20190
POR20200
POR20210
POR20220
POR20230
POR20240
POR20250
POR20260
POR20270
POR20280
POR20290
POR20300
POR20310
POR20320
POR20330
POR20340
POR20350
POR20360
POR20370
POR20380

```

```

          IF (IAT.EQ.A(IQ,J,K)) ICOR(IR) = ICOR(IR) + 1
        ENDIF
100    CONTINUE
        COR(IR) = REAL(ICOR(IR)) / (REAL(N) * 6.)
        WRITE (6,150) IR,COR(IR),ALOG10(FLOAT(IR)),ALOG10(COR(IR))
        WRITE (7,150) IR,COR(IR),ALOG10(FLOAT(IR)),ALOG10(COR(IR))
150    FORMAT (' ',14,1P,3E15.5)
500    CONTINUE
        RETURN
        END
*****
*
*       END OF PROGRAM POR_MET AND SUBROUTINES
*
*****
*
*
*       DISSOLUTION - PRECIPITATION SIMULATION
*       PROGRAM DP2
*       VERSION 2.15 FROM 01/07/86
*       TRYING TO GENERATE BROWN FRACTALS
*       THIS IMPLEMENTATION: LET TOP ROW LOSE PARTICLES
*       KEEP TRACK OF MAX Y, # OF ATTACHED PARTICLES, MIN Y
*       ALLOWS FOR CLUSTER ATTACHMENT: NO CLUSTERS (ICLUST=0), ALL CLUSTERS
*       (ICLUST=1) OR CLUSTERS LARGER THAN A CERTAIN SIZE (ICLUST>1)
*       ALLOWS FOR DISSOLUTION OF PARTICLES AS WELL
*       THE PARTICLE MOVEMENT CAN BE REGULATED BY PROB3 - PROBABILITY
*       TO TRY TO MOVE A PARTICLE
*       COR1 - CORRELATION MATRIX FOR # OF POINTS ON THE INTERFACE
*       COR2 - CORRELATION MATRIX FOR # OF INTERFACES
*
*****
*
*       PARAMETER (MXSPAC =050,MYSPAC =050,MXPRT = 1*MXSPAC*MYSPAC)
*       INTEGER*2 A(MXSPAC,MYSPAC),B(MXPRT,2),D(MXPRT,2)
* AS IS A TEMPORARY MATRIX TO CONTAIN ONLY SURFACE PARTICLES
*       INTEGER*2 AS(MXSPAC,MYSPAC)
*       INTEGER*2 MODE,IPALET,ICOLOR(3),ICLR,MAXR
*       INTEGER*2 DEVICE,I2ZERO,I2ONE,I2THRE,I2FOUR,IDIS,KCOUNT,JCOUNT
*       INTEGER*2 I2TWO
*       PARAMETER (I2ZERO=0,I2ONE=1,I2THRE=3,I2FOUR=4,I2TWO=2)
*       INTEGER*4 NPART,MXGEN,KGEN,BGROW(MXPRT),JENCNT(50),MAXHT(50)
*       INTEGER*4 NPSTUK(50),NPFRE(50),NSURPT(50),MXFREE(50)
*       INTEGER*2 MINY(50),ICNTR(-1:1)
*       LOGICAL*1 EMPTY,NOTSAV,IQUIT
*       REAL*8 DSEEDO,DSEED,GETTIM,GOTMIN,ZMANO,ZMAN
C DCONC IS THE CONCENTRATION PROFILE FOR DISSOLVED PARTICLES
C SCONC IS THE CONCENTRATION PROFILE FOR STUCK PARTICLES

```

```

POR20390
POR20400
POR20410
POR20420
POR20430
POR20440
POR20450
POR20460
POR20470
POR20480
POR20490
* POR20500
* POR20510
* POR20520
* POR20530
* POR20540
* POR20550
* POR20560
* POR20570
* POR20580
* POR20590
* POR20600
* POR20610
* POR20620
* POR20630
* POR20640
* POR20650
* POR20660
* POR20670
* POR20680
* POR20690
* POR20700
* POR20710
* POR20720
* POR20730
* POR20740
* POR20750
* POR20760
* POR20770
* POR20780
* POR20790
* POR20800
* POR20810
* POR20820
* POR20830
* POR20840
* POR20850
* POR20860
* POR20870
* POR20880

```

```

REAL*4 RUF(50),DCONC(10000),SCONC(10000)
REAL*4 COR1(100000),COR2(100000)
CHARACTER*8 IDNAM
CHARACTER*4 CDUM,MSG1,MSG2,MSG3,MSG4
CHARACTER*1 GRAP(-1:5)
DATA GRAP /'+',' ','i','#','?',' ','#',' ','-',' ','!'/
1,MSG1/'CONC'/,MSG2/'PART'/,
1 MSG3/'COR1'/,MSG4/'COR2'/
COMMON /SIZPRM/ NXSPAC,NYSPAC,MXNPRT,NPART,NNUC,NLAY
COMMON /PARAM/ NPARTO,PROB,PROB2,PROB3,CONC,IZ,MX1Y,ICLUST,KGEN,
1 MNIY,MXFRE,IAMBLK
*****
WRITE (*,*) 'IF U CHANGE ANYTHING, PLZ CHGE REV AND INSERT A COM'
REV = 2.15
WRITE (*,10) REV
10 FORMAT (' PROGRAM DP - REVISION ',F5.2)
*****
DUMMY=0.0
EMPTY = .TRUE.
NOTSAV = .FALSE.
IQUIT = .FALSE.
MODE = 1
IPALET = 0
ICOLOR(1) = 1
ICOLOR(2) = 0
ICOLOR(3) = 2
NXSPAC= MXSPAC
NYSPAC= MYSPAC
MXNPRT= MXPRT
MXFRE = 0
MAXR=NXSPAC/2
KCOUNT= IZZERO
JCOUNT= IZZERO
NSURF=0
NSURF1=0
IORD = -1
* BULK IS THE LOCAL 'Y' CONCENTRATION THAT ALMOST INSURES
* THAT THIS LINE IS CONNECTED TO THE BULK - USED WITH PARAM "IAMBLK"
BULK = .80
IDBULK = BULK * NXSPAC
DCONC(1) = 1.
SCONC(1) = 1.
DO 100 J=1,NYSPAC
DO 100 I=1,NXSPAC
100 A(I,J) = IZZERO
CALL ICOUNT (A)
CALL ICPROF (A,DCONC,SCONC)
20 IF (IQUIT) GOTO 3000
WRITE (*,*) 'ENTER CHOICE:'

```

```

POR20890
POR20900
POR20910
POR20920
POR20930
POR20940
POR20950
POR20960
POR20970
POR20980
POR20990
POR21000
POR21010
POR21020
POR21030
POR21040
POR21050
POR21060
POR21070
POR21080
POR21090
POR21100
POR21110
POR21120
POR21130
POR21140
POR21150
POR21160
POR21170
POR21180
POR21190
POR21200
POR21210
POR21220
POR21230
POR21240
POR21250
POR21260
POR21270
POR21280
POR21290
POR21300
POR21310
POR21320
POR21330
POR21340
POR21350
POR21360
POR21370

```

```

WRITE (*,*) '1- INITIALIZE A NEW PROBLEM'
WRITE (*,*) '2- RESUME AN OLD PROBLEM'
WRITE (*,*) '3- ---- QUIT ----'
WRITE (*,*) '4- GO!'
WRITE (*,*) '5- STORE THE CURRENT SIMULATION'
WRITE (*,*) '6- LIST THE CURRENTLY DEFINED PARAMETERS'
WRITE (*,*) '7- GRAPHICAL DISPLAY OF THE CURRENT SIMULATION'
CALL RDTERM(IOPT,DUM,CDUM,0,0,*20)
IF (EMPTY.AND.(IOPT.GT.3)) THEN
  WRITE (*,*) ' NO SIMULATION CURRENTLY DEFINED'
  GOTO 20
ENDIF
GOTO (1000,2000,3000,4000,5000,6000,7000), IOPT
GOTO 20
===== INITIALIZE A NEW PROBLEM =====
1000 WRITE (*,*) ' ENTER ID NAME FOR THIS RUN (DATAID-8 CHAR ',
1 ' ENCLOSED IN SINGLE QUOTES)'
  READ (*,*) IDNAM
  WRITE (*,*) 'ENTER DESIRED CLUSTER ATTACHMENT:',
1 ' (0 - NO CLUSTERS, 1 - ALL CLUSTERS, ETC.)'
  READ (*,*) ICLUST
  WRITE (*,*) 'ENTER DESIRED PARTICLE CONCENTRATION (<.5):'
  READ (*,*) CONC
  WRITE (*,*) 'ENTER DESIRED NUCLEI CONCENTRATION :'
  READ (*,*) CONUC
  WRITE (*,*) 'ENTER DESIRED # OF LAYERS :'
  READ (*,*) NLAY
  IF (NLAY.GT.2*NYSPEC/3) THEN
    WRITE (*,*) ' TOO MANY LAYERS OF BULK SPECIFIED'
    STOP
  ENDIF
  WRITE (*,*) 'ENTER DESIRED STICKING PROBABILITY .'
  READ (*,*) PROB
  WRITE (*,*) 'ENTER DESIRED UN-STICKING PROBABILITY :'
  READ (*,*) PROB2
  WRITE (*,*) 'ENTER DESIRED MOVING PROBABILITY :'
  READ (*,*) PROB3
  DSEEDO = 2345.000
  DSEED = DSEEDO
  NNUC=CONUC*NXSPAC
  IZ=NNUC+(NLAY-1)*NXSPAC
  NFREE= CONC*NXSPAC*(NYSPEC-NLAY-CONUC)
  NPART= NFREE+IZ
* NTOP IS THE NUMBER OF PARTICLES ON THE TOP (NON-NUCLEATING) PLANE
* THIS MAY BE KEPT CONSTANT IN SUCCEEDING GENERATIONS BY ADDING LOST
C PARTICLES, COUNTED BY 'CNTTOP'
  NTOP = CONC*NXSPAC+.5
  NPARTO = NPART
  DO 1050 J=1,NYSPEC

```

```

POR21380
POR21390
POR21400
POR21410
POR21420
POR21430
POR21440
POR21450
POR21460
POR21470
POR21480
POR21490
POR21500
POR21510
POR21520
POR21530
POR21540
POR21550
POR21560
POR21570
POR21580
POR21590
POR21600
POR21610
POR21620
POR21630
POR21640
POR21650
POR21660
POR21670
POR21680
POR21690
POR21700
POR21710
POR21720
POR21730
POR21740
POR21750
POR21760
POR21770
POR21780
POR21790
POR21800
POR21810
POR21820
POR21830
POR21840
POR21850
POR21860

```

```

DO 1050 I=1,NXSPAC
1050 A(I,J) = I2ZERO
CALL GENER (A,B,BGROW,DSEED)
* MXIY KEEPS TRACK OF THE MAXIMUM LEVEL THAT DENDRITE HAS GROWN TO
  MXIY = NLAY
  MNIY = NLAY
* IAMBLK KEEPS TRACK OF WHERE THE BULK APROXIMATELY BEGINS
  IAMBLK = NLAY
  KGENO = 1
* NSTOR IS A COUNTER FOR THE STORED DATA - AVGHT AND MAXHT
  NSTOR = 0
  EMPTY = .FALSE.
  ZMAN = 0.0
* RETURN TO MENU
  GOTO 20
*===== READ IN DATA FROM DISK =====
2000 READ (7,2005) IDNAM,DUMMY
      READ (7,2010) REV,EZMAN
      READ (7,2020) NXSPAC,NYSPAC,MXNPRT
      IF (NXSPAC.NE.MXSPAC.OR.NYSPAC.NE.MYSPAC) THEN
        WRITE (*,*) ' ERROR: STORED SIMULATION SPACE DIMENSION INCORRECT'
        WRITE (*,*) ' EXPECTING DIMENSIONS:',MXSPAC,' X ',MYSPAC
        WRITE (*,*) ' BUT STORED SIMULATION IS:',NXSPAC,' X ',NYSPAC
        MXNPRT = MXPRT
        NYSPAC = MYSPAC
        NXSPAC = MXSPAC
        GOTO 20
      ENDIF
      READ (7,2030) CONC,CONUC,NPARTO,NPART,FRAC
      READ (7,2040) DSEEDO,DSEED,PROB3
      READ (7,2050) ICLUST,PROB,PROB2,DUMMY
      READ (7,2060) KGEN,MXGEN,IZ,MXIY,MNIY,NSTOR,NLAY
      READ (7,2070)
      1 (JENCNT(11),NPSTUK(11),NPFRE(11),NSURPT(11)
      1,MAXHT(11),RUF(11),MINIY(11),MXFREE(11),11=1,NSTOR)
      READ (7,2075) MSG1
      READ (7,2078) DCONC(1)
      IPTO= DCONC(1)
      READ (7,2079) (DCONC(11),11=2,IPTO)
      READ (7,2078) SCONC(1)
      IPT = SCONC(1)
      READ (7,2079) (SCONC(11),11=2,IPT)
      READ (7,2075) MSG2
      READ (7,2080) ((B(11,JJ),JJ=1,2),BGROW(11),11=1,NPART)
C IORD IS THE STARTING POINT FOR FUTURE FILLING OF THE CORREL. MATRICES
  IORD =NSTOR-1
  ICOR = IORD*MAXR
  READ (7,2075) MSG3
  READ (7,2100) (COR1(1),1=1,ICOR)

```

```

POR21870
POR21880
POR21890
POR21900
POR21910
POR21920
POR21930
POR21940
POR21950
POR21960
POR21970
POR21980
POR21990
POR22000
POR22010
POR22020
POR22030
POR22040
POR22050
POR22060
POR22070
POR22080
POR22090
POR22100
POR22110
POR22120
POR22130
POR22140
POR22150
POR22160
POR22170
POR22180
POR22190
POR22200
POR22210
POR22220
POR22230
POR22240
POR22250
POR22260
POR22270
POR22280
POR22290
POR22300
POR22310
POR22320
POR22330
POR22340
POR22350

```

```

      READ (7,2075) MSG4
      READ (7,2100) (COR2(I), I=1, ICOR)

2100 FORMAT (1X,9F8.5)
2005 FORMAT (1X,A8,F5.2)
2010 FORMAT (1X,F6.2,F10.2)
2020 FORMAT (1X,2I5,18)
2030 FORMAT (1X,2F5.2,2I8,F5.2)
2040 FORMAT (1X,2D15.0,F5.2)
2050 FORMAT (1X,18,3F5.2)
2060 FORMAT (1X,2I10,18,2I5,17,15)
2070 FORMAT (1X,5I7,F7.3,2I7)
2075 FORMAT (1X,A4)
2078 FORMAT (1X,F8.0)
2079 FORMAT (1X,8F9.4)
2080 FORMAT (1X,2I5,19,2I5,19,2I5,19,2I5,19)
      DO 2200 J=1,NYSPAC
      DO 2200 I=1,NXSPAC
2200   A(I,J) = I2ZERO

*   NOW DECODE THE INFORMATION FROM THE STORED B MATRIX.
      DO 2300 I = 1,NPART
        IX = B(I,1)
        IY = B(I,2)
        A(IX,IY) = -I2ONE
*   PARTICLE STUCK??
        IF (BGROW(I).GT.0) A(IX,IY) = I2ONE
2300 CONTINUE
*   FIRST APPROXIMATION; WILL GET SET EXACTLY NEXT GENERATION
      IAMBK = MNIY
      NNUC = CONUC*NXSPAC
C     NTOP = CONC * NXSPAC + .5
      KGEN0 = KGEN + 1
      EMPTY = .FALSE.
      GOTO 20

*===== QUIT =====
3000 IF (NOTSAV) THEN
      WRITE (*,*) 'NEW SIMULATION RESULTS NOT SAVED. SAVE THEM?(Y/N)'
      IF (IGTYES(5,6).EQ.1) GOTO 5000
      ENDIF
      STOP
***** START SIMULATION *****
4000 NOTSAV = .TRUE.
C IF ONE WANTS TO KEEP THE TOP ROW AT CONSTANT CONC THEN:
C   CALL ICNTTO (A,B,BGROW,NTOP,DSEED)
C IF ONE WANTS TO KEEP THE BOTTOM ROW AT CONSTANT CONC THEN:
C   CALL ICNTBO (A,B,BGROW,NTOP,DSEED)
C   KCOUNT IS A NEIGHBOUR COUNTER   JCOUNT - SECOND NEIGHB
      CALL IUPDAT (A,B,BGROW,DSEED,D,KCOUNT,JCOUNT)

```

```

POR22360
POR22370
POR22380
POR22390
POR22400
POR22410
POR22420
POR22430
POR22440
POR22450
POR22460
POR22470
POR22480
POR22490
POR22500
POR22510
POR22520
POR22530
POR22540
POR22550
POR22560
POR22570
POR22580
POR22590
POR22600
POR22610
POR22620
POR22630
POR22640
POR22650
POR22660
POR22670
POR22680
POR22690
POR22700
POR22710
POR22720
POR22730
POR22740
POR22750
POR22760
POR22770
POR22780
POR22790
POR22800
POR22810
POR22820
POR22830
POR22840

```

```

CALL IDISP (A,AS,B,BGROW,DEVICE,REV,OLDGEN,ZMANO,CONUC
1,COR1,COR2,IORD,NSURF1,1DNAM,ZMAN,DCONC,SCONC,IPTO,MAXR)
C
C
WRITE (*,*) 'CURRENT VALUES: MXGEN=',MXGEN,' STOP FRACTION=',FRAC
WRITE (*,*) 'ENTER MAXIMUM # OF GENERATIONS      : '
READ (*,*) MXGEN
WRITE (*,*) 'ENTER MAXIMUM ALLOWED LOST FRACTION: '
READ (*,*) FRAC
*
*
* INITIALIZE INTERRUPT FLAG TO ZERO:
  INIT = IYBIT (-1)
C IORD IS A COUNTER FOR SEQUENTIAL WRITING OF CORREL MATRICES
C INITIALIZE DISSOLUTION FLAG
  IDIS=12ONE
* INITIALIZE TIMER VARIABLES
  OGEN = KGEN
* INITIAL TIME FOR TOTAL SIMULATION CALCULATION.
* ZMANO WILL ALWAYS BE SUBTRACTED FROM CURRENT TIME.
  ZMANO = GETTIM(GOTMIN)
  WRITE (*,*) 'ENTER DEVICE #:
1(0=NONE,1=PC,2=VM/TEK,3=VM/PRT,4=VM/SCREEN)'
  READ (*,*) DEVICE
* IBOTOM IS THE BOTTOM BELOW WHICH WE DONT ALLOW THIS TO PROGRESS
  IBOTOM = NLAY / 10
C DISPLAY IN LOGARITHMIC SCALE, E.G. IFREQ=3 ==> GENERATIONS DISPLAYED
C WILL BE 3, 6, 9, 30, 60, 90, 300, 600, ETC.
  IFREQ = 2
* SET UP TO AUTOMATICALLY QUIT AT THE END OF SIMULATION:
  IQUIT = .TRUE.
* INITIALIZE DECADE COUNTER
  IDECAD = 10 ** INT(ALOG10(FLOAT(KGENO)))
*
  DO 4050 I = NPART+1, MXNPRT
    BGROW(I)=0
4050 CONTINUE
*
  DO 4500 KGEN=KGENO, MXGEN
C IF ONE WANTS TO KEEP THE TOP ROW AT CONSTANT CONC THEN:
C CALL CNTTOP (A,B,BGROW,NTOP,DSEED)
C IF ONE WANTS TO KEEP THE BOTTOM ROW AT CONSTANT CONC THEN:
C CALL CNTBOT (A,B,BGROW,NTOP,DSEED)
  IF (NPART.LE.0) GOTO 20
  CALL UPDATE
* NOW UPDATE BGROW, IZ TO BE PROPERLY RECORDED
  DO 4200 I = 1, NPART
    IX = B(I,1)
    IY = B(I,2)
    POR22850
    POR22860
    POR22870
    POR22880
    POR22890
    POR22900
    POR22910
    POR22920
    POR22930
    POR22940
    POR22950
    POR22960
    POR22970
    POR22980
    POR22990
    POR23000
    POR23010
    POR23020
    POR23030
    POR23040
    POR23050
    POR23060
    POR23070
    POR23080
    POR23090
    POR23100
    POR23110
    POR23120
    POR23130
    POR23140
    POR23150
    POR23160
    POR23170
    POR23180
    POR23190
    POR23200
    POR23210
    POR23220
    POR23230
    POR23240
    POR23250
    POR23260
    POR23270
    POR23280
    POR23290
    POR23300
    POR23310
    POR23320
    POR23330

```

```

      IF (A(IX,IY).GT.I2ZERO) THEN
      IF (BGROW(I).EQ.0) THEN
      IZ=IZ+1
      BGROW(I) = KGEN
      ENDIF
      ELSE IF (A(IX,IY).LT.I2ZERO) THEN
* PARTICLE IS FREE - MAKE SURE BGROW IS 0 AND MXFRE IS CORRECT
      IF (BGROW(I) .GT. 0) THEN
      BGROW(I) = 0
      IZ=IZ-1
      ENDIF
      IF (IY.GT.MXFRE) MXFRE = IY
      ENDIF
4200 CONTINUE
-----
* NOW FIND IAMBLK VALUE
IAMBLK = MXIY
4210 ICNTR(1) = 0
      ICNTR(-1) = I2ZERO
      ICNTR(0) = I2ZERO
* COUNT THE NUMBER OF STUCK PARTICLES PER LINE - IN ICNTR(1)
DO 4250 I1=1,NXSPAC
4250 ICNTR(A(I1,IAMBLK)) = ICNTR(A(I1,IAMBLK)) + 1
      IAMBLK = IAMBLK - 1
      IF (ICNTR(1).LT.IDBULK) GOTO 4210
* IAMBLK IDENTIFIED - BELOW THAT WILL BE CONSIDERED TO BE BULK
-----
C IF IAMBLK IS TOO LOW, EXIT
IF (IAMBLK.LE.IBOTOM) GOTO 4700
IF (KGEN.GT.IDECAD*10) IDECAD = IDECAD * 10
IF (MOD(KGEN,IFREQ*IDECAD).EQ.0 ) THEN
* RESET COUNTERS EVERY NOW AND THEN SO THAT THEY DONT OVERFLOW
IORD = IORD+1
* DISPLA ALSO CALCULATES NSURF, MNIY, MXIY, NSURF1, COR1,COR2
CALL DISPLA (NSURF)
CALL CPROF(IPTO)
NSTOR = NSTOR + 1
JENCNT(NSTOR) = KGEN
NPSTUK(NSTOR) = IZ
MINIY(NSTOR) = MNIY
NPFRE (NSTOR) = NPART - IZ
MAXHT(NSTOR) = MXIY
RUF(NSTOR) = FLOAT(NSURF) / FLOAT(NXSPAC)
NSURPT(NSTOR)=NSURF1
MXFREE(NSTOR)=MXFRE
ENDIF
* LOOP EXIT CONDITION:
*
IF (NPART.LT.NPARTO*(1-FRAC).OR.MXIY+1.GE.NYSPAC) GOTO 4700
* CHECK INTERRUPT FLAG - HAS THE USER REQUESTED A HALT?
* (THIS MAY BE DONE BY THE CP COMMAND 'ST 464 1' AT A CP ATTN INTRPPOR23840

```

```

POR23340
POR23350
POR23360
POR23370
POR23380
POR23390
POR23400
POR23410
POR23420
POR23430
POR23440
POR23450
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POR23470
POR23480
POR23490
POR23500
POR23510
POR23520
POR23530
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POR23550
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POR23570
POR23580
POR23590
POR23600
POR23610
POR23620
POR23630
POR23640
POR23650
POR23660
POR23670
POR23680
POR23690
POR23700
POR23710
POR23720
POR23730
POR23740
POR23750
POR23760
POR23770
POR23780
POR23790
POR23800
POR23810
POR23820
POR23830
POR23840

```

```

      IF (ITYBIT(0).GT.0) THEN
        IQUIT = .FALSE.
        WRITE (*,*) ' USER REQUESTED HALT IN PROGRAM EXECUTION'
*      NOW SET UP LOOP PARAMETER FOR POSSIBLE LATER CONTINUE
        KGENO = KGEN + 1
        GOTO 4700
      ENDIF
4500 CONTINUE
4700 IORD=IORD+1
      CALL DISPLA (NSURF)
      CALL CPROF(IPTO)
      IF (JENCNT(NSTOR).NE.KGEN) THEN
        NSTOR = NSTOR + 1
        JENCNT(NSTOR) = KGEN
        NPSTUK(NSTOR) = IZ
        MINIY(NSTOR) = MNIY
        NPFRE (NSTOR) = NPART - IZ
        MAXHT(NSTOR) = MXIY
        RUF(NSTOR) = FLOAT(NSURF) / FLOAT(NXSPAC)
        NSURPT(NSTOR)=NSURF1
        MXFREE(NSTOR)=MXFRE
      ENDIF
* NOW GO AND STORE AND QUIT/MENU
*===== STORE THE CURRENT SIMULATION PARAMETERS =====
5000 WRITE (8,5005) IDNAM,DUMMY
      EZMAN = ZMAN / 60.DO
      WRITE (8,5010) REV,EZMAN
      WRITE (8,5020) MXSPAC,MYSPAC,MXNPRT
      WRITE (8,5030) CONC,CONUC,NPARTO,NPART,FRAC
      WRITE (8,5040) DSEEDO,DSEED,PROB3
      WRITE (8,5050) ICLUST,PROB,PROB2,DUMMY
      WRITE (8,5060) KGEN,MXGEN,IZ,MXIY,MNIY,NSTOR,NLAY
      WRITE (8,5070)
1      (JENCNT(11),NPSTUK(11),NPFRE(11),NSURPT(11)
1,MAXHT(11),RUF(11),MINIY(11),MXFREE(11),11=1,NSTOR)
      WRITE (8,5075) MSG1
      WRITE(8,5078) DCONC(1)
      IPT = DCONC(1)
      WRITE(8,5079) (DCONC(11),11=2,IPT)
      WRITE(8,5078) SCONC(1)
      IPT = SCONC(1)
      WRITE(8,5079) (SCONC(11),11=2,IPT)
      WRITE(8,5075) MSG2
      WRITE (8,5080) ((B(11,JJ),JJ=1,2),BGROW(11),11=1,NPART)
      ICOR =(IORD+1) * MAXR
      WRITE(8,5075) MSG3
      WRITE(8,5100) (COR1(1),1=1,ICOR)
      WRITE(8,5075) MSG4
      WRITE(8,5100) (COR2(1),1=1,ICOR)

```

```

POR23850
POR23860
POR23870
POR23880
POR23890
POR23900
POR23910
POR23920
POR23930
POR23940
POR23950
POR23960
POR23970
POR23980
POR23990
POR24000
POR24010
POR24020
POR24030
POR24040
POR24050
POR24060
POR24070
POR24080
POR24090
POR24100
POR24110
POR24120
POR24130
POR24140
POR24150
POR24160
POR24170
POR24180
POR24190
POR24200
POR24210
POR24220
POR24230
POR24240
POR24250
POR24260
POR24270
POR24280
POR24290
POR24300
POR24310
POR24320
POR24330

```

```

5100 FORMAT (' ',9F8.5)
5005 FORMAT (' ',A8,F5.2)
5010 FORMAT (' ',F6.2,F10.2)
5020 FORMAT (' ',2I5,I8)
5030 FORMAT (' ',2F5.2,2I8,F5.2)
5040 FORMAT (' ',F13.0,'D0',F13.0,'D0',F5.2)
5050 FORMAT (' ',I8,3F5.2)
5060 FORMAT (' ',2I10,I8,2I5,I7,I5)
5070 FORMAT (' ',5I7,F7.3,2I7)
5075 FORMAT (' ',A4)
5078 FORMAT (1X,F8.0)
5079 FORMAT (1X,8F9.4)
5080 FORMAT (' ',2I5,I9,2I5,I9,2I5,I9,2I5,I9)

```

```

NOTSAV = .FALSE.

```

```

GOTO 20

```

```

*===== LIST THE CURRENTLY DEFINED PARAMETERS =====

```

```

6000 WRITE (*,*) 'FEATURE NOT IMPLEMENTED YET'

```

```

GOTO 20

```

```

*===== GRAPHICAL DISPLAY OF SIMULATION =====

```

```

7000 WRITE (*,*) 'ENTER DEVICE #:(1=PC,2=VM/TEK,3=VM/PRT,4=VM/SCREEN)'

```

```

READ (*,*) DEVICE

```

```

* INITIALIZE TIMER VARIABLES

```

```

OGEN = KGEN

```

```

OTIM = GETTIM (GOTMIN)

```

```

CALL DISPLA (NSURF)

```

```

GOTO 20

```

```

*=====

```

```

END

```

```

SUBROUTINE GENER (A,B,BGROW,DSEED)

```

```

INTEGER*2 I2ZERO,I2ONE,I2IS

```

```

PARAMETER (I2ZERO=0,I2ONE=1)

```

```

C POPULATE A USING NPART PARTICLES AND INITIALIZE

```

```

C B TO PROPER VALUES

```

```

COMMON /SIZPRM/ NXSPAC,NYSPAC,MXNPRT,NPART,NNUC,NLAY

```

```

COMMON /PARAM/ NPARTO,PROB,PROB2,PROB3,CONC,I2,MXIY,I2CLUST,KGEN,

```

```

1 MNLY,MXFRE,IAMBLK

```

```

INTEGER*4 NXSPAC,NYSPAC,MXNPRT,NPART,BGROW(MXNPRT)

```

```

INTEGER*2 A(NXSPAC,NYSPAC),B(MXNPRT,2)

```

```

REAL*8 DSEED

```

```

C SET UP THE FIXED PARTICLES

```

```

C DEEP DOWN

```

```

NSTUK=0

```

```

DO 50 JJ=1,NLAY-1

```

```

DO 50 II=1,NXSPAC

```

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POR24340

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POR24350

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POR24360

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POR24370

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POR24380

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POR24390

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POR24400

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POR24410

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POR24420

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POR24430

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POR24440

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POR24450

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POR24460

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POR24470

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POR24480

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POR24490

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POR24500

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POR24510

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POR24520

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POR24530

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POR24540

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POR24550

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POR24560

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POR24570

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POR24580

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POR24590

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POR24600

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POR24610

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POR24620

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POR24630

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POR24640

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POR24650

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POR24660

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POR24670

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POR24680

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POR24690

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POR24700

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POR24710

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POR24720

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POR24730

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POR24740

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POR24750

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POR24760

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POR24770

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POR24780

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POR24790

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POR24800

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POR24810

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POR24820

```

```

    NSTUK=NSTUK+1
    B(NSTUK,1)=11
    B(NSTUK,2)=JJ
    BGROW(NSTUK)=1
50  A(11,JJ)=12ONE
    CONTINUE
    IF (NNUC.NE.NXSPAC) THEN
400  DO 500 I=NSTUK+1,IZ
        IX=(NXSPAC*GGUBFS(DSEED)+1.)
        IY=1
        IF (A(IX,IY).EQ.12ZERO) THEN
            A(IX,IY)=12ONE
            B(1,1)=IX
            B(1,2)=IY
            BGROW(1) = 1
        ELSE
            GOTO 400
        ENDIF
500  CONTINUE
        GOTO 600
    ELSE
        DO 550 I=1,NXSPAC
            NSTUK=NSTUK+1
            B(NSTUK,1)=I
            B(NSTUK,2)=NLAY
            BGROW(NSTUK)=1
            A(I,NLAY)=12ONE
550  CONTINUE
        ENDIF
* NOW TO SET UP THE FREE PARTICLES:
    IF (NPART.EQ.1Z) RETURN
600  DO 1000 I=1Z+1,NPART
800  IX=(NXSPAC*GGUBFS(DSEED)+1.)
        IY=(NYSPEC*GGUBFS(DSEED)+1.)
        IF (A(IX,IY).EQ.12ZERO) THEN
            A(IX,IY)=-12ONE
            B(1,1)=IX
            B(1,2)=IY
            BGROW(1) = 0
        ELSE
            GOTO 800
        ENDIF
1000 CONTINUE
    RETURN
    END

```

SUBROUTINE ISHUFFL (DSEED,B,BGROW)

```

POR24830
POR24840
POR24850
POR24860
POR24870
POR24880
POR24890
POR24900
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POR24920
POR24930
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POR24980
POR24990
POR25000
POR25010
POR25020
POR25030
POR25040
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POR25070
POR25080
POR25090
POR25100
POR25110
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POR25130
POR25140
POR25150
POR25160
POR25170
POR25180
POR25190
POR25200
POR25210
POR25220
POR25230
POR25240
POR25250
POR25260
POR25270
POR25280
POR25290
POR25300
POR25310

```

```

      INTEGER*4 NPART
      COMMON /SIZPRM/  NXSPAC,NYSPAC,MXNPRT,NPART,NNUC,NLAY
      COMMON /PARAM/  NPARTO,PROB,PROB2,PROB3,CONC,IZ,MXIY,ICLUS,KGEN,
1  MN1Y,MXFRE,IAMBLK
      REAL*8 DSEED
      INTEGER*2 B(MXNPRT,2),ITEMP
      INTEGER*4 BGROW(MXNPRT)
      RETURN
*
      ENTRY SHUFLE
      M=NPART
      KM1 = NPART-1
C
      DO 10 I=1,KM1
      NOW RANDOMLY PERMUTE
      DO 10 I=1,KM1
      J = 1+GGUBFS(DSEED)*M
      ITEMP = B(M,1)
      B(M,1) = B(J,1)
      B(J,1) = ITEMP
      ITEMP = B(M,2)
      B(M,2) = B(J,2)
      B(J,2) = ITEMP
      ITEMP = BGROW(M)
      BGROW(M) = BGROW(J)
      BGROW(J) = ITEMP
      M = M-1
10 CONTINUE
      RETURN
      END

      SUBROUTINE IUPDAT (A,B,BGROW,DSEED,D,KCOUNT,JCOUNT)
      INTEGER*2 IZERO,I2ONE,IDI3,KCOUNT,I2FOUR,I2TWO,I2THRE,JCOUNT
      PARAMETER (IZERO=0,I2ONE=1,I2FOUR=4,I2TWO=2,I2THRE=3)
C UPDATES POSITION OF THE PARTICLES; IZ IS A STUCK PARTICLE COUNTER
C MXIY RECORDS THE MAXIMUM HEIGHT OF DENDRITE, MN1Y - THE 'FLOOR LEVEL'
      COMMON /SIZPRM/  NXSPAC,NYSPAC,MXNPRT,NPART,NNUC,NLAY
      COMMON /PARAM/  NPARTO,PROB,PROB2,PROB3,CONC,IZ,MXIY,ICLUS,KGEN,
1  MN1Y,MXFRE,IAMBLK
      INTEGER*4 NXSPAC,NYSPAC,MXNPRT,NPART,BGROW(MXNPRT)
      INTEGER*2 A(NXSPAC,NYSPAC),B(MXNPRT,2),D(MXNPRT,2)
      INTEGER*2 ILIST(4),JLIST(4)
      REAL*8 DSEED
      CALL ISHUFLE (DSEED,B,BGROW)
      CALL IMOVE (A,B,BGROW,I,IX,IY,DSEED,ITRY)
      CALL INCLUS (A,D)
      CALL INEIGH (A)
      RETURN
*
      ENTRY UPDATE

```

```

POR25320
POR25330
POR25340
POR25350
POR25360
POR25370
POR25380
POR25390
POR25400
POR25410
POR25420
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POR25450
POR25460
POR25470
POR25480
POR25490
POR25500
POR25510
POR25520
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POR25600
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POR25640
POR25650
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POR25670
POR25680
POR25690
POR25700
POR25710
POR25720
POR25730
POR25740
POR25750
POR25760
POR25770
POR25780
POR25790
POR25800

```

```

CALL SHUFFLE
* THE VALUE OF NPART CHANGES AS PARTICLES LEAVE AND ENTER THE BOX
DO 1000 I=1,MXNPRT
* EVEN THOUGH NPART MAY HAVE DECREASED WE STILL WANT TO GET ALL THE
* PARTICLES CHECKED IN THIS GENERATION
5 IF (I.GT.NPART) RETURN
IX=B(I,1)
IY=B(I,2)
C IS THE PARTICLE STUCK??
***** IF SO, BEGIN ATTEMPT TO DISLodge STUCK PARTICLE
C UNLESS IT IS ON THE BOTTOM PLANE
IF (IY.EQ.1) GOTO 1000
IF (A(IX,IY).GT.I2ZERO) THEN
* IF SURROUNDED IT STAYS STUCK - AND GOTO NEXT PARTICLE
CALL COUNT (IX,IY,KCOUNT,JKOUNT,1)
IF (KCOUNT.EQ.I2FOUR) GOTO 1000
* IF THE PARTICLE IS ISOLATED, IT WILL DISSOLVE
IF (KCOUNT.EQ.I2ZERO) THEN
A(IX,IY)=-I2ONE
* TRY TO MOVE IT
GOTO 15
ENDIF
* PARTICLE IS STUCK AND ON THE SURFACE
* DOES IT DISSOLVE?
R=GCUBFS(DSEED)
* PROB2 IS THE DISSOLUTION PROBABILITY
* IF IT DOESNT DISSOLVE, THEN FORGET THIS PARTICLE.
IF (R.GE.PROB2) GOTO 1000
* IF R IS LT PROB2 THEN THE PARTICLE DISSOLVES
A(IX,IY)=-I2ONE
* NEXT QUESTION - DID DISSOLVED PARTICLE DISCONNECT A CLUSTER?
C IDIS IS A DISSOLUTION (1)/PRECIPITATION (-1) FLAG
IDIS = I2ONE
C IF IT IS A TIP OF DENDRITE, JUST TRY TO MOVE IT
IF (KCOUNT.EQ.I2ONE) GOTO 15
*****
CALL NEIGH(IX,IY,I1LIST,JLIST)
* I1LIST AND JLIST CONTAIN NEIGHBOR'S COORDINATES
*=====BEGIN AN ATTEMPT TO SEE WHAT ELSE DISSOLVES
*=====WITH CURRENT PARTICLE
IF (KCOUNT.EQ.I2TWO) THEN
DO 10 IGO=1,4
* WE KNOW THERE ARE TWO NEIGHBORS - FIND THEM!
IF (I1LIST(IGO).EQ.I2ZERO) GOTO 10
* FOUND ONE; HOW MANY NEIGHBORS DOES THIS NEIGHBOR HAVE?
IP = I1LIST(IGO)
IQ = JLIST(IGO)
CALL COUNT (IP,IQ,KCOUNT,JKOUNT,1)
IF (KCOUNT.EQ.I2ZERO) THEN

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POR25810
POR25820
POR25830
POR25840
POR25850
POR25860
POR25870
POR25880
POR25890
POR25900
POR25910
POR25920
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POR25940
POR25950
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POR25970
POR25980
POR25990
POR26000
POR26010
POR26020
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POR26080
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POR26100
POR26110
POR26120
POR26130
POR26140
POR26150
POR26160
POR26170
POR26180
POR26190
POR26200
POR26210
POR26220
POR26230
POR26240
POR26250
POR26260
POR26270
POR26280
POR26290

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*           A(IP,IQ) = -120NE
*           DONT LOOK FOR OTHER NEIGHBORS - THEY GOTTA BE STUCK
*           GOTO 15
*           ELSE
*           CALL CLUSTR (IP,IQ,IDIS)
*           ENDF
10  CONTINUE
*           ELSE
*           THERE ARE THREE NEIGHBORS FOR THE DISSOLVED PARTICLE
*           DO 12 IGO=1,4
*           WE KNOW THERE ARE THREE NEIGHBORS - FIND THEM!
*           IF (ILIST(IGO) .EQ. 12ZERO) GOTO 12
*           HOW MANY NEIGHBORS DOES THIS NEIGHBOR HAVE?
*           IP = ILIST(IGO)
*           IQ = JLIST(IGO)
*           CALL COUNT (IP,IQ,KCOUNT,JCOUNT,1)
*           IF (KCOUNT.EQ.12ZERO) THEN
*           A(IP,IQ) = -120NE
*           ELSE
*           IF (KCOUNT.LT.12THRE) CALL CLUSTR (IP,IQ,IDIS)
*           ENDF
12  CONTINUE
*===== END OF ATTEMPT TO SEE HOW MUCH DISSOLVES WITH CURR PARTICLE
*           ENDF
*##### END OF ATTEMPT TO DISLodge STUCK PARTICLE
*           ENDF
*+++++++IS THE PARTICLE FREE? THEN TRY TO MOVE IT & THEN PRECIPITATE
15  IF (A(IX,IY).LT.120NE) THEN
*           IF (GGUBFS(DSEED).LE.PROB3) CALL MOVE
C IF A PARTICLE IS LOST THEN ANOTHER ONE TAKES ITS PLACE AND IS CHECKED
*           IF (ITRY.LT.0) GOTO 5
*           AFTER EACH ADJACENT PARTICLE IS CHECKED, FIND OUT IF IT'S STUCK YET
*           PERIODIC BOUNDARY CONDITION ON THE X-AXIS
*           RIGHT HAND SIDE
*           IQ=IX+1
*           IF (IX.EQ.NXSPAC) IQ=1
*           IF (A(IQ,IY).GT.12ZERO) GOTO 950
*           LEFT HAND SIDE
*           IQ=IX-1
*           IF (IQ.EQ.0) IQ=NXSPAC
*           IF (A(IQ,IY).GT.12ZERO) GOTO 950
*
*           TOWARDS TOP - NON-NUCLEATING SIDE
*           IF (IY.LT.NYSPAC) THEN
*           IF (A(IX,IY+1).GT.12ZERO) GOTO 950
*           ENDF
*           BOTTOM - TOWARDS NUCLEATING PLANE
*           IF (IY.GT.1) THEN
*           IF (A(IX,IY-1).GT.12ZERO) GOTO 950
C IF IT NEIGHBORS THE NUCLEATING PLANE, TRY TO STICK IT

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POR26300
POR26310
POR26320
POR26330
POR26340
POR26350
POR26360
POR26370
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POR26390
POR26400
POR26410
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POR26430
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POR26470
POR26480
POR26490
POR26500
POR26510
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POR26580
POR26590
POR26600
POR26610
POR26620
POR26630
POR26640
POR26650
POR26660
POR26670
POR26680
POR26690
POR26700
POR26710
POR26720
POR26730
POR26740
POR26750
POR26760
POR26770
POR26780
POR26790

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ELSE
  GOTO 950
ENDIF
C NO STUCK NEIGHBORS, SO GO TO THE NEXT PARTICLE
GOTO 1000
***** ATTEMPT TO STICK
950 R=GGUBFS(DSEED)
IF (R.LT.PROB) THEN
*   PARTICLE HAS STUCK
   A(IX,IY)=I2ONE
C ADJUSTING IDIS FOR PRECIPITATION
  IF (ICLUST.GT.0) THEN
    IDIS = -I2ONE
    CALL CLUSTR (IX,IY,IDIS)
  ENDIF
ENDIF
C A 'SHAPE CHECK' WILL BE INTRODUCED HERE
C   IF (A(IX,IY).GT.I2ZERO) CALL SHCHEK ( )
C
*+++++++ END OF FREE PARTICLE MOVE AND PRECIPITATE
ENDIF
1000 CONTINUE
RETURN
END

SUBROUTINE IMOVE(A,B,BGROW,I,IX,IY,DSEED,ITRY)
INTEGER*2 I2ZERO,I2ONE,IDIS
PARAMETER (I2ZERO=0,I2ONE=1)
COMMON /SIZPRM/ NXSPAC,NYSPAC,MXNPRT,NPART,NNUC,NLAY
COMMON /PARAM/ NPARTO,PROB,PROB2,PROB3,CONC,IZ,MXIY,ICLUST,KGEN,
1 MNIY,MXFRE,IAMBLK
INTEGER*4 NXSPAC,NYSPAC,MXNPRT,NPART,BGROW(MXNPRT)
INTEGER*2 A(NXSPAC,NYSPAC),B(MXNPRT,2)
REAL*8 DSEED
RETURN
*
ENTRY MOVE
ITRY = 0
IX=B(1,1)
IY=B(1,2)
C CHOOSE A DIRECTION
IDIR=(4*GGUBFS(DSEED)+1.)
GOTO (1000,4000,2000,3000) IDIR
* PERIODIC BOUNDARY CONDITIONS ON X AXIS
1000 IQ=IX+1
IF (IQ.GT.NXSPAC) IQ=1
IF (A(IQ,IY).EQ.I2ZERO) THEN
POR26800
POR26810
POR26820
POR26830
POR26840
POR26850
POR26860
POR26870
POR26880
POR26890
POR26900
POR26910
POR26920
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POR27080
POR27090
POR27100
POR27110
POR27120
POR27130
POR27140
POR27150
POR27160
POR27170
POR27180
POR27190
POR27200
POR27210
POR27220
POR27230
POR27240
POR27250
POR27260
POR27270
POR27280

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```

      A(IQ,IY)=-I2ONE
C EMPTY THE PREVIOUS POSITION
      A(IX,IY)=I2ZERO
C CHANGE COORD OF THE I PARTICLE
      B(1,1)=IQ
      IX=IQ
      ENDIF
      RETURN
2000 IQ=IX-1
      IF (IQ.LT.1) IQ=NXSPAC
      IF (A(IQ,IY).EQ.I2ZERO) THEN
      A(IQ,IY)=-I2ONE
C EMPTY THE PREVIOUS POSITION
      A(IX,IY)=I2ZERO
C CHANGE COORD OF THE I PARTICLE
      B(1,1)=IQ
      IX=IQ
      ENDIF
      RETURN
C Y-AXIS MOVEMENT
3000 IF (IY+1.LE.NYSPAC) THEN
      IF (A(IX,IY+1).EQ.I2ZERO) THEN
      A(IX,IY+1)=-I2ONE
C EMRTY THE PREVIOUS POSITION
      A(IX,IY)=I2ZERO
C CHANGE COORD OF THE I PARTICLE
      B(1,2)=IY+1
      IY=IY+1
      ENDIF
      ELSE
* TOP EDGE -- LET IT MOVE OUT AND DISAPEAR!!
* BUT ONLY IF THERE IS NO IMAGINARY PARTICLE ADJOINING IT
C R=GGUBFS(DSEED)
C IF (R.GT.0) THEN
* OK, LET IT GET LOST
      A(IX,IY) = I2ZERO
* NOTE THAT THIS PARTICLE IS NOT IN THE ITH POSITION ANYMORE
* SO FILL THAT WITH THE NPART' TH PARTICLE AND DECREMENT NPART
      B(1,1) = B(NPART,1)
      B(1,2) = B(NPART,2)
      IX = B(NPART,1)
      IY = B(NPART,2)
      BGROW(1)=BGROW(NPART)
      NPART = NPART - 1
* INDICATE THAT MOVING THE ITH PARTICLE WAS NOT SUCCESSFUL (IT'S LOST)
      ITRY = - 1
C      ENDIF
      ENDIF
      RETURN

```

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POR27290
POR27300
POR27310
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POR27400
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POR27420
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POR27450
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POR27580
POR27590
POR27600
POR27610
POR27620
POR27630
POR27640
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POR27670
POR27680
POR27690
POR27700
POR27710
POR27720
POR27730
POR27740
POR27750
POR27760
POR27770

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```

4000 IF (IY-1.GE.1) THEN
      IF (A(IX,IY-1).EQ.I2ZERO) THEN
        A(IX,IY-1)=-I2ONE
C EMPTY THE PREVIOUS POSITION
        A(IX,IY)=I2ZERO
C CHANGE COORD OF THE I PARTICLE
        B(1,2)=IY-1
        IY=IY-1
      ENDIF
    ENDIF
    RETURN
  END

  SUBROUTINE INCLUS (A,D)
    INTEGER*2 I2ZERO,I2FIVE,I2ONE,IDIS
    PARAMETER (I2ZERO=0,I2ONE=1,I2FIVE=5)
    COMMON /SIZPRM/ NXSPAC,NYSPAC,MXNPRT,NPART,NNUC,NLAY
    COMMON /PARAM/ NPARTO,PROB,PROB2,PROB3,CONC,IZ,MXIY,ICLUST,KGEN,
     1 MNIY,MXFRE,IAMBLK
    INTEGER*4 NXSPAC,NYSPAC,MXNPRT,NPART,ICNTR(-1:1)
C D(NPART,2) DEPOSITORY FOR STICKING CANDIDATES
    INTEGER*2 A(NXSPAC,NYSPAC),D(NPART,2)
    RETURN

  *
  * ENTRY CLUSTER (IX,IY,IDIS)
  * INITIALIZE A SEARCH WINDOW IN LOCAL REGION OF SPACE WHERE CLUSTER IS
  * THIS WINDOW IS EXPANDED AS THE CLUSTER SEARCH GROWS
    IXMIN = IX
    IXMAX = IX
    IYMIN = IY
    IYMAX = IY
C
    JCLUST=1
    D(1,1)=IX
    D(1,2)=IY
    A(IX,IY) = I2FIVE

***** BEGIN SCAN TO FIND 5'S IN OUR SEARCH WINDOW *****
50 IFOUND=0

    DO 100 I=IXMIN,IXMAX
    DO 100 J=IYMIN,IYMAX
C CHECK WHETHER A NEWLY CONSIDERED PARTICLE IS THERE
      IF (A(I,J).NE.I2FIVE) GOTO 100
C IF YES, AND A PARTICLE NEIGHBORS IT, STICK THE NEW
      IFOUND=1
      IQ=I+1
      IF (I.EQ.NXSPAC) IQ=1
      IF (A(IQ,J).EQ.IDIS) THEN

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POR27780
POR27790
POR27800
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POR27990
POR28000
POR28010
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POR28100
POR28110
POR28120
POR28130
POR28140
POR28150
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POR28170
POR28180
POR28190
POR28200
POR28210
POR28220
POR28230
POR28240
POR28250
POR28260

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      A(IQ,J)=I2FIVE
      JCLUST=JCLUST+1
      D(JCLUST,1)=IQ
      D(JCLUST,2)=J
      IXMAX = MAX(IQ, IXMAX)
      IF (IQ.EQ.1) THEN
        IXMAX=NXSPAC
        IXMIN=1
      ENDIF
    ENDIF
  * LEFT HAND SIDE
    IQ=I-1
    IF (IQ.EQ.0) IQ=NXSPAC
    IF (A(IQ,J).EQ.IDIS) THEN
      A(IQ,J)=I2FIVE
      JCLUST=JCLUST+1
      D(JCLUST,1)=IQ
      D(JCLUST,2)=J
      IXMIN = MIN(IQ, IXMIN)
      IF (IQ.EQ.NXSPAC) THEN
        IXMAX=NXSPAC
        IXMIN=1
      ENDIF
    ENDIF
  C Y AXIS
    IF (J.EQ.NYSPAC) GOTO 75
    IQ=J+1
    IF (A(I, IQ).EQ.IDIS) THEN
      A(I, IQ)=I2FIVE
      JCLUST=JCLUST+1
      D(JCLUST,1)=I
      D(JCLUST,2)=IQ
      IYMAX = MAX(IQ, IYMAX)
    ENDIF
  75 IF (J.EQ.1) GOTO 90
    IQ=J-1
    IF (A(I, IQ).EQ.IDIS) THEN
      A(I, IQ)=I2FIVE
      JCLUST=JCLUST+1
      D(JCLUST,1)=I
      D(JCLUST,2)=IQ
      IYMIN = MIN(IQ, IYMIN)
    ENDIF
  C NOW EMPTY THE CHECKED SPOT, WITHOUT EMPTYING A
  90 A(I,J)=-IDIS
  100 CONTINUE
  *
  ***** END WINDOW SCAN FOR 5'S *****
  C IF IN THIS SCAN A CONNECTION HAS BEEN FOUND, DON'T CHECK FURTHER

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POR28270
POR28280
POR28290
POR28300
POR28310
POR28320
POR28330
POR28340
POR28350
POR28360
POR28370
POR28380
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POR28400
POR28410
POR28420
POR28430
POR28440
POR28450
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POR28590
POR28600
POR28610
POR28620
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POR28660
POR28670
POR28680
POR28690
POR28700
POR28710
POR28720
POR28730
POR28740
POR28750
POR28760

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      IF (IYMIN.LT.IAMBLK.AND.IDIS.EQ.I2ONE) GOTO 240
C NOW CHECK WHETHER NEW PARTICLES STUCK TO THE CLUSTER
      IF (IFOUND.NE.0) GOTO 50
240   IF (IDIS.GT.I2ZERO) THEN
* WE ARE TRYING TO DISSOLVE - IS CLUSTER CONNECTED TO BULK?
      IF (IYMIN.LT.IAMBLK) THEN
* YUP - PART OF BULK - LEAVE THEM STUCK
          DO 500 I1=1,JCLUST
            A(D(I1,1),D(I1,2))=I2ONE
          CONTINUE
500   ELSE
C IF NOT CONNECTED TO THE BULK, DISSOLVE IT
          DO 550 I1=1,JCLUST
            A(D(I1,1),D(I1,2))=-I2ONE
          CONTINUE
550   ENDIF
      ELSE
* WE ARE TRYING TO STICK CLUSTER - IS IT BIG ENOUGH?
      IF (JCLUST.LT.ICLUST) THEN
C UN-STICK THE ALREADY STUCK PARTICLES INCLUDING THE INITIAL ONE
C STORED IN D(1,1)
          DO 250 I=1,JCLUST
            A(D(I,1),D(I,2))=-I2ONE
          CONTINUE
250   ELSE
          DO 255 I=1,JCLUST
            A(D(I,1),D(I,2))=I2ONE
          CONTINUE
255   ENDIF
      ENDIF
      RETURN
      END

```

SUBROUTINE IDISP (A,AS,B,BGROW,DEVICE,REV,OLDGEN,ZMANO,CONUC  
1,COR1,COR2,IORD,NSURF1,1DNAM,ZMAN,DCONC,SCONC,IPTO,MAXR)  
INTEGER\*2 I2ZERO,I2ONE,I2THRE,I2FOUR,IDIS,I2EIGT  
PARAMETER (I2ZERO=0,I2ONE=1,I2THRE=3,I2FOUR=4,I2EIGT=8)  
COMMON /SIZPRM/ NXSPAC,NYSPAC,MXNPRT,NPART,NNUC,NLAY  
COMMON /PARAM/ NPARTO,PROB,PROB2,PROB3,CONC,IZ,MXIY,ICLUST,KGEN,  
1 MN1Y,MXFRE,IAMBLK  
REAL\*4 DCONC(10000),SCONC(10000),COR1(100000),COR2(100000)  
REAL\*8 ZMAN,GETTIM,GOTMIN,ZMANO,ZMANOW,DELTAT  
INTEGER\*4 NXSPAC,NYSPAC,BGROW(MXNPRT)  
INTEGER\*2 A(NXSPAC,NYSPAC),B(MXNPRT,2),ICNTR(-1:1),KCOUNT,JCOUNT  
INTEGER\*2 AS(NXSPAC,NYSPAC)  
INTEGER\*2 MODE,IPALET,ICOLOR(3),ICLR,I1,JJ,IZERO,IFIVH  
INTEGER\*4 NPART,MXGEN,KGEN

POR28770  
POR28780  
POR28790  
POR28800  
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POR29230  
POR29240  
POR29250



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      NSURF = 0
C NSURF1 IS THE # OF SURFACE POINTS
      NSURF1 = 0
      DO 300 I = 1, NPART
          IX = B(1,1)
          IY = B(1,2)
          IF (A(IX,IY).GT.IZZERO .AND. IY.GE.MN1Y-2) THEN
C THE TRUE 'ROUGHNESS' IS THE # OF INTERFACES STUCK/REST OF THE WORLD
C (FREE PART OR EMPTY SPACE)
              NSURF=NSURF+(4-KCOUNT)
              KJCNT = KCOUNT + JCOUNT
              IF (KJCNT.LT.IZEIFGT.AND.KJCNT.GT.IZZERO) THEN
C FILLING THE "SURFACE MAP" AS(I,J)
C 1 IS ADDED TO 4-KCOUNT TO AVOID 0 FOR 4-COORDINATED SURFACE PARTICLES
                  AS(IX,IY) = IZFOUR-KCOUNT+IZONE
              ENDIF
          ENDIF
      300 CONTINUE
C CALCULATE THE CORRELATION FUNCTION
      CALL CORE
* NOW DISPLAY -
      GOTO (1000,2000,3000,4000) DEVICE
* DEVICE 0 - JUST RETURN
      RETURN
*----- PC VERSION:

      1000 CONTINUE
*      CALL CLS
          WRITE (*,*) '          ', KGEN, NPART0, NPART

*      DO 1115 KK=1, NPART
          ICLR = ICOLOR(A(B(KK,1),B(KK,2))+2)
          CALL PSET(B(KK,1),B(KK,2), ICLR)
*1115 CONTINUE
          RETURN

*----- MAINFRAME VERSION - TEK SCREEN
      2000 IONE = 1
          ICLR = 1
          ITWO = 2
          IZERO = 0
          IFIVH = 500
*      CALL INIT (IZERO,TWO)
*      II = 1
*      JJ = 2 * (NYSPEC + 5)
*      CALL PLOT (II,JJ,IZERO)
*      II = 500
*      CALL PLOT (II,JJ,ITWO)

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POR29760
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POR29900
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POR29930
POR29940
POR29950
POR29960
POR29970
POR29980
POR29990
POR30000
POR30010
POR30020
POR30030
POR30040
POR30050
POR30060
POR30070
POR30080
POR30090
POR30100
POR30110
POR30120
POR30130
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POR30170
POR30180
POR30190
POR30200
POR30210
POR30220
POR30230
POR30240
POR30250

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*      JJ = 3 *NYSPEC
*      DO 2115 J=1,NYSPEC
*      DO 2115 I=1,NXSPAC
*          II =7*I
*          JJ=5*J
*          IF (A(I,J).EQ.1) CALL PLOT(II,JJ,IONE)
*2115  CONTINUE
*      CALL TSET(IFIVH,IONE,ITWO)
*          IO = 6
*          ASSIGN 4010 TO IFORM
*          GOTO 9000
* ---- MAINFRAME -- PRINTER (NO GRAPHICS SCREEN)
3000  IO = 3
*          ASSIGN 4015 TO IFORM
*          GOTO 9000
* ---- MAINFRAME -- SCREEN ONLY
4000  IO = 6
*          ASSIGN 4010 TO IFORM
9000  AVGHT = FLOAT(IZ) / FLOAT(NXSPAC)
*          PORO=100.*(1. - AVGHT/MX1Y)
*          RUFN=FLOAT(NSURF)/FLOAT(NXSPAC)
*          WRITE (10,4105) IDNAM,EZMAN,SPEED,REV,KGEN
*          WRITE (10,4120) NXSPAC,NYSPEC,NLAY,NPARTO,ICLUST,PROB,PROB2,PROB3,
*          1CONC,CONUC
*          WRITE (10,4140) AVGHT,MX1Y,MN1Y,PORO,RUFN
*          WRITE (10,4150) NPART,IZ,NPART-IZ
*          IF (DEVICE.LT.2) RETURN
*          IF (DEVICE.EQ.4.AND.NXSPAC.GT.78) GOTO 4004
*          IF (DEVICE.EQ.3.AND.NXSPAC.GT.130) GOTO 4004
C FOR QUIK DISPLAY ON SCREEN, SHOW ONLY THE 'ROUGH' PART
*  ALWAYS SHOW ONLY UP TO THE MAXIMUM PARTICLE HEIGHT
*          MNJ=MN1Y-1
*          MXJ = MAX (MXFRE,MX1Y)
*          IF (10.EQ.3) MNJ=1
*          WRITE (10,IFORM) ((GRAP(A(I,J)),I=1,NXSPAC),J=MNJ,MXJ)
*          WRITE (10,IFORM) ((GRAP(AS(I,J)),I=1,NXSPAC),J=MNJ,MXJ)
4004  WRITE (10,4006)
4006  FORMAT ('1')
*          WRITE (10,*) '0.          ---- CONCENTRATION-----
1
1-----1.' .5
*          WRITE (10,*) '-----
1-----
1-----
1PT = DCONC(1)
IEND = 130
*          ASSIGN 4008 TO IFORM
*          IF (DEVICE.EQ.4) THEN
*              IEND = 80

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POR30260
POR30270
POR30280
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POR30390
POR30400
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POR30580
POR30590
POR30600
POR30610
POR30620
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POR30660
POR30670
POR30680
POR30690
POR30700
POR30710
POR30720
POR30730
POR30740

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      ASSIGN 4007 TO IFORM
    ENDIF
    DO 4009 I = IPT0+1, IPT
      IPOS = DCONC(I)*(IEND-1)+1
    C IF IPOS IS TOO BIG, IPOS+2 IS LARGER THAN IEND AND NEEDS ADJUSTMENT
      IF (IPOS.EQ.IEND) IPOS=IEND-2
        CPLIT = CBLANK(1:IPOS) // '*' // CBLANK(IPOS+2:IEND)
        IPOS = SCONC(I)*(IEND-1) + 1
      IF (IPOS.EQ.IEND) IPOS=IEND-2
        CPLIT = CPLIT(1:IPOS) // '+' // CPLIT(IPOS+2:IEND)
      WRITE (IO,IFORM) CPLIT
    4007 FORMAT (1X,A80)
    4008 FORMAT (1X,A130)
    4009 CONTINUE
    4010 FORMAT (' ',50A1)
    4015 FORMAT (' ',050A1)
    4105 FORMAT (' ',A8,' TIME: ',F8.2,' MIN.', ' SPEED=',F8.3,' SEC/GEN',
      1 ' VERSION#',F5.3,' GEN#',I7)
    4120 FORMAT (' ',LAT. SZE',15,' X',15,' #LYRS',15,' NPART',17,
      1 ' CLST SZE',14,' PRB (PP,DIS,MOVE)',3F5.2
      1 ' CONC',F5.3,' CONUC',F5.3)
    4140 FORMAT (' ',AVG.HT=',F7.3,' MAXHT=',15,' MNHT=',15,' PORO=',F7.3,
      1 ' ROUGH=',F9.3)
    4150 FORMAT (' ',TOTAL # ',18,' # STUCK=',18,' # FREE=',18)
    RETURN
  END

  SUBROUTINE ICOUNT (A)
    INTEGER*2 I2ZERO,I2ONE,IDIS,KCOUNT,ICNTR(-1:1),JCOUNT,I2TWO
    PARAMETER (I2ZERO=0,I2ONE=1,I2TWO=2)
    COMMON /SIZPRM/ NXSPAC,NYSPAC,MXNPRT,NPART,NNUC,NLAY
    COMMON /PARAM/ NPARTO,PROB,PROB2,PROB3,CONC,IZ,MX!Y,ICLUST,KGEN,
      1 MN!Y,MXFRE,IAMBLK
    INTEGER*4 NXSPAC,NYSPAC,MXNPRT,NPART
    INTEGER*2 A(NXSPAC,NYSPAC)
    RETURN
  C
    ENTRY COUNT (IX,IY,KCOUNT,JCOUNT,JUMP)
    KCOUNT=I2ZERO
    JCOUNT=I2ZERO
    ICNTR(1)=I2ZERO
    ICNTR(-1) = I2ZERO
    ICNTR(0) = I2ZERO
  C LOOK TO THE RIGHT
    IQ=IX+1
    IF (IX.EQ.NXSPAC) IQ=1
    ICNTR(A(IQ,IY)) = ICNTR(A(IQ,IY)) + I2ONE
  C LOOK TO THE LEFT
    IQ=IX-1

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      IF (IQ.EQ.0) IQ=NXSPAC
      ICNTR(A(IQ,IY)) = ICNTR(A(IQ,IY)) + I2ONE
C LOOK UP
      IQ=IY+1
C THE SPACE IS CONSIDERED VOID OF 'SOLUTE' OUTSIDE THE BOX
      IF (IY.EQ.NYSPAC) GOTO 100
      ICNTR(A(IX,IQ)) = ICNTR(A(IX,IQ)) + I2ONE
C LOOK DOWN
100   IQ=IY-1
C THE BOX IS INFINITELY FULL UNDER THE BOTTOM
      IF (IQ.EQ.0) THEN
          ICNTR(1) = ICNTR(1) + I2ONE
          KCOUNT = ICNTR(1)
          GOTO 150
      ENDIF
      ICNTR(A(IX,IQ)) = ICNTR(A(IX,IQ)) + I2ONE
      KCOUNT = ICNTR(1)
150   IF (JUMP.EQ.1) RETURN
C LOOKING ON THE DIAGONAL
C ICNTR SHOULD BE RESET
      ICNTR(1)=I2ZERO
      ICNTR(-1) = I2ZERO
      ICNTR(0) = I2ZERO
C NORTH-EAST
      IQX=IX+1
      IQY=IY+1
      IF (IY.EQ.NYSPAC) GOTO 200
      IF (IX.EQ.NXSPAC) IQX=1
      ICNTR (A(IQX,IQY))=ICNTR(A(IQX,IQY)) + I2ONE
C NORTH-WEST
      IQX=IX-1
      IQY=IY+1
      IF (IQX.EQ.0) IQX=NXSPAC
      ICNTR (A(IQX,IQY))=ICNTR(A(IQX,IQY)) + I2ONE
C SOUTH-EAST
200   IQX=IX+1
      IQY=IY-1
      IF (IQY.EQ.0) GOTO 300
      IF (IX.EQ.NXSPAC) IQX=1
      ICNTR (A(IQX,IQY))=ICNTR(A(IQX,IQY)) + I2ONE
C SOUTH-WEST
      IQX=IX-1
      IQY=IY-1
      IF (IX.EQ.1) IQX=NXSPAC
      ICNTR (A(IQX,IQY))=ICNTR(A(IQX,IQY)) + I2ONE
      GOTO 350
300   ICNTR(1)=ICNTR(1)+I2TWO
350   JCOUNT=ICNTR(1)
400   RETURN

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END
SUBROUTINE INEIGH (A)
INTEGER*2 I2ZERO, I2ONE, IDIS, ILIST(4), JLIST(4)
PARAMETER (I2ZERO=0, I2ONE=1)
COMMON /SIZPRM/ NXSPAC, NYSPAC, MXNPRT, NPART, NNUC, NLAY
COMMON /PARAM/ NPARTO, PROB, PROB2, PROB3, CONC, IZ, MXIY, ICLUST, KGEN,
1 MNIY, MXFRE, IAMBLK
INTEGER*4 NXSPAC, NYSPAC, MXNPRT, NPART
INTEGER*2 A(NXSPAC, NYSPAC)
RETURN
C
ENTRY NEIGH (IX, IY, ILIST, JLIST)
*
* RETURNS THE COORDINATES OF THE FOUR NEIGHBORS IF THEY EXIST
* 1- TOP 2- RIGHT 3-BOTTOM 4-LEFT
C LOOK TO THE RIGHT
IQ=IX+1
IF (IX.EQ.NXSPAC) IQ=1
IF (A(IQ, IY).EQ.I2ONE) THEN
  ILIST(2) = IQ
  JLIST(2) = IY
ELSE
  ILIST(2) = I2ZERO
ENDIF
C LOOK TO THE LEFT
IQ=IX-1
IF (IQ.EQ.0) IQ=NXSPAC
IF (A(IQ, IY).EQ.I2ONE) THEN
  ILIST(4) = IQ
  JLIST(4) = IY
ELSE
  ILIST(4) = I2ZERO
ENDIF
C LOOK UP
IQ=IY+1
C THE SPACE IS CONSIDERED VOID OF 'SOLUTE' OUTSIDE THE BOX
ILIST(1) = I2ZERO
IF (IY.LT.NYSPAC) THEN
  IF (A(IX, IQ).EQ.I2ONE) THEN
    ILIST(1) = IX
    JLIST(1) = IQ
  ENDIF
ENDIF
C LOOK DOWN
ILIST(3) = I2ZERO
IQ=IY-1
C THE BOX IS INFINITELY FULL UNDER THE BOTTOM
* BUT NEIGHBOR COORDINATES ARE NOT VALID BELOW IX = 1
* SO LEAVE ILIST = 0 FOR THE Y=0 ROW

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POR31990
POR32000
POR32010
POR32020
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POR32060
POR32070
POR32080
POR32090
POR32100
POR32110
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POR32140
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POR32210
POR32220
POR32230

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IF (IQ.EQ.0) THEN
  RETURN
ENDIF
IF (A(IX,IQ).EQ.I2ONE) THEN
  ILIST(3) = IX
  JLIST(3) = IQ
ENDIF
RETURN
END

SUBROUTINE ICPROF(A,DCONC,SCONC)
  INTEGER*2 I2ZERO,I2ONE,ICNTR(-1:1)
  PARAMETER (I2ZERO=0,I2ONE=1)
  COMMON /SIZPRM/ NXSPAC,NYSPAC,MXNPRT,NPART,NNUC,NLAY
  COMMON /PARAM/ NPARTO,PROB,PROB2,PROB3,CONC,IZ,MXIY,ICLUST,KGEN,
  1 MNIY,MXFRE,IAMBLK
  INTEGER*4 NXSPAC,NYSPAC,MXNPRT,NPART
  INTEGER*2 A(NXSPAC,NYSPAC)
  REAL*4 DCONC(10000),SCONC(10000)
  RETURN

ENTRY CPROF (IPTO)
* ON EXIT, IPTO WILL PT TO THE FIRST ENTRY OF LATEST SET
*****
* CALCULATE A CONCENTRATION PROFILE FROM MNIY TO THE TOP
* LOCAL CONCENTRATION IS A FUNCTION OF Y:
* (NUMBER OF FREE ON A ROW)
* FOR EACH Y, DCONC(Y)=-----
* (NUMBER OF FREE+NUMBER BLANK SPACES)
*****
* DCONC DATA STRUCTURE: DCONC(1) = TOTAL # OF VALID PTS IN DCONC
* DCONC(2) = TOTAL #OF PTS IN DATASET 1
* DCONC(3) ... DCONC(M)=FIRST SET'S CONC PROF
* DCONC(M+1) = TOTAL # OF PTS IN DATSET #2
* DCONC(M+2)...DCONC(N) = 2ND SET'S CONC PROF
*****
  IPT = DCONC(1) + 1
  IPTO = IPT
  DO 100 J = MNIY,MXFRE
    ICNTR(-1) = I2ZERO
    ICNTR(0) = I2ZERO
    ICNTR(1) = I2ZERO
    DO 90 I = 1,NXSPAC
      ICNTR(A(I,J)) = ICNTR(A(I,J)) + I2ONE
90 CONTINUE
    NAIR = ICNTR(0)
    NFRE = ICNTR(-1)
    NSTIK = ICNTR(1)

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      IPT = IPT + 1
      SCONC(IPT) = 1-FLOAT(NSTIK) / FLOAT(NXSPAC)
      IF (NFRE+NAIR .EQ. 0) THEN
        DCONC(IPT) = 0.001
      ELSE
        DCONC(IPT)= FLOAT(NFRE) / FLOAT(NFRE + NAIR)
      ENDIF
100 CONTINUE
      DCONC(1) = IPT
      DCONC(IPT0) = IPT - IPT0
      SCONC(1) = IPT
      SCONC(IPT0) = IPT - IPT0
      RETURN
      END

      SUBROUTINE ICORE (AS,MAXR,NSURF1,COR1,COR2,IORD)
      COMMON /SIZPRM/ NXSPAC,NYSPAC,MXNPRT,NPART,NNUC,NLAY
      COMMON /PARAM/ NPARTO,PROB,PROB2,PROB3,CONC,IZ,MXIY,ICLUST,KGEN,
1 MN1Y,MXFRE,IAMBLK
      REAL*4 COR1(100000),COR2(100000)
      INTEGER*2 AS(NXSPAC,NYSPAC),MAXR,I2ZERO
      PARAMETER (I2ZERO=0)
      INTEGER*4 NPART,MXGEN,KGEN,ICALC1,ICALC2
      RETURN

      ENTRY CORE

      IARD=IORD*MAXR
      C CHOOSE A PARTICLE
      DO 200 IR=1,MAXR
        ICRR=IARD+IR
        ICALC1= 0
        ICALC2= 0
        DO 100 I=1,NXSPAC
          DO 100 J=1,NYSPAC
            IF (AS(I,J).EQ.I2ZERO) GOTO 100
          C X -NEIGHBORS
          C RIGHT
            IQ = I + IR
            IF (IQ.GT.NXSPAC) IQ=IQ-NXSPAC+1
            IF (AS(IQ,J).NE.I2ZERO) THEN
              ICALC1=ICALC1+1
              ICALC2=ICALC2+AS(IQ,J)-1
            ENDIF
          C LEFT
            IQ = I - IR
            IF (IQ.LE.0) IQ=NXSPAC+IQ-1
            IF (AS(IQ,J).NE.I2ZERO) THEN

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                ICALC1=ICALC1+1
                ICALC2=ICALC2+AS(IQ,J)-1
            ENDIF
C Y -NEIGHBORS
C UP
                IQ = J + IR
                IF (IQ.GT.NYSPAC) IQ=IQ-NYSPAC+1
                IF (AS(I,IQ).NE.I2ZERO) THEN
                    ICALC1=ICALC1+1
                    ICALC2=ICALC2+AS(I,IQ)-1
                ENDIF
C DOWN
                IQ = J - IR
                IF (IQ.LE.0) IQ=NYSPAC+IQ-1
                IF (AS(I,IQ).NE.I2ZERO) THEN
                    ICALC1=ICALC1+1
                    ICALC2=ICALC2+AS(I,IQ)-1
                ENDIF
100    CONTINUE
C AVERAGE COR1(R) OVER 4 NEIGHBOR DIRECTIONS
        COR1(ICRR)=ICALC1/ (4.0 * NSURF1)
C AVERAGE COR2(R) OVER 8 NEIGHBOR DIRECTIONS
        COR2(ICRR)=ICALC2/ (8.0 * NSURF1)
200    CONTINUE
        RETURN
        END

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POR33450
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POR33470

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