

## Electrical characterization of linear and non-linear random networks and mixtures

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### SUMMARY

A material composed of a mixture of distinct homogeneous media can be considered as a homogeneous one at a sufficiently large observation scale. The problem of mixture characterization has been exactly solved in case of linear random mixtures, that is, materials for which the various components are isotropic, linear and mixed together as an ensemble of particles having random shapes and positions. In the present work the authors briefly review the linear theory and then consider mixtures of non-linear media. In particular they give formulas for obtaining their constitutive equations for current density, electrical displacement, and magnetic induction. These relations have been derived by means of heuristic considerations on random networks and they have been verified with simulations obtaining a high degree of accuracy. Copyright © 2003 John Wiley & Sons, Ltd.

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### 1. INTRODUCTION

A widely dealt topic concerning the physical behaviour of heterogeneous materials (mixtures) is that of calculating their (d.c.) conductivity starting from the knowledge of the conductivity of each medium composing the mixture as well as of the structural properties of the mixture itself (percentage of each medium, shapes and relative positions of the single parts of the various media). Clearly, it concerns with isotropic linear media, which combine to form linear mixtures. We find in literature a large number of approximate analytical expressions for the effective conductivity of composed media as a function of the conductivity of its homogeneous constituents and some stoichiometric parameter [1–3].

It must be underlined that from a merely mathematical standpoint, the problem of calculating the mixture conductivity is identical to a number of others, for instance to that regarding permeability (in a magnetostatic situation), permittivity (in the electrostatic case), thermal conductivity (in a steady-state thermal regime) and so on. This point is rather well known: we remember that the formal identity of all cases bears on the fact that in each case the

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fundamental physical function dealt with is some  $V(x, y, z)$  which is harmonic in each single homogeneous region: for example,  $V$  is the electrical potential, or the magnetic one, or the temperature; and the boundary conditions for this  $V$  are formally identical in the various examples considered here: they all state that across the boundary between two different regions the product of the normal derivative of  $V$  and of the proper parameter (conductivity, permittivity, permeability, thermal conductivity etc.) is continuous across the boundary.

If  $V(x, y, z)$  is the electrical potential, one has  $\nabla^2 V = 0$  in each homogeneous region and  $\sigma \partial V / \partial n$  continuous at any interface between different media ( $\sigma$  is the conductivity); these equations can be studied with finite difference methods, so that the mixture can be replaced with a cubic lattice network (or a square lattice network, in a two-dimensional case) composed of linear resistors. This is the transition between actual continuous media and lumped networks.

The paper is devoted to an investigation on the linear and non-linear random networks and mixtures. Roughly speaking, a random (or statistical) mixture is a material composed of little particles, having completely haphazard sizes, positions and shapes; each particle is entirely composed of one of some given homogeneous materials. This definition has a clear intuitive meaning, though it does not withstand an accurate criticism. This fact explains why, as a matter of fact, several different theoretical results have been obtained by various authors, e.g. concerning the permittivity or the conductivity of a random mixture [4].

Some elements that may characterize a random material are:

- the nature of the materials that compose the mixture (they may be linear or non-linear, isotropic or anisotropic);
- the concentrations of the components;
- the dimensionality of the mixture (a mixture is said two-dimensional when its properties do not depend on a given direction; a mixture is said three-dimensional when its properties depend on all the co-ordinates  $x, y, z$ ).

If we are dealing with continuous random media the analysis of the equations by means of the finite difference method leads to lumped random networks. Therefore, the interest to random networks chiefly arises from the study of these random mixtures.

From a historical point of view we review some formulas describing a mixture composed of two linear isotropic components; one of the most famous is the Maxwell formula [1,5] for a strongly diluted suspension of spheres (three-dimensional case):

$$v \frac{\sigma_1 - \sigma_2}{2\sigma_1 + \sigma_2} = \frac{\sigma_1 - \bar{\sigma}}{2\sigma_1 + \bar{\sigma}} \quad (1)$$

where  $\sigma_1$  is the conductivity of the suspending medium,  $\sigma_2$  is the conductivity of the embedded spheres,  $v$  the volume fraction of medium 2 and  $\bar{\sigma}$  is the equivalent conductivity of the mixture. A similar equation holds true for a mixture of parallel circular cylinders (two-dimensional case):

$$v \frac{\sigma_1 - \sigma_2}{\sigma_1 + \sigma_2} = \frac{\sigma_1 - \bar{\sigma}}{\sigma_1 + \bar{\sigma}} \quad (2)$$

Other relations have been found by Bruggeman and Hanai and should maintain the validity also for less diluted suspensions [1]; the three-dimensional case leads to the

formula:

$$1 - v = \frac{\sigma_2 - \bar{\sigma}}{\sigma_2 - \sigma_1} \left( \frac{\sigma_1}{\bar{\sigma}} \right)^{1/3} \quad (3)$$

and the two-dimensional case to the following:

$$1 - v = \frac{\sigma_2 - \bar{\sigma}}{\sigma_2 - \sigma_1} \left( \frac{\sigma_1}{\bar{\sigma}} \right)^{1/2} \quad (4)$$

Finally, we recall the Looyenga–Landau–Lifshitz formulas that should be approximately used in the same hypothesis of the previous expressions [1,6]; they are only an approximation, explicitly derived under the hypothesis that  $\sigma_1 \cong \sigma_2$ :

$$\bar{\sigma}^{1/3} = (1 - v)\sigma_1^{1/3} + v\sigma_2^{1/3} \quad (5)$$

for the three-dimensional case and

$$\ln \bar{\sigma} = (1 - v) \ln \sigma_1 + v \ln \sigma_2 \quad (6)$$

for the two-dimensional case. Equation (6) is not explicitly derived by Looyenga–Landau–Lifshitz, but it can be easily found by repeating their calculation for the two-dimensional case. It is interesting to observe, for the following purposes, that by letting  $v = 1/2$  the above formula (6) yields  $\bar{\sigma} = \sqrt{\sigma_1 \sigma_2}$ ; this relation, rather surprisingly, will be exactly verified for a particular class of random mixtures in the following section. The proof will be based on duality concepts for two-dimensional mixtures [7].

The structure of the paper is the following: in Sections 2–4 we describe the analysis performed on linear networks and mixtures and we show the proposed numerical solutions drawing a comparison with the analytical relations. In Section 5 we generalize the linear results to the non-linear networks and mixtures using the standard small-signal technique. Furthermore, in Section 6, another generalization is made, from symmetric to asymmetric components: some general relations have been found out to describe the average behaviour of non-linear networks. Finally, some non-linear simulations are presented to verify the heuristic predictions.

## 2. DUALITY FOR TWO-DIMENSIONAL MIXTURES

Firstly we consider a medium having a conductivity  $\sigma(x, y)$  arbitrarily variable with  $x$  and  $y$  but independent of  $z$ . The average conductivity  $\bar{\sigma}$  is defined as the conductance for unit square in the  $xz$  plane, for a unit spacing between the electrodes.

Figure 1 represents the system under consideration. In a d.c. situation, one has:  $\nabla \cdot \mathbf{J} = 0$ ,  $\mathbf{J} = \sigma \mathbf{E}$ ,  $\mathbf{E} = -\nabla V$  where  $\mathbf{J}$  is the current density,  $\mathbf{E}$  is the electrical field and  $V$  is the potential; therefore, with reference to Figure 1:

$$\begin{aligned} \frac{\partial}{\partial x} \left( \sigma \frac{\partial V}{\partial x} \right) + \frac{\partial}{\partial y} \left( \sigma \frac{\partial V}{\partial y} \right) &= 0 \\ V(x, l) &= V_0 \\ V(x, 0) &= 0 \end{aligned} \quad (7)$$

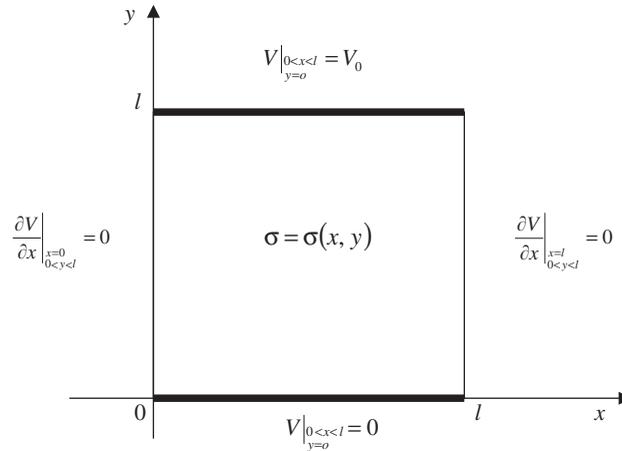


Figure 1. A two-dimensional random mixture (size  $l \times l$ ) characterized by the non-homogeneous conductivity  $\sigma(x, y)$  and embedded between two electrodes  $V = 0$  and  $V_0$ . We may observe the boundary conditions on the lateral sides, where no current can flow.

$$\begin{aligned}\frac{\partial V}{\partial x}(0, y) &= 0 \\ \frac{\partial V}{\partial x}(l, y) &= 0\end{aligned}$$

The first two boundary conditions are imposed by the presence of two electrodes at the potential 0 ( $y = 0$ ) and  $V_0$  ( $y = l$ ) and the others mean that there is no current flowing across the vertical sides of the structure. This differential problem has only one solution. The current flowing between the electrodes per unit square is

$$J = \frac{1}{l} \int_0^l \sigma(x, y) \frac{\partial V(x, y)}{\partial y} dx \quad (8)$$

which is independent on the variable  $y$ ; the average conductivity is defined as

$$\bar{\sigma} = \frac{Jl}{V_0} = \frac{1}{V_0} \int_0^l \sigma(x, y) \frac{\partial V(x, y)}{\partial y} dx \quad (9)$$

Now, if the first equation in (7) is written as follows:

$$\frac{\partial}{\partial x} \left( -\sigma \frac{\partial V}{\partial x} \right) = \frac{\partial}{\partial y} \left( \sigma \frac{\partial V}{\partial y} \right) \quad (10)$$

it is apparent that the two-dimensional differential form  $\sigma(\partial V/\partial y)dx - \sigma(\partial V/\partial x)dy$  is exact, which means that there exists a function  $U(x, y)$  such that

$$\begin{aligned}\frac{\partial U}{\partial x} &= \sigma \frac{\partial V}{\partial y} \\ \frac{\partial U}{\partial y} &= -\sigma \frac{\partial V}{\partial x}\end{aligned}\quad (11)$$

This function  $U$  is fully determined but for an arbitrary integration constant. Furthermore,  $U$  fulfils a differential equation very similar to that of  $V$ :

$$\frac{\partial}{\partial x} \left( -\frac{1}{\sigma} \frac{\partial U}{\partial x} \right) = \frac{\partial}{\partial y} \left( \frac{1}{\sigma} \frac{\partial U}{\partial y} \right) \quad (12)$$

Now we are going to analyse the boundary conditions for  $U$ . From Equations (7) and (11), one finds that along each vertical side  $U$  is constant (since there  $\partial V/\partial x = 0$ , hence  $\partial U/\partial y = 0$ ). So we can impose  $U = 0$  on the left side ( $x = 0$ ); this determines the additive constant inherent to  $U$ ; along the right side ( $x = l$ ),  $U$  has some unknown but constant value, referred to as  $U_0$ . On the other hand, along the horizontal sides, one has  $\partial U/\partial y = 0$  (since there  $V$  is constant with respect to  $x$ , hence  $\partial V/\partial x = 0$ ). So, we have found two dual cases: the original one with horizontal electrodes, conductivity  $\sigma$  and potential  $V$ ; the dual case with vertical electrodes, conductivity  $1/\sigma$  and potential  $U$ . The original structure has average conductivity:

$$\bar{\sigma} = \frac{Jl}{V_0} = \frac{1}{V_0} \int_0^l \sigma(x, y) \frac{\partial V(x, y)}{\partial y} dx = \frac{U_0}{V_0} \quad (13)$$

The dual system fulfils the relation

$$\bar{\sigma}^* = \frac{J^*l}{U_0} = \frac{1}{U_0} \int_0^l \frac{1}{\sigma(x, y)} \frac{\partial U(x, y)}{\partial x} dy = \frac{V_0}{U_0} \quad (14)$$

Therefore, the following duality theorem holds true: if the original structure has conductivity  $\sigma(x, y)$ , horizontal electrodes and average conductivity  $\bar{\sigma}$ , the dual structure with conductivity  $1/\sigma(x, y)$  and vertical electrodes has average conductivity  $1/\bar{\sigma}$ .

This theorem may be applied to the case of a random two-dimensional mixture: we consider a random mixture with two components with conductances respectively  $\sigma_1$  and  $\sigma_2$  (with volume fraction  $v$  of the second medium). We assume by hypothesis that *the only structural information on the mixture is the volume fraction of the second medium*, since either medium is composed of particles completely randomised in size, position and shape. So, we state that  $\bar{\sigma}$  is a function of  $\sigma_1$ ,  $\sigma_2$  and  $v$  only:

$$\bar{\sigma} = F(\sigma_1, \sigma_2, v) \quad (15)$$

This statement is actually a *definition* of random mixture: a mixture composed by parts so randomised, as above said, that the only significant structural information is merely the volume fraction  $v$  ( $1 - v$  for the first medium). We understand that this definition could hardly be defended against a severe criticism; however it is heuristically useful to calculate average parameters, as it is shown in the sequel.

Now,  $F$  must be homogeneous, of degree one with respect to  $\sigma_1$  and  $\sigma_2$  (this is the so-called Wiener postulate [2]). This implies

$$\bar{\sigma} = \sigma_1 F\left(1, \frac{\sigma_2}{\sigma_1}, v\right) \quad (16)$$

Another mixture having  $v$  as concentration of the first medium is characterized by another conductance  $\Sigma$  such that

$$\Sigma = \sigma_2 F\left(1, \frac{\sigma_1}{\sigma_2}, v\right) \quad (17)$$

The dual mixture of the original one is once again described by the relation  $F$  because of the randomness and of the isotropy, so that

$$\bar{\sigma}^* = \frac{1}{\bar{\sigma}} = F\left(\frac{1}{\sigma_1}, \frac{1}{\sigma_2}, v\right) = \frac{1}{\sigma_1} F\left(1, \frac{\sigma_1}{\sigma_2}, v\right) \quad (18)$$

From Equations (17) and (18) one finds  $\bar{\sigma}\Sigma = \sigma_1\sigma_2$ .

If we consider a random two-dimensional mixture with  $v = 1/2$  we must have  $\bar{\sigma} = \Sigma$ ; therefore, in such case

$$\bar{\sigma} = \Sigma = \sqrt{\sigma_1\sigma_2} \quad (19)$$

This is the main result of this paragraph: a two-dimensional random mixture, composed of two media each at 50% volume concentration, has a conductivity given by the geometrical mean between  $\sigma_1$  and  $\sigma_2$ .

### 3. RANDOM MIXTURES OF LINEAR HOMOGENEOUS ISOTROPIC MEDIA

This section deals with random mixtures composed of  $N$  homogeneous media, having volume fractions  $v_1, v_2, \dots, v_n$  ( $\sum v_i = 1$ ) and conductivities  $\sigma_1, \sigma_2, \dots, \sigma_n$ , respectively. Our aim is to find an expression of the mixture conductivity in terms of the above quantities. We review here a particular solution given in literature [8], which is very suitable to make the generalization to the non-linear cases considered in the subsequent sections.

Firstly, we again consider the case of a random mixture (either two-dimensional or three-dimensional) composed by two media  $\sigma_1$  and  $\sigma_2$  with concentrations  $v_1 = 1 - v_2$  and  $v_2$ , respectively. For such a mixture we can use a relation of this kind for the average conductivity:  $\sigma = F(\sigma_1, \sigma_2, v_2)$ . The function  $F$ , which is *a priori* unknown in its structure, must fulfil the following constraints:

$$\begin{aligned} \sigma_1 &= F(\sigma_1, \sigma_2, 0) \\ \sigma_2 &= F(\sigma_1, \sigma_2, 1) \\ F(h\sigma_1, h\sigma_2, v_2) &= hF(\sigma_1, \sigma_2, v_2) \end{aligned} \quad (20)$$

The third one is the Wiener's postulate already used (it simply states the fact that, if one changes the system of units, then  $\sigma$  must change as  $\sigma_1, \sigma_2$  do). In order to develop the study, first a discrete model is assumed for the mixture: it is considered as a lumped-element resistive

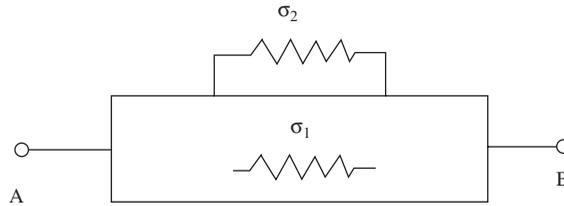


Figure 2. A generic one-port representing a whole network where a resistor  $\sigma_2$  and several resistors  $\sigma_1$  are present and randomly distributed.

network, composed of resistors arranged in a regular order; for example, the nodes form a cubic lattice (three-dimensional mixture) or a square lattice (two-dimensional mixture). The network elements are specified according to one of the following criteria:

- (I) each resistor of the network has conductance  $\sigma_k$  ( $k = 1$  or  $2$ ) with probability  $v_k$ ;
- (II) the network contains  $100 \cdot v_k$  percent resistors having conductance  $\sigma_k$ .

Moreover, the resistors are spatially distributed at random in the network. Both criteria are used to develop the study, whilst only the second one has been used for the computer simulations whose results will be presented later on. However, when the number of resistors is large, as we suppose, the two criteria should practically coincide in the average.

Let us examine the situation for very low values of  $v_2$ ; according to the criterion II, in the discrete model the lowest  $v_2$  is reached when the lumped network contains only a single ' $\sigma_2$ ' element. In this case the whole network can be represented as shown in Figure 2.

According to standard network-theory theorems, the conductance  $\sigma_{AB}$  between terminals  $A$  and  $B$  is a bilinear function of  $\sigma_2$  [9]:

$$\sigma_{AB} = \frac{a'\sigma_2 + b'}{c'\sigma_2 + d'} \quad (21)$$

where  $a', b', c'$  and  $d'$  depend on  $\sigma_1$ .

The difference of average conductivity between the case of a homogeneous medium with conductivity  $\sigma_1$  and the case of a mixture with a single element  $\sigma_2$  is

$$\sigma_{AB}|_{\text{only one } \sigma_2} - \sigma_{AB}|_{\text{all } \sigma_1} = \frac{a'\sigma_2 + b'}{c'\sigma_2 + d'} - \sigma_1 = \frac{a\sigma_2 + b}{c\sigma_2 + d} \quad (22)$$

or, dividing by  $c$  (we assume  $c \neq 0$ ; it could be easily shown that the case  $c = 0$  leads to an expression of  $F$  which is surely incorrect for a random mixture):

$$\sigma_{AB}|_{\text{only one } \sigma_2} - \sigma_{AB}|_{\text{all } \sigma_1} = \frac{(a/c)\sigma_2 + (b/c)}{\sigma_2 + (d/c)} \quad (23)$$

Therefore, from now on, we can consider the relation:

$$\left. \frac{\partial F}{\partial v_2} \right|_{v_2=0} = \frac{A\sigma_2 + B}{\sigma_2 + D} \quad (24)$$

where  $A$ ,  $B$  and  $D$  are functions of  $\sigma_1$  only.

We try to identify  $A$ ,  $B$  and  $D$  using the properties of  $F$ : if  $\sigma_1 = \sigma_2$  it follows that  $F(\sigma_1, \sigma_1, v_2) = \sigma_1 = \text{cost}$  and then  $\partial F / \partial v_2 = 0$ ; therefore  $A\sigma_1 + B = 0$  and  $B = -\sigma_1 A$ . Consequently, the first result is:

$$\left. \frac{\partial F}{\partial v_2} \right|_{v_2=0} = A \frac{\sigma_2 - \sigma_1}{\sigma_2 + D} \quad (25)$$

But  $F$  is a homogeneous function of  $\sigma_1$  and  $\sigma_2$  and clearly  $\partial F / \partial v_2$  has the same property, so that, we can write for any value of the constant  $h$ :

$$A(h\sigma_1) \frac{h\sigma_2 - h\sigma_1}{h\sigma_2 + D(h\sigma_1)} = hA(\sigma_1) \frac{\sigma_2 - \sigma_1}{\sigma_2 + D(\sigma_1)} \quad (26)$$

or, after some straightforward computation:

$$A(h\sigma_1)\sigma_2 + A(h\sigma_1)D(\sigma_1) = hA(\sigma_1)\sigma_2 + A(\sigma_1)D(h\sigma_1) \quad (27)$$

Dividing the above expression by  $\sigma_2$  and letting  $\sigma_2 \rightarrow \infty$  we find  $A(h\sigma_1) = hA(\sigma_1)$ , hence  $A(\sigma_1) = \alpha\sigma_1$  where  $\alpha$  is some constant; in Equation (27) we can let  $\sigma_2 = 0$  obtaining:  $A(h\sigma_1)D(\sigma_1) = A(\sigma_1)D(h\sigma_1)$  which is equivalent to  $\alpha h\sigma_1 D(\sigma_1) = \alpha\sigma_1 D(h\sigma_1)$  so:  $D(\sigma_1) = \beta\sigma_1$  being  $\beta$  some other constant.

Summing up, one has

$$\left. \frac{\partial F}{\partial v_2} \right|_{v_2=0} = \alpha\sigma_1 \frac{\sigma_2 - \sigma_1}{\sigma_2 + \beta\sigma_1} \quad (28)$$

where, as it has been said above,  $\alpha$  and  $\beta$  are constants (independent on  $\sigma_1$  and  $\sigma_2$ ).

Now we are ready to consider a generic mixture with  $N$  different media. Equation (28) plays an essential role in the further development of the theory. Each medium, in the generic mixture, has conductivity  $\sigma_i$  and volume concentration  $v_i$ ;  $\sigma$  is the equivalent conductivity of the mixture.

If we add to the mixture a little volume  $dv$  with conductivity  $\sigma_k$  we create a new heterogeneous material formed by the original mixture (conductivity  $\sigma$ ) and a volume with conductivity  $\sigma_k$ ; this new mixture can be analysed considering it as a two component one; therefore, its conductivity, referred to as  $\sigma'_k$ , is given by

$$\sigma'_k = F(\sigma, \sigma_k, dv/1 + dv) \cong F(\sigma, \sigma_k, dv) \quad (29)$$

This procedure is not new in principle, since it has been used, e.g. by Maxwell, Looyenga and Bruggemann even for non infinitesimal volumes of the medium added to the original mixture; here it is used in a weaker sense, i.e. for an infinitesimal added volume. The above considerations are not justifications of the procedure, which remains heuristic and whose validity stems on the agreement with the computer simulation discussed later on.

Now, let us suppose that the procedure of adding a volume  $dv$  is made for each medium ( $k = 1, 2, \dots, n$ ), giving a probability  $v_k$  to the case of conductivity  $\sigma_k$ . The average conductivity is clearly the very same  $\sigma$  of the original mixture, hence one may state that Equation (30)

below holds true:

$$\sigma = \sum_{k=1}^N \sigma'_k v_k = \sum_{k=1}^N F(\sigma, \sigma_k, dv) v_k = \sum_{k=1}^N \left[ F(\sigma, \sigma_k, 0) + \frac{\partial F}{\partial v} \Big|_{\substack{\sigma \\ \sigma_k \\ 0}} dv \right] v_k \quad (30)$$

or, after some manipulation

$$0 = \sum_{k=1}^N \frac{\partial F}{\partial v} \Big|_{\substack{\sigma \\ \sigma_k \\ 0}} v_k \quad (31)$$

and, equivalently

$$\frac{1}{(\beta + 1)\sigma} = \sum_{k=1}^N \frac{v_k}{\sigma_k + \beta\sigma} \quad (32)$$

Equation (32) is, in implicit form, the required expression which gives the average conductivity  $\sigma$  in terms of the  $\sigma_k$  and of the  $v_k$ .

The evaluation of the constant  $\beta$ , which for the moment is unknown, is discussed now. In a one-dimensional structure the result must be

$$\frac{1}{\sigma} = \sum_{k=1}^N \frac{v_k}{\sigma_k} \quad (33)$$

since it is a very simple case of ‘conductances in cascade’.

Indeed, a one-dimensional mixture is composed of a superposition of sheets with given conductances, and the current flows perpendicular to the sheets.

So, in this condition we can let  $\beta = 0$ . Moreover, it is known that for two-dimensional structures with  $v = 1/2$  and  $N = 2$  the relation  $\sigma = \sqrt{\sigma_1 \sigma_2}$  must hold true (see Section 2 of this paper); this leads, after some straightforward calculations, to  $\beta = 1$ . Hence, we state that  $\beta = 1$  for any two-dimensional structure. This theoretical result has been tested over a large number of different situations through a computer simulation as discussed in a following section.

On the basis of this discussion, it seems reasonable to assume that  $\beta + 1$  actually is the *dimensionality* of the mixture. Therefore, we conjecture that  $\beta = 2$  for three-dimensional structures. As we have previously recalled, all these formulas are correct for many physical parameters: permeability (in a magnetostatic situation), permittivity (in the electrostatic case), thermal conductivity (in a steady-state thermal regime) and so on. Therefore, at this point a summary of the final results with generic symbol  $p$  (and not with conductivity symbol  $\sigma$ ) is useful:

*Two-dimensional case:*

The equation for the scalar field  $f$  involved (the electrical potential in the previous discussion) and the definition of the average parameter (the conductivity for example) are:

$$\bar{\nabla}(p \bar{\nabla} f) = 0 \quad \bar{p} = \int_0^1 p \frac{\partial f}{\partial y} dx \quad (34)$$

Here  $p$  is a random scalar field, which describes the random mixture.

The proposed solution of the problem is

$$\frac{1}{2\bar{p}} = \sum_{k=1}^N \frac{v_k}{p_k + \bar{p}} \quad (35)$$

*Three-dimensional case:*

The equation for the scalar field involved and the definition of the average parameter are:

$$\bar{\nabla}(p\bar{\nabla}f) = 0 \quad \bar{p} = \int_0^1 \int_0^1 p \frac{\partial f}{\partial z} dx dy \quad (36)$$

where the  $z$ -axis is orthogonal to the planes where  $f=0$  and 1 and  $p$  is the random scalar field. The obtained solution is given by

$$\frac{1}{3\bar{p}} = \sum_{k=1}^N \frac{v_k}{p_k + 2\bar{p}} \quad (37)$$

Equations (35) and (37) are  $N$ th degree algebraic equations in the unknown  $\bar{p}$  (it is not difficult to verify that a single positive or null solution exists for both equations).

We remember that the above results had been heuristically obtained with the following assumptions on the statistical composition of the heterogeneous material: we subdivide the whole medium in many smaller pieces; each of these part is homogeneous and has the parameter  $p_k$  with probability  $v_k$ . The value of the parameter in a given little piece is statistically independent on the values assigned in the other pieces. This means, using the law of large numbers, that the stoichiometric coefficient of the  $k$ th components is  $v_k$ . Finally, we assumed that a large network or a mixture subdivided in many little pieces (with size very smaller than the whole compound) normally behaves as the mean value given by Equation (35) and (37).

In other words, the density probability of the value of the parameter in a given piece of material has the form  $W(p) = \sum_{k=1}^N p_k \delta(p - p_k)$  where the  $p_k$ 's and the  $v_k$ 's are the above defined quantities and  $\delta(x)$  is the Dirac delta function: this means that the probability for  $p$  to assume values between  $p$  and  $p + dp$  is exactly  $W(p) dp$  and the values are statistically independent for different parts of the whole material.

These are the main assumptions used to derive our predictions on the behaviour of the heterogeneous material. Obviously, if one considers values of the parameter extracted from a density probability different from the discrete one above described, the results presented in this work are no more valid. Therefore, any continuous density probability is not correct for our analysis. In conclusion, we observe that other limits of validity are the independence of the values assumed in each part and the large size of the networks, which simulates a large scale of observation of the mixture.

#### 4. RANDOM MIXTURES AND RANDOM NETWORKS

A random mixture can be numerically analysed by means of the finite difference method [10]; so doing, the procedure leads to lumped electrical networks, which approximately describe the mixture. In a two-dimensional case such a network can be represented as in Figure 4.

The voltage  $V$  is assumed constant and we observe the current  $I$  when the components are randomly varied. Each component is a linear resistor with the value taken at random (with the probability proportional to the concentration) in a set of  $N$  values corresponding to the  $N$  different materials.

Figure 4 represents the case  $N=2$ . Let  $M$  be the number of components in any row and  $Q$  the number of components in any column of the two-dimensional grid; moreover, let  $G_1$  and  $G_2$  be the conductances of the introduced linear resistors; the analysis outlined in the previous section gives the following average solution if one considers, for example,  $v=1/2$ :

$$I = \frac{M+1}{Q} \sqrt{G_1 G_2} V \quad (38)$$

This means that the behaviour of the network is unchanged if one consider all the components with conductance  $\sqrt{G_1 G_2}$ .

Similarly, for the three-dimensional case we may consider a grid with dimensions  $M \times L \times Q$  and we randomly introduce two components  $G_1$  and  $G_2$  with equal probability. The analytical result for the average characteristic is given by

$$I = \frac{(M+1)(L+1)}{Q} \left[ \frac{G_1 + G_2}{8} + \frac{1}{4} \sqrt{\left( \frac{G_1 + G_2}{2} \right)^2 + 8G_1 G_2} \right] V \quad (39)$$

In both two- and three-dimensional cases, Equations (38) and (39) have been verified by numerical simulation; we use the Monte-Carlo method averaging over several networks with different distribution of resistors; each resistor, in a given network, has been randomly chosen with equal probability between two possible values ( $G_1$  and  $G_2$ ). For each network we calculate the whole actual conductance corresponding to the particular choice of the resistors: when a network is established it is solved by means of the relaxation method applied to the equations written in each node (it is well known that this method always leads to convergence in the case of linear networks). Finally, after having considered a large number of networks, we calculated the arithmetic mean of the obtained values for the conductance; this average value corresponds to a linear characteristic which is very similar to the analytical one given by Equation (38) or (39). Therefore, such simulations have verified the heuristic predictions with high degree of accuracy. Furthermore, other simulations have been performed with different concentrations for the media and with a greater number of composing media. A particular case is found when one medium has zero conductivity, say  $\sigma_2=0$ , and the other one has positive conductivity  $\sigma_1$ ; this case is interesting because it is the most critical one for linear mixtures [1]. For two-dimensional structures Equation (35) becomes  $\sigma^2 + \sigma\sigma_1(2v_2 - 1) = 0$  which has two solutions:  $\sigma = 0$  and  $\sigma_1(1 - 2v_2)$ .

Since it is reasonable to assume that  $\sigma$  be a non-negative, continuous function of  $v_2$ , the true solution is given by  $\sigma = \sigma_1(1 - 2v_2)$  if  $0 < v_2 < 1/2$  and  $\sigma = 0$  if  $1/2 < v_2 < 1$ . Let us consider now the case when  $\sigma_2=0$  in two-component three-dimensional mixtures; Equation (37) becomes  $2\sigma^2 + \sigma\sigma_1(3v_2 - 2) = 0$  with two solutions:  $\sigma = 0$  and  $\sigma = \sigma_1(1 - 3v_2/2)$ ; requiring, as before, that  $\sigma$  be a non negative continuous function of  $v_2$  the correct solution is  $\sigma = \sigma_1(1 - 3v_2/2)$  if  $0 < v_2 < 2/3$  and  $\sigma = 0$  if  $2/3 < v_2 < 1$ . Figure 3 shows the comparison between these analytical results and the numerical ones obtained with the corresponding two and three-dimensional random networks. Once again, the agreement with the equations is very satisfactory.

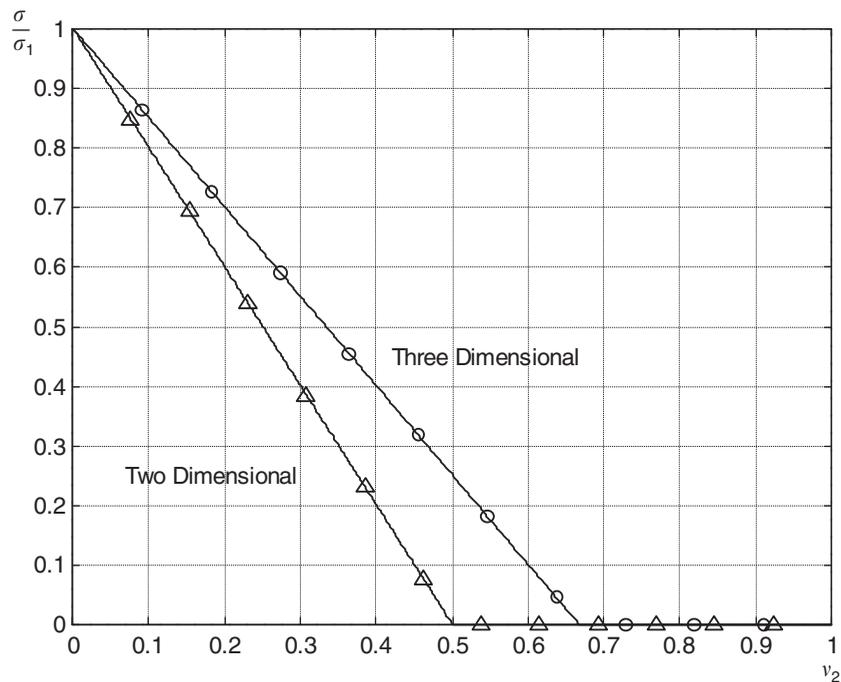


Figure 3. Comparison between analytical results (continuous lines) and numerical ones (triangles in two-dimensional case and circles in three-dimensional case) for the two component linear mixture, having  $\sigma_2=0$ , described in the main text. For the two-dimensional case we used a square lattice with 100 nodes and the result represents the mean value over 20 realizations of the network; in the three-dimensional case the cubic lattice contained 125 nodes and we used 25 different networks.

## 5. ISOTROPIC NON-LINEAR NETWORKS AND MIXTURES

The problem of mixture characterization has been previously solved in case of linear random mixtures, that is, materials for which the various components are isotropic, linear and mixed together as an ensemble of particles having random shapes and positions. In the present section we consider mixtures of isotropic but non-linear media and by using statistical-network methods we obtain formulas for evaluating their constitutive equations. Each medium is described by a constitutive relation of the kind  $J = f_k(E)$  (or  $D = f_k(E)$ ,  $B = f_k(H)$  and so on) between scalar quantity because of the isotropy. The function  $f_k(v)$  represents the non-linearity of the material and must be considered an odd function  $f_k(-v) = -f_k(v)$ .

We consider  $N$  different media mixed together with concentrations  $v_1, v_2, \dots, v_n$  and  $N$  odd functions  $f_1, f_2, \dots, f_n$ : we are interested in the non-linear equivalent characteristic of the whole structure  $J = f(E)$ . We can verify that

$$J = f(E) = \int_0^E g(y) dy \quad (40)$$

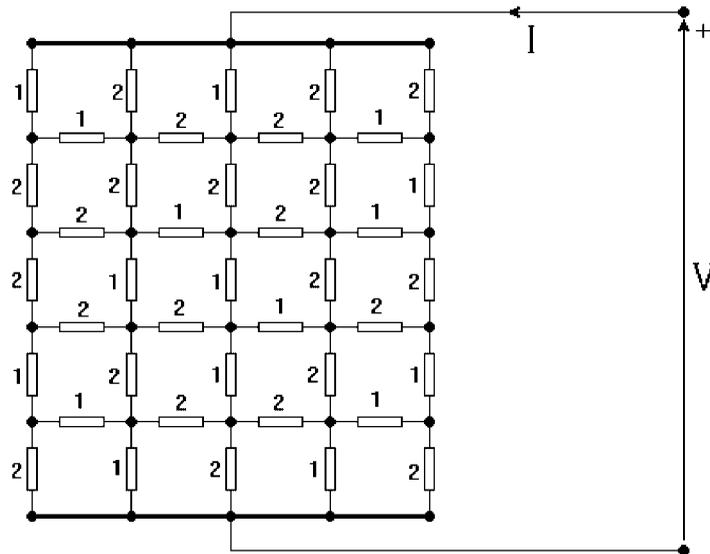


Figure 4. A two-dimensional square mesh random network: the symbols 1 and 2 identify two different kinds of linear or non-linear resistive one-port.  $V$  and  $I$  represent the whole voltage and current, respectively.

$g(y)$  satisfies Equation (41) in the two-dimensional case:

$$\frac{1}{2g(y)} = \sum_{k=1}^N \frac{v_k}{f'_k(y) + g(y)} \quad (41)$$

and Equation (42) in the three-dimensional case:

$$\frac{1}{3g(y)} = \sum_{k=1}^N \frac{v_k}{f'_k(y) + 2g(y)} \quad (42)$$

To justify such statements, first of all, we consider a square-mesh network (composed by  $M$  components along each row and  $Q$  components along each column) of memoryless non-linear one ports, each one described by some voltage–current relationship  $i = f_k(v)$  being the  $f_k$ 's,  $k = 1, \dots, N$ , increasing differentiable functions of their argument. So, there are  $N$  different kinds of non-linear resistors. It is assumed that both  $Q$  and  $M$  are very large, in order to simulate a large observation scale, and that the various resistors are completely randomised as regards their spatial distribution in the network.

The whole structure is endowed with two electrodes (see Figure 4 where the case  $N = 2$  is considered: one deals with a random distribution of two types of non-linear resistors indicated in the figure by the labels 1 and 2).

Finally, it is assumed that the fraction  $v_k$  of the  $k$ th type components (i.e. the ratio of their number to the total number of non-linear resistors in the network) is fixed for all values of  $k$ .

In this situation, for any choice of the distribution of the various non-linear resistors in the network, the one-port in figure is a memoryless one, say a resistor (in general again

non-linear) for which there is some relationship,  $I = F^*(V)$ , relating to the overall voltage  $V$  and current  $I$ .

Now, if we take into consideration the various possible spatial distributions of the single resistors in the network (being  $v_1, v_2, \dots, v_k$  fixed) then the network becomes a statistical one, which models a two-dimensional mixture of non-linear media, having the  $v_i$ 's as volume fractions of each component. The question is: what is the relationship  $I = F(V)$  of the statistical one-port, or preferably what is the average of the function  $F^*(V)$ ?  $F(V)$  characterizes the statistical non-linear resistor in Figure 4 (In a continuous scheme, this function, once the appropriate scaling operations are performed, becomes the constitutive equation  $J = F(E)$ , of a two-dimensional random mixture: see below).

To calculate  $F(V)$ , we notice that in the particular case when all media are linear, in such a way that  $f_k(v) = g_k v$  (being the  $g_k$ 's positive constants, actually, conductances) the electrical behaviour of the structure is well known, as already mentioned.

Now, let us consider the non-linear case. If  $V$  is changed to  $V + dV$ , then the current  $I = F(V)$  will change up to some value  $I + dI$ . Now, one can apply the standard small-signal technique, considering a network where the overall voltage is  $dV$ , the overall current is  $dI$ , and each resistor is substituted with its differential conductance  $g_k = f'_k(v)$ .

Each 'small signal' resistor has, in the average, a given voltage bias: the mean voltage across each one-port (in the original network) is precisely  $V/Q$ , in the columns; as regards the resistors in the rows, their average voltage is zero so that their mean contribution may be neglected.

So, the small-signal network corresponding to the original one is linear and composed of resistors having conductances  $g_k = f'_k(V/Q) \geq 0$  (because the function  $f_k$  are increasing). But for such a network, one can apply the theory of linear random network, which yields, for the (differential) conductance  $g = (dI/dV)(Q/(M + 1))$ , the following equation:

$$\frac{1}{2g(V/Q)} = \sum_{k=1}^N \frac{v_k}{f'_k(V/Q) + g(V/Q)} \quad (43)$$

so that

$$I = \frac{M + 1}{Q} \int_0^V g(x) dx \quad (44)$$

With the substitution  $x = Qy$ , the above equations can be reformulated as follows:

$$\frac{1}{2g(y)} = \sum_{k=1}^N \frac{v_k}{f'_k(y) + g(y)} \quad (45)$$

$$I = F(V) = (M + 1) \int_0^{V/Q} g(y) dy$$

These equations allow us to calculate  $I = F(V)$ , though implicitly: specifically, for any  $y$  in the range  $0 \leq y \leq V/Q$  one solves the (algebraic, of degree  $N - 1$ ) equation for  $g(y)$ , then calculates  $I$  performing the indicated integration.

Similarly, for a three-dimensional network  $MxLxQ$  we have:

$$\frac{1}{3g(y)} = \sum_{k=1}^N \frac{v_k}{f'_k(y) + 2g(y)} \quad (46)$$

$$I = F(V) = (M + 1)(L + 1) \int_0^{V/Q} g(y) dy$$

Equations (45) and (46) completely solve the problem of the two- and three-dimensional non-linear random networks composed by symmetrical components. These relationships have been also verified by means of some computer simulations described in a following section.

From the development of this theory one can easily find the solutions for random mixtures. If we consider  $a$  as differentiation step, the density of current  $J$  in the three-dimensional case corresponds to  $I/((M + 1)a(L + 1)a)$  in the network, the field corresponds to  $V/(Qa)$  and so on for the other quantities; therefore

$$J = f(E) = \int_0^E g(y) dy \quad (47)$$

The relationships for  $g(y)$  are unchanged with respect to Equations (45) and (46).

The following relationships give two examples of application of the random network analysis; for two components with the same probability in the two-dimensional case we have

$$I = (M + 1) \int_0^{V/Q} \sqrt{\frac{df_1}{dx} \frac{df_2}{dx}} dx \quad (48)$$

and in the three-dimensional case

$$I = (M + 1)(L + 1) \int_0^{V/Q} \left[ \frac{1}{8} \left( \frac{df_1}{dx} + \frac{df_2}{dx} \right) + \frac{1}{4} \sqrt{\frac{1}{4} \left( \frac{df_1}{dx} + \frac{df_2}{dx} \right)^2 + 8 \frac{df_1}{dx} \frac{df_2}{dx}} \right] dx \quad (49)$$

Some of these relationships will be numerically verified in a following section.

## 6. RANDOM NETWORKS WITH ASYMMETRIC NON-LINEAR RESISTORS

The previous results describing the non-linear random networks can be generalized to the case of asymmetric components. Now, we suppose that the condition  $f_k(-v) = -f_k(v)$  is not necessarily fulfilled. Hence, we may have  $f_k(-v) \neq -f_k(v)$  with  $f_k(0) = 0$ . Because of the randomness, in average sense, this network will have components with concentration  $v_k/2$  for each polarity and consequently the terms, in the relationship (45) or (46), are split as follows. In the two-dimensional case

$$I = F(V) = (M + 1) \int_0^{V/Q} g(y) dy \quad (50)$$

where

$$\frac{1}{2g(y)} = \sum_{k=1}^N \left[ \frac{v_k/2}{f'_k(y) + g(y)} + \frac{v_k/2}{f'_k(-y) + g(y)} \right] \quad (51)$$

and in the three-dimensional case

$$I = F(V) = (M + 1)(L + 1) \int_0^{V/Q} g(y) dy \quad (52)$$

where

$$\frac{1}{3g(y)} = \sum_{k=1}^N \left[ \frac{v_k/2}{f'_k(y) + 2g(y)} + \frac{v_k/2}{f'_k(-y) + 2g(y)} \right] \quad (53)$$

We may observe that the whole characteristic of the network is, in an average sense, symmetric in spite of the asymmetries of the components: the effect of the constitutive asymmetries is overcome by the randomness of the interconnections of the lumped components, leading at a macroscopic level to a symmetric equivalent characteristic. As already said, all the results obtained for the non-linear case are valid under the following assumptions on the statistics of the random networks (or mixtures): each component of the network (or piece of medium) has a given non-linearity extracted with a fixed probability among a set of functions  $f_1, f_2, \dots, f_n$ ; moreover, the extractions for components or pieces of material are statistically independent. Finally, the further limit of validity is given by the large dimension of the network, which represents the mixture.

## 7. EXAMPLES OF NON-LINEAR RANDOM NETWORKS

From the numerical point of view we may analyse a random non-linear network by means of the Monte-Carlo method applied as follows: one generates several different realizations of the network with the specified non-linear component having their appropriate given concentrations (or probabilities). For each network obtained in such a way we have to find out the whole voltage-current relationship: this means that, one must solve the network for many values of the applied voltage. Fixed a given value of this voltage we can solve the network by means of the non-linear relaxation method. Repeating this procedure for all the interesting values of the potential we obtain the characteristic of the given realization of the network. Averaging over the results corresponding to many realizations we may find the mean or average voltage-current relationship of the whole network. At this point one can draw a comparison with the theoretical result described in the previous sections. Here two particular examples have been considered:

*First example.*

We consider a planar network (size  $M \times Q$ ) with two kinds of components having symmetrical characteristics described by algebraic expressions of degree three:

$$f_1(v) = \alpha v + \beta v^3, \quad f_2(v) = \gamma v + \delta v^3 \quad (54)$$

Moreover we consider equal fractions of the two components. One can see Figure 4 where a realization of the network is shown. In such a case Equation (48) gives the theoretical solution

of the problem. The integral, which appears in Equation (48), can be solved in closed form obtaining the following result:

$$I = 3(M + 1)\sqrt{\beta\delta} \left\{ \frac{a}{3} [2b^2 F(p, q) - (a^2 + b^2)E(p, q)] + \frac{u}{3} (u^2 + a^2 + 2b^2) \sqrt{\frac{a^2 + u^2}{b^2 + u^2}} \right\} \quad (55)$$

where

$$u = \frac{V}{Q}, \quad a^2 = \frac{\alpha}{3\beta}, \quad b^2 = \frac{\gamma}{3\delta}, \quad p = \arctg \frac{u}{b}, \quad q = \frac{\sqrt{a^2 - b^2}}{a}, \quad a > b \quad (56)$$

$F(p, q)$  and  $E(p, q)$  represent the first and second kind elliptic integrals [11], defined as follows:

$$F(p, q) = \int_0^p \frac{d\alpha}{\sqrt{1 - q^2 \sin^2 \alpha}} = \int_0^{\text{sen } p} \frac{dx}{\sqrt{(1 - x^2)(1 - q^2 x^2)}} \quad (57)$$

$$E(p, q) = \int_0^p \sqrt{1 - q^2 \sin^2 \alpha} d\alpha = \int_0^{\text{sen } p} \frac{\sqrt{1 - q^2 x^2}}{\sqrt{1 - x^2}} dx$$

In actual computer simulation the following values for the parameters, which appear in the description of the network, have been used:  $Q = 6$ ,  $M = 5$ ,  $\alpha = 2$ ,  $\beta = 1$ ,  $\gamma = 1$ ,  $\delta = 2$ ; the results are shown in Figure 5.

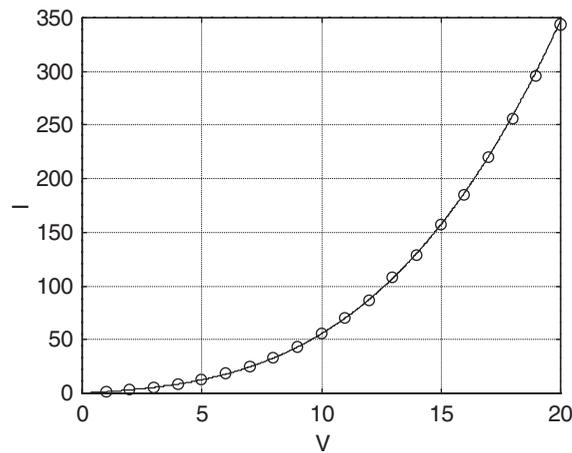


Figure 5. Voltage ( $V$ )–current ( $I$ ) relationship for a two-dimensional random network with dimensions  $Q = 6$ ,  $M = 5$  composed by equal fractions non-linear resistors described by  $i = 2v + v^3$  and  $i = v + 2v^3$  (continuous line: theoretical relationship given by Equation (55); circles: results of computer simulations obtained by means of the non-linear relaxation method over 40 different realizations of the network).

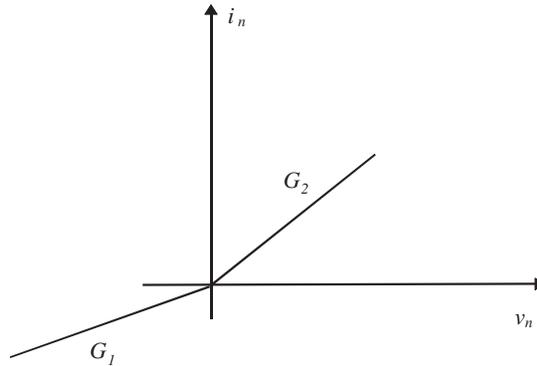


Figure 6. Piecewise linear non-symmetric voltage ( $v_n$ )-current ( $i_n$ ) characteristic of the lumped element considered in the three-dimensional cubic lattice described in the main text.  $G_1$  and  $G_2$  are the conductances for negative and positive voltage, respectively. The orientation of each component in the whole network is randomly obtained.

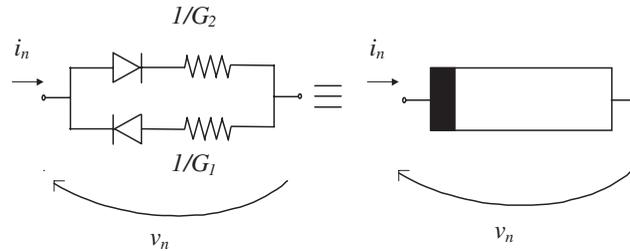


Figure 7. Schematics of the lumped elements used in the network of Figure 8. Their characteristic is shown in Figure 6.

*Second example.*

Now, we consider a three-dimensional mesh formed by only one type of non-symmetric component having piecewise linear characteristic given by

$$i_n = f(v_n) = \begin{cases} G_1 v_n & \text{if } v_n < 0 \\ G_2 v_n & \text{if } v_n > 0 \end{cases} \quad (58)$$

One can find the simple plot of this characteristic in Figure 6; moreover, this component corresponds to the one-port shown in Figure 7. The diodes shown in the equivalent circuit of Figure 7 are ideal, i.e. they act as short circuits if the current is positive and as open circuits if the voltage is negative. We may observe that if  $G_1 = 0$  the characteristic reduces to the model of an ideal diode in series to a resistor  $1/G_2$ . The goal is the evaluation of the overall average electrical characteristic related to the three-dimensional network of Figure 8, made of the randomly interconnected non-linear asymmetric components of Figure 7.

A one-port can be associated with the whole network by means of two arbitrary electrodes parallel to the planes of the cubic lattice. Their potential difference is  $V$  and their associated current is  $I$ .

The analytical solution of the problem for a network  $M \times L \times Q$  is given by Equations (52) and (53) with  $N = 1$  and  $v_1 = 1$ . After some straightforward calculations we easily obtain, as

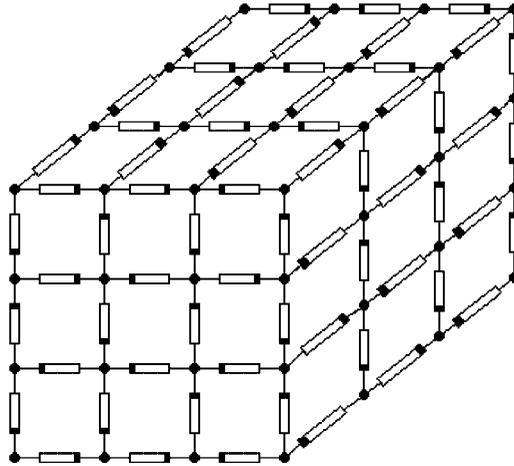


Figure 8. Schematic of the three-dimensional cubic network of identical asymmetric lumped elements described in Figures 6 and 7. As one can see the orientation of each element is a random variable.

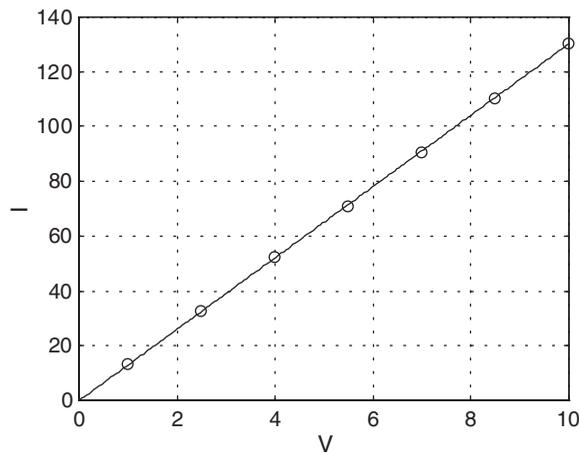


Figure 9. Example of computer simulation for the voltage ( $V$ )–current ( $I$ ) relationship (performed using the non-linear relaxation method) of the cubic network of Figure 8 where  $G_1 = 1$ ,  $G_2 = 4$ ,  $Q = 6$ ,  $M = 5$  and  $L = 5$  (continuous line: theoretical relationship given by Equation (59); circles: results of the Monte-Carlo simulation performed over 50 different realizations of the network).

result, a linear characteristic given by

$$I = \frac{(M + 1)(L + 1)}{Q} \left[ \frac{G_1 + G_2}{8} + \frac{1}{4} \sqrt{\left( \frac{G_1 + G_2}{2} \right)^2 + 8G_1G_2} \right] V \tag{59}$$

In Figure 9 one can find the results of computer simulations performed using the following values of the parameters:  $Q = 6$ ,  $M = 5$ ,  $L = 5$ ,  $G_1 = 1$ ,  $G_2 = 4$ . This result proves that the network performs as a linear constant conductance as expected by the heuristic theory.

Moreover, this result, if extrapolated to biological media, suggests a new hint for explaining why there is not any inconsistency between the typical electric characterization of biological tissues as almost linear macroscopic media, by means of their effective conductivity and permittivity, and the non-linearities of the biochemical processes occurring in the tissue cells [12,13]. In fact the non-linearities may be not observable by means of macroscopic electric measurement because of the randomised spatial orientation and location of the processes.

## 8. CONCLUSIONS

The present work describes the derivation of a heuristic theory, which characterizes linear and non-linear random mixtures and networks. Such a theory, both in linear and in non-linear case has been verified with a high satisfactory degree of accuracy by means of a series of computer simulations. All results are derived starting from a circuit-theoretic approach: in fact, the conclusions are based on the similarity between a random mixture and a proper electrical network. The generalization from the linear case to the non-linear one is obtained using the standard small signal technique, which leads to a general solution of the non-linear random network with symmetric and non symmetric lumped components. We wish to point out that most of the heuristic considerations described in the main text generates correct results (verified only by computer simulations) because of the randomness of the system and of the large dimensions of the network which smear out the refined effects of the non-linearities and of the complicated interconnections.

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## REFERENCES

1. Van Beek LKH. Dielectric behaviour of heterogeneous systems. *Progress in Dielectric* 1967; **7**:71–114.
2. Tareev B. *Physics of Dielectric Materials*. MIR Publishers: Moscow, 1979.
3. Dukhin SS. Dielectric properties of disperse systems. *Surface & Colloid Science* 1973; **3**:83–165.
4. Bianco B, Parodi M. A unifying approach for obtaining closed-form expressions of mixtures permittivity. *Journal of Electrostatics* 1984; **15**:183–195.
5. Maxwell JC. *A Treatise on Electricity and Magnetism*. Clarendon Press: Oxford, 1881.
6. Landau LD, Lifshitz EM. *Electrodynamics of Continuous Media*. Pergamon Press: London, 1960.
7. Bianco B, Degani G. Conductivity of mixtures: exact calculation in a particular case. *Alta Frequenza* 1981; **2L**:113–115.
8. Bianco B, Parodi M, Degani G. A circuit approach for deriving the electrical properties of statistical mixtures. *Alta Frequenza* 1982; **2LI**:80–85.
9. Belevitch V. *Classical Network Theory*. Holden-Day: San Francisco, 1967.
10. Forsythe GE, Wasov WR. *Finite-Difference Methods for Partial Differential Equations*. John Wiley & Sons: New York, 1967.
11. Abramowitz M, Stegun IA. *Handbook of Mathematical Functions*. Dover: New York, 1970.
12. Bianco B, Chiabrera A, Giordano S. D.C. Characterization of random mixtures of piecewise non-linear media. *Bioelectromagnetics* 2000; **21**:145–149.
13. Foster KR, Schwan HP. Dielectric permittivity and electrical conductivity of biological materials. In *Handbook of Biological Effects of Electromagnetic Fields*, Polk G, Postow E (eds). CRC Press: Boca Raton, 1986, p. 27–98.