

# The Frequency Dependent Permittivity and AC Conductivity of Random Electrical Networks

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*It has been shown that the universal dielectric response (UDR) of heterogeneous materials can be reproduced by electrical networks consisting of randomly positioned resistor and capacitors. The random network represents a microstructure that contains both insulating (the capacitor) and conductive regions (the resistor). This paper presents an investigation into the frequency dependent properties of large numbers of resistor-capacitor (R-C) networks. Parameters investigated include permittivity, conductivity and phase angle, with particular emphasis on observed power-law behavior and a comparison with previous work in this area.*

**Keywords** Dielectrics; permittivity; modelling; universal response

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## Introduction

Heterogeneous materials which contain both conductive and dielectric (insulating) phases are present in a wide variety of materials, including ceramics, polymers and composites. Many of these systems, including ferroelectric ceramics and polymers [2–4], display a very similar frequency dependent conductivity and permittivity, often termed Jonscher’s “Universal Dielectric Response” (UDR) [1]. An example of UDR behavior is shown in Fig. 1 which shows a doped yttria stabilized zirconia ionic conductor at a variety of temperatures. A typical UDR response is that at low frequencies the bulk ac conductivity is frequency independent,  $\sigma(\omega)$ , but at higher frequencies the ac conductivity increases, following power-law behaviour such that,

$$\sigma(\omega) \propto \omega^n \quad (1)$$

The overall response the type of data in Fig. 1 is often described by Eq. (2).

$$\sigma(\omega) = \sigma(0) + A\omega^n \quad (2)$$

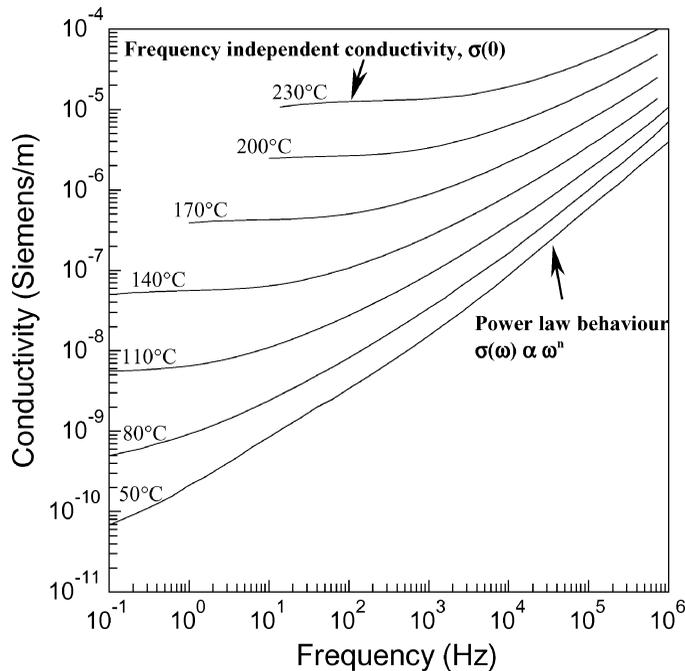
where  $\omega$  is angular frequency,  $0.6 < n < 1.0$  and  $A$  is a constant. The real part of the permittivity ( $\epsilon'$ ) can also be expressed by a power-law decay [5] such that,

$$\epsilon' \propto \omega^{n-1} \quad (3)$$

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**Figure 1.** Ac conductivity a doped zirconia at various temperatures.

The universality of the observed behavior is considered incompatible with conventional Debye behaviour, the distribution of relaxation times (DRT) and stretched exponential approach [6]. Many-body theories, fractal behaviour and hopping models have also been proposed [7, 8]. It is important that any proposed model for the UDR must also explain its commonality and for this reason random resistor-capacitor (R-C) networks have been recently examined by Almond et al. [9–11]. The R-C networks represent a microstructure that contains both dielectric (the capacitor) and conductive regions (the resistor). For the doped zirconia (Fig. 1), the disorder is on a small scale whereby mobile oxygen vacancies are distributed in the insulating zirconia host.

### Random Network Circuit Construction

Resistor-capacitor circuits for analysis of their electrical behaviour with frequency were constructed using a MATLAB script that allowed a large number (256) of circuits to be produced quickly and efficiently. The script had input variables for the circuit size (number of components along one length of the square), the values of capacitance and resistance of the components and the resistor/capacitor contents. In the networks presented in this paper the capacitors were 1nF and the resistors 1 k $\Omega$ , as in previous work on R-C networks [9, 10]. The MATLAB script constructed a network of components from its top left corner. All the vertical components were placed first with the horizontal elements added second. The components were placed between “nodes.” The nodes can be visualised as a square grid/lattice of dots. The script inserted capacitors or resistors randomly between these nodes with the ratio of resistors to capacitors being defined at the beginning of the construction.

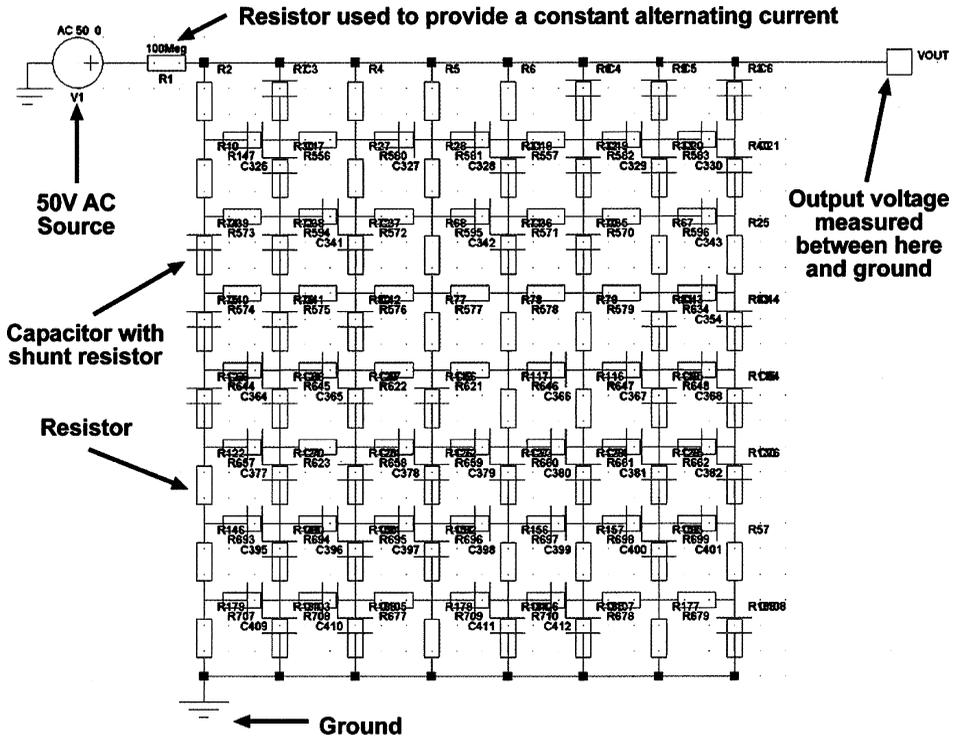


Figure 2. Example 8 × 8 resistor-capacitor network (64 components).

An example of an 8 × 8 random-resistor capacitor network is shown in Fig. 2, although the actual networks used this work are larger (16 × 16). Once the basic square circuit of the required composition and size has been formed the ancillary circuit components were added. These included the  $V_{OUT}$  connection to allow the electrical potential across the network to be measured and an external 50 V AC source which applied an alternating potential across the network. A 100 MΩ resistor was placed between the 50 V AC source and the network to maintain a constant current.

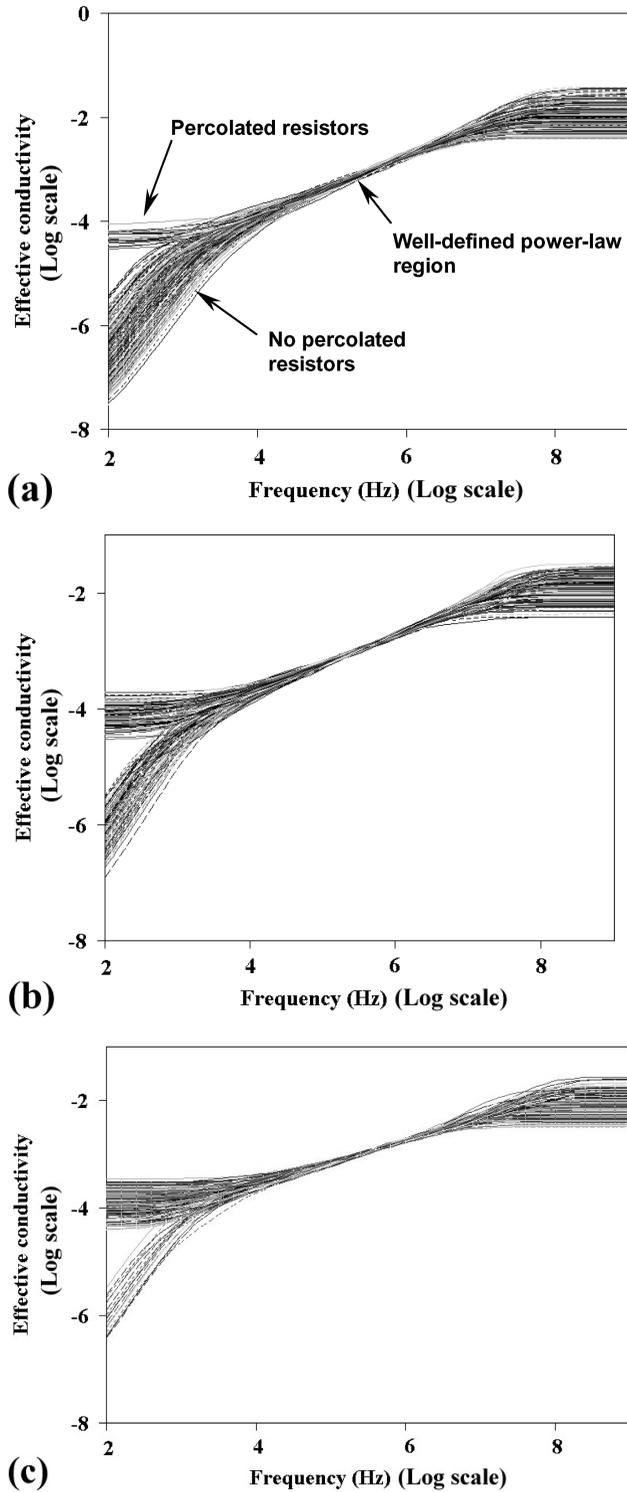
After the circuit has been constructed, SIMetrix software was used to solve the network simulation. The software used a simulation algorithm from SPICE (Simulation Program (with) Integrated Circuit Emphasis) version 3 f.5. The program simulates the behaviour of the circuit over the range of frequencies chosen for the analysis type (typically  $10^2$  to  $10^9$  Hz in this work).

### Typical response of R-C networks

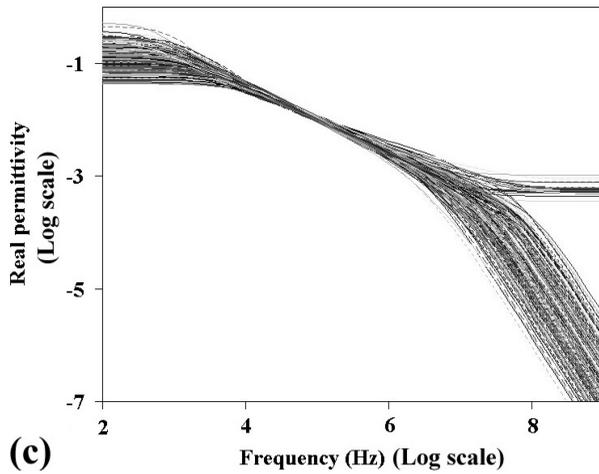
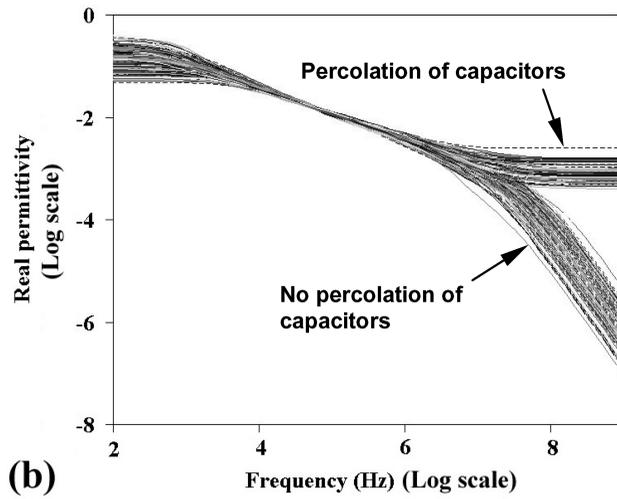
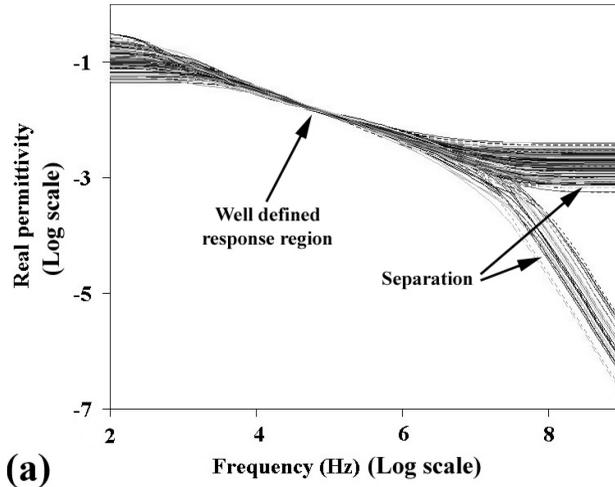
The following data are for three fixed compositions of resistors and capacitors, namely:

- (i) 40%R:60%C (percolated capacitors across the network are likely)
- (ii) 50%R:50%C
- (iii) 60%R:40%C (percolated resistors across the network are likely)

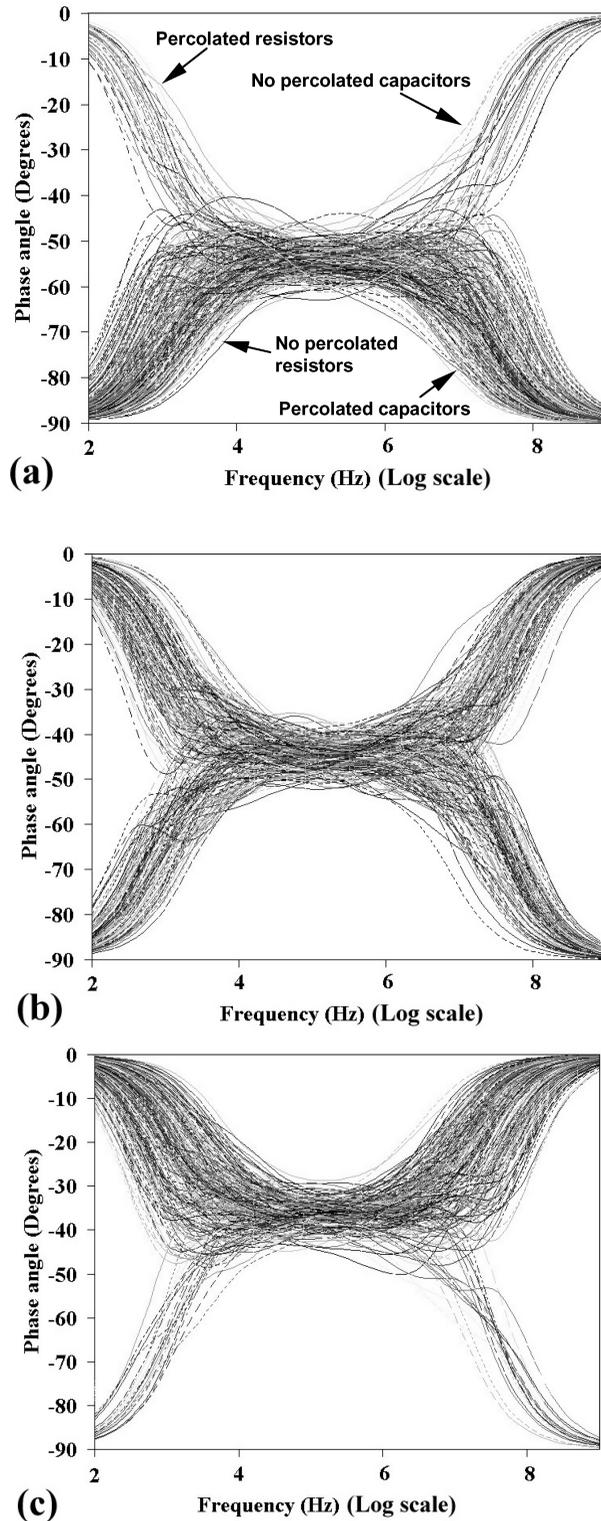
The frequency dependent effective permittivity (network capacitance), ac conductivity and phase angle of the networks as a function of R:C ratio are presented in Figs. 3–5. These



**Figure 3.** Network conductivity as a function of frequency (a) 40%R:60%C (b) 50%R:50%C (c) 60%R:40%C.



**Figure 4.** Network capacitance as a function of frequency (a) 40%R:60%C (b) 50%R:50%C (c) 60%R:40%C.



**Figure 5.** Network phase angle as a function of frequency (a) 40%R:60%C (b) 50%R:50%C (c) 60%R:40%C.

figures have been generated from data for 256 individual network simulations for each composition with a side length of 16 components (256 components in total). The variability in responses observed in Figs. 3–5 for networks of identical composition are due to the random configuration of individual networks.

### AC Conductivity of the Networks

Figure 3a–c shows the ac conductivity frequency dependence for networks of the different resistor-capacitor contents examined. To understand the patterns emerging from these figures, it is important to note that while the resistor conductivity ( $R^{-1}$ ) is frequency independent, the ac conductivity of the capacitors (the admittance,  $\omega C$ ) increases linearly with frequency. Therefore, at low frequencies the conductivity of the resistors is large compared to the capacitors ( $R^{-1} \gg \omega C$ ) and they can be regarded as short circuits [8]. At high frequencies the ac conductivity of capacitors is much larger than the resistors ( $R^{-1} \ll \omega C$ ) and the resistors can be regarded as open circuits [9]. We will now consider the response of the 60%C:40%R networks in Fig. 3a, where the capacitors are in the majority and their percolation across the network is likely. At low frequencies ( $<10^3$  Hz) a small number of the 256 networks exhibit a frequency independent plateau in conductivity (as is observed experimentally in Fig. 1). This is a result of a percolated path of resistors across the network. At these low frequencies, the ac conductivity of the capacitors is sufficiently low that current flows through the resistor percolation path ( $R^{-1} \gg \omega C$ ). A larger number of the networks exhibit a rapidly decreasing conductivity at frequencies below ( $10^3$  Hz), due to a lack of a percolated resistor path. The network conductivity is dominated by the very low ac conductivity of the capacitors in this frequency range, which continually decreases with frequency. Since the networks in Fig. 3a have a high fraction of capacitors (60%), it is not surprising that this behaviour is observed more often (compared to the plateau in conductivity).

As the frequency increases ( $>10^4$  Hz) the network response converges to a well-defined region. In this region the ac conductivity (admittance) of the capacitors is similar to that of the resistors ( $R^{-1} \sim \omega C$ ) and the current flows through both the resistors and capacitors (the condition  $R^{-1} = \omega C$  is met at  $10^{5.2}$  Hz). Since current flows through all the components, the configuration of the resistors and capacitors in individual networks is less important, resulting in a network independent response [9]. The network conductivity begins to increase linearly with frequency region (power law behaviour), due to the increasing conductivity of the capacitors. The sharply defined power law response is considered an emergent property of the random networks and Bounmrane [10] proposed the term ‘emergent scaling’ to describe this behaviour. This pattern of a low frequency plateau followed by a power law region compare qualitatively with the experimental data in Fig. 1.

At high frequencies ( $>10^7$  Hz), the ac conductivity of the capacitors is much higher than that of the resistors ( $R^{-1} \ll \omega C$ ). The network conductivity returns to a frequency independent behaviour, determined by the resistors connected via the high conductivity capacitors in the network. Since the low and high frequency response is determined by the random resistor distribution in each network, there is significant variation in the behaviour of each network. If a percolated path of capacitors across the network is present, the conductivity of the network should continue to increase with frequency. The lack of this behaviour at high frequencies may be due to the configuration of the model.

Figure 3b–c show similar network results for increasing resistor contents. With a 50%R:50%C composition (Fig. 3b), approximately half the networks exhibit the low

frequency plateau, while for the 60%R:40%C mixture (Fig. 3c) most networks exhibit the plateau as a result a percolated path of resistors across the networks. The well defined power-law region is observed for all of the networks examined at the three different compositions. The gradient of the power law region decreases with decreasing capacitor content and will be considered more quantitatively later.

### Permittivity (Normalised Network Capacitance)-Frequency Dependence

At low frequencies the relative permittivity of the network is unaffected until a frequency of  $10^3$  Hz is reached (Fig. 4). Again, in this low frequency region the permittivity is determined by the ac conduction path which travels preferentially through the resistors which may be percolated across the network or linked by a small number of low admittance capacitors. At high frequencies, current passes preferentially through the capacitors in series, resulting in a lower network permittivity compared to the low frequency value. Separation of the response at high frequencies ( $>10^7$  Hz) is observed between different networks of the same composition at high frequency due to the fact that some networks contain percolated capacitors (a plateau in permittivity) and no percolated capacitors (relative permittivity continues decreasing with frequency). Although significant difference between different random networks is observed at low and high frequencies the well defined, network independent, power law response is again observed at intermediate frequencies  $10^4$ – $10^6$  Hz. The power law decay in permittivity with frequency is also widely observed in many materials, as in Eq. (2).

### Phase Angle Frequency Dependence

The phase angle frequency dependences for the different network combinations (Fig. 5) follows a similar behaviour based on the previous discussions. For the 60%C:40%R composition (Fig. 5a) a small number of networks (those which exhibit a low frequency conductivity plateau in Fig. 3a) have a percolated path of resistors across the network. Resistor percolation results in a phase angle of  $0^\circ$  (purely resistive behaviour) at low frequencies. Most of the networks in Fig. 5a, however, have no percolation of resistors and at low frequencies the phase of the network is dominated by the low admittance capacitors, resulting in a phase angle of  $90^\circ$  (purely capacitive). At high frequencies, percolation paths of capacitors across the network dominate and most networks exhibit a phase angle of  $90^\circ$ . A small number of networks do not have percolated capacitors and at high frequency the phase is  $0^\circ$  since it is dominated by the relatively high impedance of the resistors.

At intermediate frequencies ( $10^4$ – $10^7$  Hz) the phase angle is relatively constant. Comparing Figs. 5a–c, the constant phase angle in this frequency ranges decreases as a result of an increase in the fraction of resistors in the network [9].

### Analysis of Power-Law Behaviour

The networks examined here show some qualitative similarity with the UDR of many materials, such as the conductivity frequency dependence of the zirconia in Fig. 1. It has been reported [9, 10] that for random R–C networks the power law exponent,  $n$ , of the relative permittivity and conductivity response is dependent on the network composition and that  $n$  in Eqs. (1–3) is related to the fraction of capacitors in the network.

**Table 1**  
Fraction of Capacitors and Power Law fit for the  
Three Different Network Compositions

Network type (%R:%C)	Power law fit, n
60:40	0.399
50:50	0.487
40:60	0.594

The suggested format for this relationship is that given in Eq. (4), where  $\sigma^*$  is the complex conductivity of the network and  $\omega$  the angular frequency ( $2\pi f$ ) of the stimulating field.

$$\sigma^*(\omega) \propto (i\omega)^n \quad (4)$$

To examine the relationship between the fraction of capacitors in the network and the power law behaviour observed in the network response, a trend line was fitted to the conductivity data of Fig. 3 in the well defined frequency range  $10^3$ – $10^7$  Hz. The gradient, n, of the power law fit to the conductivity-frequency data are compared against network composition in Table 1, with good agreement.

## Conclusions

Large numbers of two-dimensional networks of random resistors and capacitors have been successfully constructed and the frequency response of permittivity, conductivity and phase angle simulated. The simulated results of the frequency response of permittivity, conductivity and phase angle and are in agreement with recently published work on R-C networks. The conductivity and permittivity frequency dependence is similar to the UDR behaviour observed in a wide range on materials. At low ( $R^{-1} \gg \omega C$ ) and high ( $R^{-1} \ll \omega C$ ) frequencies, significant variation in network behaviour is observed between networks of the same composition. This is due to the variation in the percolation paths of resistors and capacitors in individual networks. The results indicate an intermediate frequency regime ( $R^{-1} \sim \omega C$ ) of well-defined power law behaviour, ‘emergent scaling’ [10]. In this frequency range, since both the resistors and capacitors contribute to the overall conductivity and permittivity, random networks construction of the same composition behave almost identically. The power-law exponent, n, is related to the fraction of capacitors in the model implying a similar relationship in the universal dielectric response of real materials, which consist of conductive and insulating regions in its macrostructure, microstructure or nanostructure. Experimental evidence of this relationship has recently been reported [12]. An understanding of these networks can provide an understanding of the role of microstructure in determining the frequency dependence of permittivity and conductivity of heterogeneous materials, including ferroelectric ceramics, polymers and composites.

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