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## Dielectric constant of quasi-critical percolation systems

V A Garanov†, A A Kalachev†, A M Karimov†, A N Lagarkov†,  
S M Matitsin†, A B Pakhomov‡, B P Peregood‡§, A K Sarychev†,  
A P Vinogradov† and A M Virnik†

† Institute of High Temperatures, USSR Academy of Sciences, Izhorskaya 13/19,  
Moscow 127412, USSR

‡ A F Ioffe Physico-Technical Institute, USSR Academy of Sciences, Polytechnicheskaya  
26, Leningrad 194021, USSR

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**Abstract.** A way of obtaining binary composite materials having some properties of the critical percolation system is considered. These materials can be obtained by high-density current action on an initially well conducting mixture. This action destroys part of the contacts between adjacent conducting granules, bringing the system to the quasi-critical state. It is shown that quasi-critical systems may have high values of dielectric constant  $\epsilon \approx 10^2$ – $10^6$  in the frequency range  $0 < f < 10^{10}$  Hz. The formation and dielectric properties of the systems are studied both by computer simulation and experimentally.

### 1. Introduction

The dielectric properties of disordered binary mixtures of conducting and non-conducting granules have been extensively studied (Shklovskii and Efros 1976, Grannan *et al* 1981, Vinogradov *et al* 1984, Zeng *et al* 1989, and references therein). The current interest in these systems is partly due to the prospect of using them in manufacturing new dielectric materials. A distinguishing feature of these systems is the existence of a percolation threshold  $P_c$ . The composite material can only possess bulk conductivity if the volume concentration  $P$  of a conducting component exceeds the percolation threshold  $P_c$ .

The mixture with the concentration  $P$  close to the percolation threshold  $P_c$  (hereafter referred to as a critical percolation system) has a large dielectric constant  $\epsilon_{\text{eff}}$ . In the infinite system the calculated value of  $\epsilon_{\text{eff}}$  tends to infinity as the concentration tends to the threshold. It is known that  $\epsilon_{\text{eff}}$  depends on frequency in a wide frequency range (Shklovskii and Efros 1976, Vinogradov *et al* 1984, 1988, Zeng *et al* 1989, and references therein).

For convenience, we shall here briefly summarize some properties of percolation systems. In the vicinity of the percolation threshold the effective dielectric constant  $\epsilon_{\text{eff}}$  and the bulk conductivity  $\sigma_{\text{eff}}$  of a percolation system obey the power laws (Shklovskii

§ Deceased.

and Efros 1976, Grannan *et al* 1981, Vinogradov *et al* 1984, 1988)

$$\begin{aligned} \varepsilon_{\text{eff}}(\tau) &\sim |\tau|^{-q} \\ \sigma_{\text{eff}}(\tau) &\sim \begin{cases} \tau^t & \tau > 0 \\ 0 & \tau < 0 \end{cases} \end{aligned} \quad (1)$$

where  $q$  and  $t$  are the critical indices, and  $\tau = (P - P_c)/P_c$ .

In the systems with  $P > P_c$  an infinite cluster of conducting particles is formed. The mean size of the infinite cluster's inhomogeneity, known as the correlation length, diverges near the threshold (Shklovskii and Efros 1984, Stauffer 1985), so that  $\xi \sim |\tau|^{-\nu}$ , where  $\nu = 0.88$  (Stauffer 1985).

According to the theory (see, e.g. Stauffer 1985), the effective values  $\varepsilon_{\text{eff}}$  and  $\sigma_{\text{eff}}$  for a finite critical system depend on its size  $L$ , for  $L < \xi$ :

$$\begin{aligned} \varepsilon_{\text{eff}}(L) &\sim (L/a_0)^{q/\nu} \\ \sigma_{\text{eff}}(L) &\sim (L/a_0)^{-t/\nu} \end{aligned} \quad (2)$$

where  $q/\nu = 0.85$  (Herrmann *et al* 1984) and  $a_0$  is the granule diameter. The fluctuations of  $\sigma_{\text{eff}}$  and  $\varepsilon_{\text{eff}}$  are of the order of their mean values.

The manufacturing of critical percolation systems with properties (1) have been described, for example, by Grannan *et al* (1981), Nicklasson and Grangvist (1984) and Vinogradov *et al* (1984). However, as far as we know, mixtures possessing properties (2) have not been obtained as yet. The point is that the inevitable technological fluctuations of concentration are much larger than statistical fluctuations, whatever method of mixing and compaction is used. An example is a cubic sample with an edge length  $L = 1$  cm consisting of granules of size  $a_0 = 1 \mu\text{m}$ . Typical relative values of statistical spatial fluctuations of concentration in these systems are of the order of  $N^{-1/2} \sim (L/a_0)^{-3/2} \approx 10^{-6}$ . Obviously, this homogeneity is not likely to be achieved in real mixtures prepared by familiar technologies (Grannan *et al* 1981, Nicklasson and Grangvist 1984, Vinogradov *et al* 1984). Consequently, the critical concentration  $P_c$  cannot be reached simultaneously in different regions of the sample†.

Thus, if we consider any sample with concentration  $P$  slightly lower than the percolation threshold, there would exist a set of separate regions with high values of  $\varepsilon(r)$ .

This fact, however, does not lead to a significant increase in the effective dielectric constant  $\varepsilon_{\text{eff}}$  of the sample, since  $\varepsilon_{\text{eff}}$  is not equal to the volume average value  $\langle \varepsilon(r) \rangle$ .

That is why the manufacture of systems with a high dielectric constant and experimental verification of relationships (2) appear to be difficult.

Recently the modification of random-fuse networks by an electric current has been discussed in the literature for  $d = 2$  (de Arcangelis *et al* 1985, 1989, Duxbury and Leath 1987, Duxbury *et al* 1987, Kahng *et al* 1988, de Arcangelis and Herrmann 1989) and for  $d = 3$  (Vinogradov *et al* 1989b). The dynamics of the breakdown process and the value of the breakdown voltage have been shown to depend upon the system size and the type of disorder in the system. In the thermodynamic limit the breakdown strength of the network goes to zero. It seems that the resulting structure should show a critical behaviour of the dielectric constant.

Our investigations show that the modified systems (hereafter referred to as quasi-critical percolation systems) may have high values of dielectric constant  $\varepsilon_{\text{eff}} \approx 10^2\text{--}10^6$  in

† Similar troubles, for example, arise in the investigation of critical phenomena in the vicinity of the vapour-liquid critical point (Anisimov 1987)

the frequency range  $0 \text{ Hz} < f < 10^{10} \text{ Hz}$ . We have obtained such systems experimentally and also simulated them on a computer.

## 2. Quasi-critical percolation systems

In this paper some results of a theoretical and experimental investigation concerning the possibility of modifying percolation systems with initial metal concentration  $P$  significantly higher than the percolation threshold  $P_c$  are presented. We have tried to obtain a composite material with properties (1) and (2), but with concentration  $P > P_c$ .

Following de Arcangelis *et al* (1985), Duxbury and Leath (1987) and Duxbury *et al* (1987), we consider a disordered mixture of metal and insulator granules with metal concentration  $P > P_c$  placed in an electric field. When a voltage is applied to the sample, the electric current will flow through the metal granules forming percolation channels. If a current flowing through any conducting bond (an electrical contact between two adjacent metal granules) belonging to the backbone is large enough, this bond will be broken. For example, this may melt and be destroyed because magnetohydrodynamical instability of the conductor shape develops in the magnetic field of the current (Abramova *et al* 1975). Finally, in the case of a current density  $J \geq 5 \times 10^7 \text{ A cm}^{-2}$ , material evaporation will most rapidly lead to bond breaking (Kolgatin *et al* 1989). In any case, for a given voltage pulse duration there is a value of the voltage which causes breakdown in the sample conductivity as a result of successive destruction of a number of bonds belonging to the backbone.

In this work we attempt to study the dielectric properties of the network modified by the electric current after it has lost its conductance.

Let us consider the AC properties of the system. Bonds with maximum current values are broken first of all. Their breaking gives a maximally large value of the displacement current within the appearing gaps. Therefore, increasing the voltage slowly makes it possible to break all the percolation channels gradually and to obtain a system with a high value of  $\varepsilon_{\text{eff}}$  which satisfies the condition (2), as will be shown below.

## 3. Numerical simulation of quasi-critical systems

Quasi-critical systems were studied by computer experiment with  $L \times L \times L$  cubic fuse networks, where  $L$  is the number of bonds along the cube edge. The  $1 - P$  portion of resistors were randomly broken. The voltage was applied to two opposite faces while periodic boundary conditions were imposed on the others. Then Kirchhoff's equations were solved by the conventional technique (Vinogradov *et al* 1984, 1989b) and the current values in all the bonds were determined. The bonds with a current above the critical value  $I_{\text{cr}}$  were broken, upon which calculations were repeated with the former voltage. If the current within any bond exceeded  $I_{\text{cr}}$ , this bond was broken too and calculations were repeated again. When the largest current  $I_{\text{max}}$  decreased below  $I_{\text{cr}}$ , the exterior voltage was multiplied by a factor  $I_{\text{cr}}/I_{\text{max}}$ . This caused destruction of a new portion of bonds. The described computational procedure of stepping up the voltage was carried out until all the conducting circuits in the backbone were broken (see Vinogradov *et al* (1989b) for details). So this procedure slightly differs from that used by Duxbury *et al* (1987) and de Arcangelis *et al* (1985). Artificially decreasing the external potential is impossible in our algorithm. We think that the procedure reflects the experimental situation more adequately.

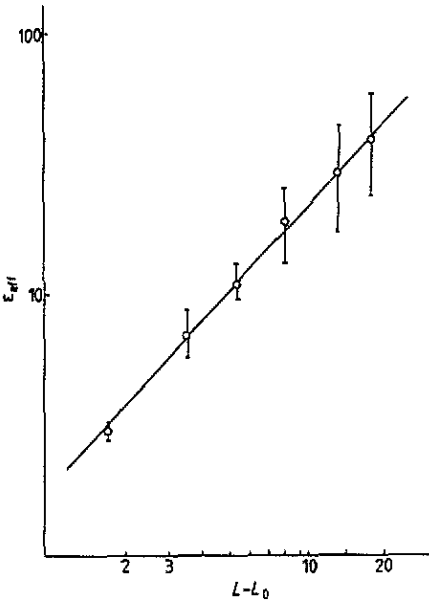


Figure 1. Size dependence of dielectric constant of a modified system (Monte Carlo simulation results).

After the rupture of all the percolation channels the effective dielectric constant of the obtained system was calculated. Without percolation channels, the system capacity  $C_{\text{eff}}$  is proportional to the conductance  $\Sigma_{\text{eff}}$  of the conjugate system built from the initial system by replacing the conductors with ideal conductors and by placing the resistors of finite resistance value  $R_{\text{fin}}$  in the dielectric gaps:

$$C_{\text{eff}}/\epsilon_d = \Sigma_{\text{eff}}R_{\text{fin}}$$

where  $\epsilon_d$  is the dielectric constant of the dielectric material present in the mixture. The method of calculating  $\Sigma_{\text{eff}}$  is well known (see, e.g., Vinogradov *et al* 1984). The value of  $\epsilon_{\text{eff}}$  is equal to  $C_{\text{eff}}/L$ .

The numerical results displayed in figure 1 demonstrate an increase in  $\epsilon_{\text{eff}}$  with increasing system size. The open circles represent the value of  $\epsilon_{\text{eff}}$  averaged over the number  $N(L)$  of different realizations of the system obtained in computer simulation for each  $L$ . The numbers  $N(L)$  for  $L = 3, 5, 7, 10, 15, 20$  equal 98, 237, 403, 82, 39 and 54, respectively. The bars shown contain twice the standard deviations of the mean value of  $\epsilon_{\text{eff}}(L)$ .

Following the speculation by Duxbury *et al* (1987), one can easily imagine the dynamics of the process in the limiting case when the initial concentration  $P$  is close to unity. In this case, only a few insulating bonds are present in the conductive medium. If the voltage applied to the system is large enough, bonds will be destroyed in the regions of high current density around the insulating bonds. Successive breaking of bonds around non-conducting granules causes expansion of a crack in the plane perpendicular to the external field. When this expanding interlayer reaches the boundaries of the system, the system splits into two parts and its conductivity in the direction of the field vanishes as argued by Duxbury and Leath (1987). If we imagine a conducting cube with an insulating interlayer interposed between the capacitor plates, it will be easy to show that its effective dielectric constant is directly proportional to the size  $L$ :

$$\epsilon_{\text{eff}}(L) \sim (L/a_0)^\gamma \quad \text{with } \gamma \approx 1. \quad (3)$$

At the percolation threshold (where  $L \ll \xi = \infty$ ) all the 'red' bonds in the sample (Skal and Shklovskii 1974, de Gennes 1976, Stanley 1977, Pike and Stanley 1981) should break simultaneously when the voltage reaches its critical value and the backbone splits into a set of conducting clusters. Using the work by Sarychev *et al* (1987) one may obtain for the number of the clusters an estimate  $k \propto L^{1/\nu}$ . In the case when the concentration  $p$  differs from both unity and  $p_c$  and when  $L > \xi$ , the whole sample may be considered as a superlattice of blobs with unit-cell size of  $\xi$ . This superlattice may be regarded as a system of random fuses in which conductivities as well as failure-inducing voltage drops are continuously distributed. Both ductile and brittle regimes of fracture may be observed in a system of this kind depending on the size  $L$  (Kahng *et al* 1988). The ductile damage is observed at low  $L$ , and at  $L \rightarrow \infty$  the brittle fracture takes place. In the intermediate case, fracture is a two-stage process. Accumulation of non-localized damage that indicates a ductile regime is the essence of the first stage. At the second stage, only one brittle crack grows in the superlattice (Vinogradov *et al* 1989a, b). The resulting crack is a fractal specified by the fractal dimension  $f > 2$  and the mean width  $\xi$ . The volume of the crack is filled with the percolative medium with dielectric constant  $\epsilon_{\text{eff}} = \epsilon(\tau) + \epsilon_b$ , where the additional term  $\epsilon_b$  is the contribution due to the creation of new dielectric gaps in place of the conducting bonds which have been broken. So the dielectric constant of the sample can be estimated as

$$\epsilon_{\text{eff}} \sim [\epsilon(\tau) + \epsilon_b] L^{f-1} / \xi(\tau). \quad (4)$$

The actual structure of the crack and the resulting number of finite clusters outside the crack are still in question.

Our calculations show (see figure 1) that, for  $p = 0.41$ ,

$$\epsilon_{\text{eff}}(L) \sim (L - L_0)^\gamma \quad (5)$$

where  $L_0 = 1.36$ ,  $\gamma = 1.08 \pm 0.26$ , the value of  $\gamma$  is in agreement with our speculation above†. The power law behaviour of  $\epsilon_{\text{eff}}(L)$  obtained is similar to the relationship (2). In this sense the systems are quasi-critical. The values of  $\epsilon_{\text{eff}}$  for a given  $L$  are three to four times larger than that reported by Vinogradov *et al* (1984) for the critical value. Two reasons may be mentioned. First, the metal concentration  $P$  is sufficiently high compared with  $P_c$ . One should have in mind that the action of the electric current destroys no more than 1–2% of conducting bonds; thus there is more metal in the modified system than in the critical system. Secondly, there is a certain 'trend' of modification. As has been mentioned above, the bonds in the system which are destroyed preferentially are those which, if absent, provide a maximum increase in  $\epsilon_{\text{eff}}$ .

Figure 2 depicts the frequency dependence of the effective dielectric constant for a particular system given by numerical experiments ( $L = 15$ ). The computational algorithm used cannot give the lower edge of the dispersion region. It may only be asserted that frequency dependence of  $\epsilon_{\text{eff}}$  is essential for  $f \geq 10^{-2} \sigma_M$ , where  $\sigma_M$  is the conductivity of the metal phase of the mixture. This dependence can be approximated by the function  $\epsilon \sim f^{-1/3}$ , which is valid for critical percolation systems (Shklovskii and Efros 1976,

† We should like to mention that the conditions for single bond destruction in our experiments are somewhat different from the model involved in the computer simulations. In order to fit the experimental situation more correctly, one should take into account the possibility of dielectric breakdown of the gaps that the broken bonds are substituted for. In this case the electric current in the sample is not interrupted after the creation of the crack. The process of sample modification may then be imagined as a sequence of crack creations. As a result the whole body of the sample must be filled with a medium having the properties of the medium inside the crack.

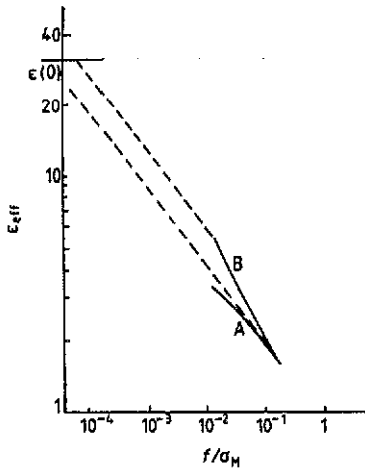


Figure 2. Calculated dispersion curves for a particular initial system (curve A) and for a relevant quasi-critical percolation system (curve B) ( $P_0 = 0.41$ ;  $L = 15$ ). ---, possible extrapolations.

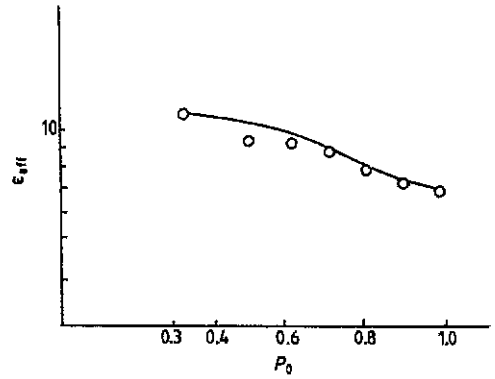


Figure 3. Calculated values of dielectric constant of modified system versus initial conducting-bond concentration ( $L = 7$ ).

Vinogradov *et al* 1984, Zeng *et al* 1989). Using this relationship to extrapolate curve B (figure 2) into the lower-frequency region, we can determine the dispersion range edge value  $f_{\text{edge}}$  to be of the order of  $10^{-5} \sigma_M$ . Near this frequency the function  $\epsilon(f)$  intersects the level of the static dielectric constant value.

Thus, numerical simulations have shown that it is possible to modify the percolation system under the action of an external field so as to impart to it some typical properties of the critical system. These properties are a high value of the dielectric constant  $\epsilon_{\text{eff}}$ , abnormal dispersion  $\epsilon_{\text{eff}}(f)$  and power-law size dependence  $\epsilon_{\text{eff}}(L)$ . All of these are evidence that the systems obtained are quasi-critical.

We wish to emphasize once more that, although percolation channels are absent, the concentration of conducting (metal) particles in a quasi-critical percolation system may substantially exceed the ordinary value of the percolation threshold  $P_c$ . One may also say that external action (electric field) moves the percolation threshold from  $P_c$  to  $P$ . The  $\epsilon_{\text{eff}}$ -value of a quasi-critical percolation system slightly depends on the value  $P - P_c$  (figure 3), which is why the relation (1) cannot in this case be written down.

#### 4. Experimental results

Inspired by the results of computer simulation we proceed to carry out experiments. Conducting samples of the composite material were exposed to electric current pulses of different amplitudes; changes in the backbone structure were controlled by measuring the resistance. The dielectric constants of both the initial samples and the samples obtained after the electric discharge were measured.

Composite material samples were manufactured by a plasma-spraying technique (Virnik *et al* 1989). The aluminium and corundum powders of particle size about  $100 \mu\text{m}$  were conveyed to the plasma torch, melted there and deposited onto an insulator substrate. The fusion of adjacent metal particles meant that the conductivity of contacts

was of the order of conductivity of metal inclusions. Thus we can suppose that the composite material obtained imitates that studied in the computer experiments.

To obtain systems exhibiting a quasi-critical behaviour, we used samples possessing bulk conductivity. The metal concentration  $P_0$  in these samples was higher than the percolation threshold ( $P_0 \approx 17$  vol.%, while the value of the percolation threshold  $P_c$  equalled 14%, and the initial mean resistivity was about  $(1-2) \times 10^{-3} \Omega \text{ cm}$ ). The samples were exposed to single high-power electric current discharges generated by a capacitor bank with a capacitance of  $1.25 \mu\text{F}$  and an inductance in the discharge circuit of  $0.7 \mu\text{H}$ . The charging voltage was varied from 10 to 50 kV, the discharge current amplitude at maximum charging voltage being about 60 kA. A typical current oscillogram is shown in figure 4. Samples were squeezed between two isolating plates to avoid destruction by electrodynamic forces. A longitudinal voltage was applied to the opposite sample edges through retained soft foil contacts, as shown in figure 5. One can estimate the mean current density in a percolation channel at a given current amplitude, starting from the measured value of conductivity. For  $I = 50$  kA the mean value  $J_{\text{mean}}$  was about  $10^7-10^8 \text{ A cm}^{-2}$ . It has already been demonstrated (Kolgatin *et al* 1989) that, at this current density, explosive evaporation of metal is more rapid than the development of magnetohydrodynamic instability of a conductor in the magnetic field of the current. Thus we expect evaporation of contacts to be the main mechanism of bond destruction in our case.

So, the current causes multiple destruction of the conducting contacts. This process is analogous to the volume destruction of metal compacted samples under the action of a high-density current (Vinogradov *et al* 1989a). However, the current flow was not even interrupted during the discharge when all the conducting channels were broken. We attribute this to the electric arc formed between the gap edges (Abramova *et al* 1975, Kolgatin *et al* 1989). After the electric current pulse has finished, the sample has a new conductance value lower than that of the initial state. The residual resistance tends to infinity if the electric pulse amplitude exceeds 40 kA.

The initial values for the dielectric constants of the samples were measured using a specially developed resonator technique applicable to materials with high values of loss tangent. The measured initial value was  $\epsilon' \approx 2$ ,  $\epsilon'' \approx (2-3) \times 10^{3\ddagger}$  in the microwave range for the electric field vector parallel to the sample plane.

In agreement with numerical results, the samples acquire higher dielectric constant values after the current action. These were measured by means of the contactless technique (Kalachev *et al* 1987, 1989) in a frequency range from  $4 \times 10^9$  to  $1 \times 10^{10}$  Hz $\ddagger$ . The lower-frequency edge was defined in the experiment by the relation between the wavelength and sample dimensions. Measurements at frequencies  $f > 10^{10}$  Hz were limited by the low transparency of the samples (Kalachev *et al* 1987, 1989).

The dielectric constants are plotted in figure 6 for five samples. The value for the real part of microwave dielectric constant varied from 200 to 400 in these samples (it should

$\ddagger$  If the initial static conductivity measured is  $\sigma \approx 10^{15} \text{ s}^{-1}$ , the imaginary part of the dielectric constant can for  $f \approx 10^{10}$  be evaluated as  $\epsilon'' \sim 2\sigma/f = 10^5$ . Discrepancy between this estimate and experimental data may be associated with the skin effect at a given frequency (see Vinogradov *et al* (1989c) for more details).

$\ddagger$  The value  $f = 10^{10}$  Hz is approximately equal to the estimate for the dispersion region edge  $f_{\text{edge}} \approx 10^{-5} \sigma_M \approx 10^{10}$  Hz, given by extrapolation of computational data. We have used the relationship  $\epsilon(f) \sim f^{-1/3}$  for this extrapolation (see figure 2). The value of conductivity  $\tilde{\sigma}_M \sim \sigma_M(\delta/a_0) \sim \sigma_M 10^{-2} \approx 10^{15} \text{ s}^{-1}$  renormalized with respect to the skin effect (Vinogradov *et al* 1989c) is used in the estimate instead of  $\sigma_M$  (here  $\delta/a_0$  is the ratio of the microwave field penetration depth to the granule size).



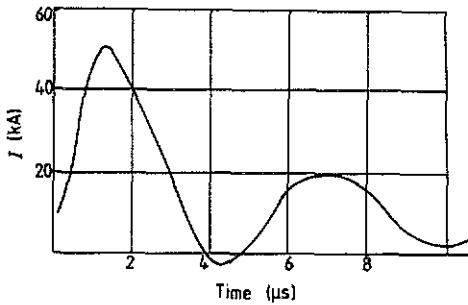


Figure 4. A typical current oscillogram.

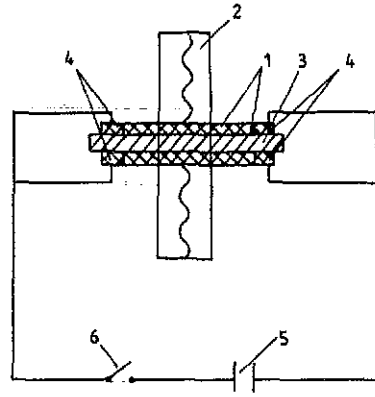


Figure 5. A schematic diagram of the installation for sample breakdown: 1, 2, dielectric cassette; 3, sample; 4, retained soft foil contact; 5, capacitor bank; 6, commutator.

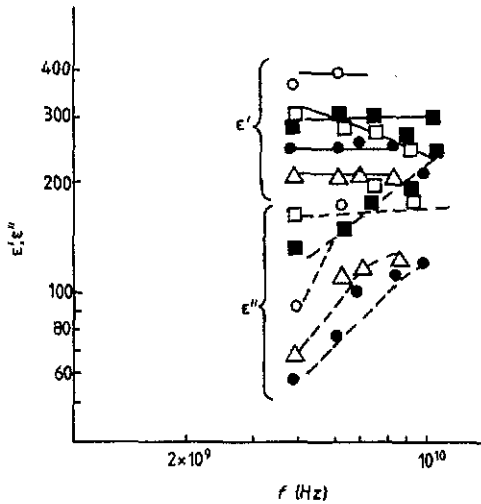


Figure 6. Measured dispersion of real and imaginary parts of dielectric constant for five samples:  $\square$ , sample 1;  $\circ$ , sample 2;  $\blacksquare$ , sample 3;  $\bullet$ , sample 4;  $\triangle$ , sample 5.

be recalled that in the initial state this value was not greater than 2). This value may be compared with those for ferroelectrics.

It can be seen in figure 6 that the real part of dielectric constant  $\epsilon'$  of samples 2–5 is constant in all the measurements while the imaginary part  $\epsilon''$  increases with increasing frequency. This behaviour of  $\epsilon'(f)$  allows us to suggest that the measured values of  $\epsilon'$  are equal to the static values. In a similar way to percolation systems (Shklovskii and Efros 1976), we can expect that the condition  $\epsilon' \approx \epsilon''$  should be valid in the dispersion range. It means that significant changes of  $\epsilon'$  for samples 2–5 would take place at  $f \geq 10^{10}$  Hz while, for sample 1, dispersion of  $\epsilon'$  is observed and  $\epsilon''$  reaches its maximum value for  $f \leq 10^{10}$  Hz.

The size dependence of the physical properties have not been experimentally studied in this work.

However, the high dielectric constant values achieved and the frequency dispersion of  $\epsilon_{\text{eff}}$  are good reasons to believe that the experimental composite material is in the

quasi-critical state defined and described above. If the theory is correct, dielectric constant values up to  $10^6$  can be obtained for quasi-critical systems using granules of smaller size and the above method of producing quasi-critical systems.

## 5. Conclusion

In this paper the dielectric properties of quasi-critical percolation systems are considered for the first time. The quasi-critical state is achieved by opening a small portion of conducting bonds belonging to the backbone and an initially conducting mixture. It has been shown that dispersion in these systems resembles that in critical percolation systems with a dispersion range extended into the region of much lower frequencies.

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