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RESEARCH PAPER

MICROSCOPIC MODEL OF DIELECTRIC α-RELAXATION IN DISORDERED MEDIA

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Abstract

The micro/mesoscopic theory of dielectric relaxation has been developed. Based on the fractional kinetics it gives a possibility to obtain the desired expression for the complex dielectric permittivity (CDP) and describe the asymmetric peaks that are created presumably by the so-called "excess wing" located in high-frequency region. The well-known empirical Cole-Davidson expression and its generalization for the CDP were obtained from this theory. This theory is based on self-similar phenomenon and multi-channel organization of relaxation process in disordered dielectrics. The relaxation parameters are connected with the structural parameters of the medium considered.

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Key Words and Phrases: Cole-Cole expression; Cole-Davidson expression; dielectric permittivity; fractals; fractal kinetics; excess wing

1. Introduction

One of the most striking features of the dielectric relaxation phenomenon existing in disordered materials such as complex liquids and amorphous polymers is the failure of the Debye theory [7] of dielectric relaxation that

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enables to describe adequately the low frequency spectrum. The experimental data are deviated considerably from the exponential (Debye) pattern and exhibits a broad distribution of relaxation times. The dielectric spectroscopy experiments show also of an asymmetrically broadened relaxation peak, conventionally defined as the α-relaxation peak that flattens into an excess wing at high frequencies, [16, 22].

Most of the theoretical and experimental works use a small number of empirical expressions such as the Cole-Davidson (CD) [5], Havriliak-Negami (HN) [8] or Kohlrausch-Williams-Watts (KWW) [25] formulae for fitting of the asymmetric α-relaxation peak. All of these phenomenological fitting formulae are obtained by the method of introducing a fractional "stretching" exponent into the standard Debye relaxation in the time or frequency domain. In the frequency domain the relaxation is described in terms of the normalized complex dielectric permittivity (NCDP)

$$\hat{\varepsilon}(i\omega) = \frac{\varepsilon(i\omega) - \varepsilon_\infty}{\varepsilon_0 - \varepsilon_\infty}, \tag{1.1}$$

where $\varepsilon(i\omega)$ determines the complex dielectric permittivity (CDP), ε_0 and ε_∞ are low-frequency and high-frequency limits of the CDP $\varepsilon(i\omega)$, correspondingly. Relaxation in the time domain is described by the normalized relaxation function $\phi(t)$ defined as

$$\phi(t) = \begin{cases} P(t)/P(0), & t \geq 0, \\ 0, & t < 0. \end{cases} \tag{1.2}$$

Here $P(t)$ denotes an experimental relaxation function (the electrical polarization). Experiments in the time domain measuring of $\phi(t)$ are related to experiments in the frequency domain measuring $\mathtt{Re}[\hat{\varepsilon}(i\omega)]$ (or $\mathtt{Im}[\hat{\varepsilon}(i\omega)]$) through the formula [2]

$$\hat{\varepsilon}(i\omega) = \hat{L}\left[-\frac{d\phi(t)}{dt}; i\omega\right] = 1 - i\omega\hat{L}\left[\phi(t); i\omega\right], \tag{1.3}$$

where $\hat{L}\left[\phi(t); i\omega\right]$ is the Laplace transform of the relaxation function. Modern broadband dielectric spectroscopy [23] utilizes time and frequency domain measurements simultaneously.

Dielectric loss spectra very often show a marked excess contribution at frequencies some decades above the peak frequency of the α-relaxation, [16]. Experimentally, this phenomenon is defined as the "excess wing". It has been noted in early works [5], but up to now there is no commonly accepted model and general explanations (based on the micro/mesoscopic theory or principles) of the excess wing phenomenon in a wide class of glass-forming and other disordered materials.

As it has been marked above for an adequate description of α - relaxation peak figuring in the dielectric loss spectrum some empirical relationships for description of the NCDP were suggested. In the paper [6], Davidson and Cole discussed the two-parameter expression

$$\hat{\varepsilon}(i\omega) = \frac{1}{(1 + i\omega\tau_\beta)^\beta}\,, \tag{1.4}$$

for the NCDP containing a single stretching exponent $0 < \beta \leq 1$ and single relaxation time constant $0 < \tau_\beta < \infty$. Another popular three-parameter fitting empirical formula for the frequency-dependent dielectric permittivity was suggested and discussed by Havriliak and Negami [8]:

$$\hat{\varepsilon}(i\omega) = \frac{1}{(1 + (i\omega\tau_H)^\alpha)^\beta}\,, \tag{1.5}$$

with two stretching exponents $0 < \alpha, \beta \leq 1$ and one relaxation time $0 < \tau_H < \infty$. Many works on dielectric relaxation utilize also the earlier Cole-Cole formula [3] (obtained by setting $\beta = 1$ in equation (1.5)), which, however, it leads to the symmetrically broadened peak in contrast with the most experimental observations of α-asymmetrical peaks. We should note also that the formulae given above do not allow in describing adequately the "excess wing" phenomenon.

Among first expressions suggested for description of the "excess wing" phenomenon it is necessary to mark three-parameter formula for description of the two-step Debye's relaxation

$$\hat{\varepsilon}(i\omega) = \frac{1 + i\omega\tau_2(1 - C)}{(1 + i\omega\tau_1)(1 + i\omega\tau_2)}\,, \tag{1.6}$$

where $0 < \tau_1 < \tau_2 < \infty$ are the two relaxation times and C is a parameter that fixes the relative dielectric strength of the two-step relaxation processes. Of course, this formula is equivalent to the popular relaxation time distribution model

$$\hat{\varepsilon}(i\omega) = \int\limits_0^\infty \frac{g(\tau)d\tau}{1 + i\omega\tau}\,, \tag{1.7}$$

with a sum of two δ-distributions for the probability density relaxation times function

$$g(\tau) = \left(1 - \frac{C}{1 - \tau_1/\tau_2}\right)\delta(\tau - \tau_1) + \left(1 - \frac{C}{1 - \tau_1/\tau_2}\right)\delta(\tau - \tau_2). \tag{1.8}$$

At present time the most adequate mathematical tool for construction of micro/mesoscopic models and following from them the fitting functions for the CDP is associated with fractal theory and fractional calculus. This tool, in turn, is tightly related with self-similar (fractal) character of relaxation

phenomena in disordered media. The scientist R. Hilfer in the frame of his theory of the "fractional dynamics" [9, 13, 14] derived the three-parameter fitting function for the NCDP [11]

$$\hat{\varepsilon}(i\omega) = \frac{1 + (i\omega\tau_\gamma)^\gamma}{1 + (i\omega\tau_\gamma)^\gamma + i\omega\tau'_\gamma}\,. \tag{1.9}$$

It contains one "stretching" power-law exponent γ and two relaxation times $0 < \tau_\gamma < \tau'_\gamma < \infty$. The given expression as it has been shown in [12] is in a good agreement with experimental dielectric data for propylene carbonate at $T = 193K$. It describes well not only the asymmetric α peak, but the "excess wing" in high-frequency region as well.

All expressions shown above were written in frequency region. They can be transformed to time-domain based on relationship (1.3). The corresponding expressions for the NCDP in time-domain were given in paper [10].

The most suitable way for the theoretical justification of the suggested expressions is based on idea of construction of kinetic equations for the corresponding relaxation functions. Using the mathematical apparatus of the fractional calculus [21] one can derive the kinetic equations for relaxation function (1.2), which, in turn, lead (by means of relationship (1.3)) to empirical equations for the NCDP (for example, to Cole-Cole(CC), Cole-Davidson(CD) and Havriliak-Negami(HN)). For example, the simplest kinetic equation for relaxation function (1.2) leading to the CC expression has the following form

$$\frac{d\phi(t)}{dt} + \tau^{-\alpha}\,{}_0D_t^{1-\alpha}\,\phi(t) = 0, \tag{1.10}$$

where ${}_0D_t^{1-\alpha}$ is the Riemann-Liouville differential operator of non-integer order, [21]:

$${}_0D_t^{1-\alpha}\,f(t) = \frac{1}{\Gamma(\alpha)}\frac{d}{dt}\int\limits_0^t \frac{f(t')}{(t-t')^{1-\alpha}}dt',\;\; 0 \le \alpha < 1, \tag{1.11}$$

here $\Gamma(\alpha)$ determines the gamma-function. The possibility of construction of the differential equation of such type is tightly related with the mathematical property of the non-integer differential operator [21]

$$\hat{L}\left[{}_0D_t^{1-\alpha}\,f(t); i\omega\right] = (i\omega)^{1-\alpha}\hat{L}\left[f(t); i\omega\right]. \tag{1.12}$$

By analogy, one can write kinetic equations with "fractional" operators leading in the frequency region to the NCDP expressions suggested empirically for the CD and HN functions. So the problem of construction of the corresponding micro/mesoscopic model is formulated as follows: based on the Mori-Zwanzig general approach the corresponding model can be derived and justified theoretically and confirmed on available dielectric data

experimentally. It has been done in papers [15, 19, 18, 20]. The suggested model is based on idea of self-similar (fractal) spatial-temporal organization of relaxation process in disordered dielectric media. In the frame of this model we obtain the kinetic equation (1.10) and gave the physical interpretation of the power-law exponent entering previously into the CC empirical expression. It is necessary to note that we found the relationship of this power-law exponent with the structural parameters of the disordered medium considered. The previous developed in papers [15, 19, 18] can be generalized for receiving the mathematical expressions for the CDP, which can describe the asymmetric relaxation peaks containing "excess wing" in high-frequency region.

The basic aim of this paper can be formulated as follows: (a) to generalize the model of dielectric relaxation based on the fractional kinetics [15, 19, 18] for the case of the asymmetric behavior of dielectric loss peak in frequency region; (b) to describe these peaks by the corresponding expressions obtained for the CDP including the "excess wing" phenomenon.

In the frame of the linear response approximation, the fluctuations of polarization caused by the thermal motion are the same as for the macroscopic reconstruction induced by the electric field [24, 4]. Thus, one can equate the relaxation function and the macroscopic dipole correlation function (DCF) $\psi(t)$ as follows

$$\phi(t) \cong \psi(t) = \frac{\langle \mathbf{M}(t)\mathbf{M}(0)\rangle}{\langle \mathbf{M}(0)\mathbf{M}(0)\rangle}\,, \tag{1.13}$$

where $\mathbf{M}(t)$ is the macroscopic fluctuating dipole moment of the sample volume unit, which is equaled to the vector sum of all entering in it molecular dipoles. The rate and laws governing the DCF $\psi(t)$ are directly related to the structural and kinetic properties of the sample and characterize the macroscopic properties of the system under study. The most general, from our point of view, the way for calculation of the temporal correlation functions (TCF) (the DCF (1.13) is obviously referred to the TCF as a partial case) is based on the formalism of projection operators developed earlier by Mori [17] and Zwanzig [26]. This approach can be considered as the most general because it is based on the first principles. In the frame of this approach the equation for the TCF has the form [1]

$$\frac{d\psi(t)}{dt} = -\int\limits_0^t K(t-t')\psi(t')dt', \tag{1.14}$$

where $K(t)$ determines the memory function (MF) satisfying to the normalized condition

$$\tau \int\limits_0^\infty K(t)dt = 1. \tag{1.15}$$

Here the normalized parameter τ characterizes (it can be nonexponential)) some characteristic relaxation time. If the memory in the system considered is absent (Marcovian process) then $K(t) = \tau^{-1}\delta(t)$ that reproduce the exponential behavior of the TCF. In real situation we deal with processes having a memory. The presence of memory leads to the nonexponential behavior of the TCF (and for DCF, in particular). Let us transform equation (1.14) to more convenient form by means of integration in the limits from 0 to t and using the property (1.15):

$$\psi(t) = -\int_0^t M(t-t')\psi(t')dt', \tag{1.16}$$

where the function

$$M(t) = \int_t^\infty K(u)du, \tag{1.17}$$

is determined as the integral memory function (IMF).

If now integrate equation (1.10) from t to ∞, then it can be transformed to the same form (1.16)

$$\phi(t) = -\tau^{-\alpha}\ {}_0D_t^{-\alpha}\ \phi(t), \tag{1.18}$$

where ${}_0D_t^{-\alpha}$ denotes the Riemann-Liouville non-integer order integration operator, [21]

$${}_0D_t^{-\alpha}\ f(t) = \frac{1}{\Gamma(\alpha)}\int_0^t \frac{f(t')}{(t-t')^{1-\alpha}}dt'. \tag{1.19}$$

Comparing equation (1.18) with (1.16), we conclude that the CC expression is restored with the help of the following equations written for the IMF and MF, correspondingly

$$M_{CC}(t) = -\frac{\tau^{-\alpha}t^{\alpha-1}}{\Gamma(\alpha)}\,,\ \ K_{CC}(t) = \frac{\tau^{-\alpha}t^{\alpha-2}}{\Gamma(\alpha-1)}\,. \tag{1.20}$$

In a such way, based on the Zwanzig-Mori formalism and taking into account the presence of the memory function we conclude that the nature of non-Debye relaxation is related closely with memory effects that are expressed mathematically in the form of power-law behavior of MF (1.20).

Again, using the results obtained in papers [15, 19, 18] we conclude that the power-law behavior of the IMF appears in the result of averaging of the hierarchy-subdominanted relaxation processes having the distribution function of the type

$$g(t) = \sum_{l=-L_{min}}^{L_{max}} \frac{N_l}{N}\delta(\tau-\tau_l), \tag{1.21}$$

where $N_l = N_0 b^l$ determines the number of dipoles referring to the l-th level of the hierarchy-organized relaxation process, $\tau_l = \tau_0 \xi^l$ defines the corresponding relaxation time describing this level, N characterizes the total number of dipoles, $b, \xi > 1$ the structural self-similar parameters (spatial and temporal (or dynamical), correspondingly) of the substance considered. The parameters N_0 and τ_0 in (1.21) characterize the mesoscopic level of the system and the summation limits exceed the unit value, i.e. $L_{min}, L_{max} \gg 1$. The mathematical justification of this relaxation model was given in [15]. This relaxation process we define as the "Cole-Cole mechanism". In the result of averaging with distribution function (1.21) the IMF accepts the form (1.20), where the power-law exponent coincides with the "fractal dimension" of the spatial-temporal self-similar (fractal) ensemble

$$\alpha = d = \frac{\ln b}{\ln \xi}, \tag{1.22}$$

and the macroscopic relaxation time τ is related to the structural parameters of the medium considered as

$$\tau^{-d} = -\frac{N_0 d}{N \ln \xi} \hat{G}(1-d)\Gamma(d)\tau_0^{-d}. \tag{1.23}$$

Here $\hat{G}(s)$ is the Mellin-transform of the function $\tau M(t)$ from (1.17).

2. The generalized Cole-Cole expression

The generalization of the formalism developed above one can start from the following observation. In the "CC mechanism" the relaxation process is developed successively capturing on each stage the increasing number of dipoles. This process evokes the growth of the dynamical fractal cluster of the dipoles that have passed through the stage of relaxation. It is natural to make a supposition that there is a situation when for all dipoles of the sample considered another mechanism of relaxation is switched on. For example, it can be the process of relaxation to thermostat (heat reservoir), surrounding the dipole system considered with another relaxation time τ'. For certainty, we accept that this independent mechanism follows to the simplest Debye's (exponential) law. Now we have two relaxation processes when the "CC relaxation mechanism" is accompanied by the relaxation process to a heat reservoir. From the mathematical point of view in this case we have the product of two relaxation processes

$$\phi(t) = \phi_{CC}(t) \exp(-t/\tau'), \tag{2.1}$$

where the function $\phi_{CC}(t)$ figuring on the right determines the "CC relaxation mechanism". Finding from here the desired function $\phi_{CC}(t)$ and inserting it into expression obtained into equation (1.18) we obtain finally

the integral equation for relaxation function describing two relaxation processes

$$\phi(t) = -\tau^{-\alpha} e^{-t/\tau'} \, {}_0D_t^{-\alpha} \, e^{t/\tau'} \phi(t) = -\tau^{-\alpha} \left(\frac{1}{\tau'} + {}_0D_t^1 \right)^{-\alpha} \phi(t), \quad (2.2)$$

which can be transformed easily to the following form

$$\frac{d\phi(t)}{dt} + \tau^{-\alpha} \frac{d}{dt} \left(\frac{1}{\tau'} + {}_0D_t^1 \right)^{-\alpha} \phi(t) = 0. \quad (2.3)$$

From equation (2.2) one can notice that the IMF for two relaxation processes of such type accepts the form $M(t) = M_{CC}(t) \exp(-t/\tau')$. The stationary solution of equation (2.3) in frequency region can be presented as

$$\hat{L}[\phi(t); i\omega] = \frac{1}{i\omega \left(1 + (i\omega\tau + \tau/\tau')^{-\alpha}\right)}. \quad (2.4)$$

Taking into account the relationship (1.3) and solution (2.4), one can obtain easily the following expression for the NCDP function which generalizes the CC formula

$$\hat{\varepsilon} = \frac{1}{1 + (i\omega\tau + \Delta)^{\alpha}}, \quad (2.5)$$

where $\Delta = \tau/\tau'$. For demonstration of the influence of the parameter Δ on the relaxation peak form we give on Figure 1 the frequency dependence plot of the imaginary part of the NCDP function, which depends on two parameters Δ and $\lg(\omega)$. Two other parameters are chosen as $\tau = 0.01$ and $\alpha = 0.8$. From this plot it is clearly seen that decreasing of leads to a relatively small shifting of relaxation peak to the high-frequency direction with simultaneous decreasing of its intensity forming finally a plateau region. It is necessary to note that the form of the absorption peak remains symmetrical.

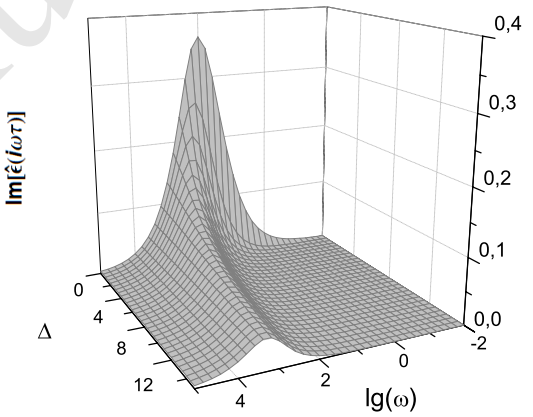

Fig. 1: The frequency dependence of the imaginary part of the NCDP (2.5) at various values of parameter Δ for $\tau = 0.01$ and $\alpha = 0.8$.

3. Multi-channel mechanism of relaxation and the Cole-Davidson expression

The following possible generalization of the previous model can be obtained easily from the following observation. Let us suppose that relaxation process in the dielectric substance considered captures $r+1$ parallel channels and relaxation process in each channel follows to the independent "CC mechanism" having own set of relaxation parameters. For this case the IMF describing the total multi-channel process is written in the form

$$M(t) = \sum_{k=0}^{r} M_{CC}(t, \alpha_k, \tau_k), \tag{3.1}$$

where the right-hand part of this expression is presented in the form

$$M_{CC}(t, \alpha_k, \tau_k) = -\Gamma^{-1}(\alpha_k)\tau_k^{-\alpha_k} t^{\alpha_k - 1}.$$

The substitution of (3.1) into equation (1.16) and subsequent differentiation leads to the following kinetic equation for the relaxation function describing the multi-channel relaxation process

$$\frac{d\phi(t)}{dt} + \sum_{k=0}^{r} \tau_k^{-\alpha_k}\, {}_0D_t^{1-\alpha_k}\, \phi(t) = 0. \tag{3.2}$$

The solution of equation (3.2) in frequency region determines the following expression for the Laplace-image of relaxation function

$$\hat{L}[\phi(t); i\omega] = \frac{1}{i\omega\left(1 + \sum\limits_{k=0}^{r} (i\omega\tau_k)^{-\alpha_k}\right)}, \tag{3.3}$$

which by means of relationship (1.3) leads to the following expression for the NCDP

$$\hat{\varepsilon}(i\omega) = \frac{1}{1 + \left[\sum\limits_{k=0}^{r} (i\omega\tau_k)^{-\alpha_k}\right]^{-1}}. \tag{3.4}$$

We should note that the Hilfer's expression (1.9) for the NCDP is the partial case of expression (3.4) written for two relaxation channels with parameters $\alpha_0 = 1$, $\tau_0 = \tau'_\gamma$ and $\alpha_1 = 1 - \gamma$, $\tau_1^{-\alpha_1} = \tau_\gamma^\gamma / \tau'_\gamma$ but the physical principles of his theory are different in comparison with ideas that are suggested in our theory. Expression (3.4) for the NCDP naturally explains the "excess wing" phenomenon that appears only in the high-frequency region. For proving it is sufficient to consider two limiting cases $\omega \to 0$ and $\omega \to \infty$. Taking the corresponding limits we obtain

$$\begin{aligned} &\lim_{\omega\to 0} \mathtt{Im}[\hat{\varepsilon}(i\omega)] \sim \omega^{\alpha_{max}}, \qquad \alpha_{max} = \mathtt{max}\{\alpha_0, \alpha_1, \ldots, \alpha_r\}, \\ &\lim_{\omega\to \infty} \mathtt{Im}[\hat{\varepsilon}(i\omega)] \sim \omega^{-\alpha_{min}}, \qquad \alpha_{min} = \mathtt{min}\{\alpha_0, \alpha_1, \ldots, \alpha_r\}. \end{aligned} \tag{3.5}$$

From (3.5) it follows that the high-frequency wing of the α-relaxation peak decreases more slowly in comparison with the low-frequency part.

One can suggest the following organization of multi-channel relaxation process. As is has been mentioned above the "CC relaxation mechanism" forms the dynamical fractal cluster of dipoles that have passed through the relaxation process. The number of these dipoles is equaled to $N_l = N_0 b^l$ ($-L_{min} \le l \le L_{maz}$), where b is the scaling factor and $\alpha = d = \ln b/\ln \xi$. It is known that the fractal has the discrete scale invariance property and it is transformed to itself if the scale is changed in b^{k+1} ($k = 0, 1, 2, \ldots$) times. It is natural to suppose that in the simplest case the relaxation channels are organized in accordance with the following law

$$\begin{aligned} &\alpha_k = \ln b^{k+1}/\ln \xi = (k+1)d, \\ &\tau_k = \tau, \;\; k = 0, 1, 2, \ldots, r \to \infty. \end{aligned} \tag{3.6}$$

The condition $\tau_k = \tau$ it is possible to realize at certain combination of the relaxation parameters τ_k. Then taking into account the scenario (3.6) and realizing the trivial summation in (3.4) one can get the following expression for the NCDP

$$\hat{\varepsilon}(i\omega) = \frac{1}{(i\omega\tau)^d}\,. \tag{3.7}$$

If now, in addition, we switch on the Debye's relaxation mechanism into the surrounding medium with relaxation time τ' then by complete analogy with (3.7) one can transform expression (2.4) to the form

$$\hat{\varepsilon}(i\omega) = \frac{1}{(i\omega\tau + \Delta)^d}\,, \tag{3.8}$$

where $\Delta = \tau/\tau'$. Expression (3.8) for the NCDP contains the empirical CD expression at $\tau' \simeq \tau$. Figure 2 reproduces the frequency dependent plot of the imaginary part of the NCDP (3.8) at various values of the parameter Δ and at the fixed values of $d = 0.6$ and $\tau = 0.01$. In comparison with the previous Figure 1, Figure 2 demonstrates a small displacement of the asymmetric peak in high-frequency region and it is decreasing clearly with increasing of the Δ value.

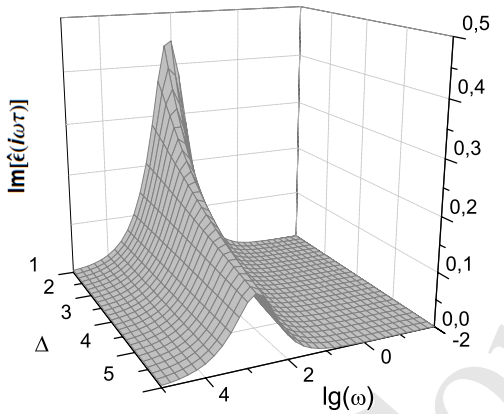

Fig. 2: The frequency dependence of the imaginary part of the NCDP (3.8) at various values of parameter Δ for $d = 0.6$ and $\tau = 0.01$.

4. Conclusion

Generalizing the previous ideas developed in papers [15, 19, 18], we suggest a more general model of dielectric relaxation that contains a set of parallel and independent channels of relaxation with self-similar mechanism of energy percolation. From this model one can obtain rather general expression for the NCDP (3.4) that can be applied with success for experimental description and explanation of the asymmetric-relaxation peaks containing the "excess wing" in high-frequency region. Partial scenario (3.6) realizing the multi-channel mechanism of relaxation with simultaneous relaxation of the dipole system considered to heat reservoir (thermostat) leads to the generalized CD expression (3.8) for the NCDP. So, dielectric relaxation phenomena can be considered as an interesting and perspective direction of the application of the kinetic equations containing non-integer differentiation and integration operators. As one can notice from this approach, the fractional kinetic equations were *derived* from the model of disordered medium containing a set of self-similar structures that participate in the hierarchically-organized dynamical process of relaxation. We did *not* impose the non-integer differentiation and integration operators into the corresponding kinetic equations and tried to derive the desired equations based on the fractal (spatial and dynamical) structure of the disordered medium. We think that these ideas can be useful also for description of other types of relaxations taking place in different disordered media, in particular, for proper description of the mechanical relaxation phenomena.

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