EFFECT OF DISORDER ON DIFFUSION AND VISCOSITY IN CONDENSED SYSTEMS

I. AVRAMOV and A. MILCHEV

Institute of Physical Chemistry, Bulgarian Academy of Sciences, 1040 Sofia, Bulgaria

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A simple theoretical model describing the influence of disorder on transport properties (viscosity, diffusion coefficients, etc.) in undercooled melts and crystals is suggested. The basic assumption is that structural disarray results in a random probability distribution of energy barriers for diffusion characterized by dispersion \( \sigma \) around some mean value \( \langle E \rangle \). It is shown that the effect of \( \sigma \) on the mean jump frequency \( \langle \nu(E) \rangle \) may lead to corrections of many order of magnitude as compared to the hopping frequency calculated traditionally in terms of the average activation energy \( \langle E \rangle \) only. The temperature course of \( \langle \nu(E) \rangle \) is examined making use of the relation between \( \sigma \) and the entropy of the system \( S \). Thus an analytical formula is obtained which properly describes molecular transport in both the crystalline and the amorphous state. Even in a simplified form, \( \eta = \eta_0 \exp(\beta/T^*) \), it reproduces well the existing data on temperature variation of viscosity \( \eta \) (or self-diffusion) in glassforming melts.

In another aspect – in terms of the percolation theory – the model describes the diffusion of a foreign particle in a rigid host structure and yields also a qualitative estimate of the variation of the percolation threshold \( E_p \) with the degree of amorphisation \( \sigma \).

1. Introduction

Recently there has been a vivid interest in modelling of amorphous condensed media which reveal some unique properties important for practical applications (e.g. amorphous semiconductors [1], solid electrolytes on the basis of oxide glasses [2], metallic glasses [3] or fibre optics [4]). It has long been known that disordered systems of different nature show remarkable similarities in some physical properties, such as, e.g., the temperature course of various thermodynamic quantities in undercooled melts [5]. The analysis of existing data on transport properties in disordered systems, meanwhile the kinetic behaviour of, say, dense fluids near a glass transition is quite complex and its description from microscopic principles remains an unsolved problem in condensed matter physics [6,7]. The most encouraging attempts at present for a rigorous microscopic treatment apply predominantly to simplified models of disordered systems. (a hard sphere liquid [8], a kinetic Ising model [9], or a Lennard–Jones fluid [10]) and, although providing a deeper insight into the problem, are still incapable of describing the behaviour of typical glassforming melts. For this latter purpose experimentalists usually employ empirical expressions such as those of Ferry in polymers [12]) or the result of Adam and Gibbs [13], or models based on a “free volume” description of liquids [14–17].

In the present paper we suggest a very general phenomenological model for the description of transport in condensed systems in a state of either dynamic (self-diffusion), or static (percolation) disorder within the framework of the activated complex theory [18,19]. The idea that spatial dis-
order inevitably leads to some probability distribution of the activation energies for elementary jumps, exploited in a previous paper [20], is hereby further developed. The activation energy dispersion σ which should nearly vanish in the crystal is expressed by means of the entropy S of the system, and thus related to existing temperature measurements. Assuming that a hopping particle has always Z channels available to move from a present position, each channel with a different energy barrier E according to the probability distribution function (PDF), \( P(E) \), we calculate the average transition rate \( v(\sigma(T)) \) as indicative for viscosity or diffusion in the system.

In the spirit of the comprehensive arguments of Druger et al. [21], we identify self-diffusion in liquids with the case of dynamic disorder when the mean time for system evolution renewal \( \tau_{ren} \) is comparable with the average particle hopping time \( \tau_{hop} \). In cases when \( \tau_{ren} \gg \tau_{hop} \), that is, in cases of static disorder, one deals with a situation where a description in terms of a percolation theory should be appropriate. In this case we estimate the impact of a varying degree of disorder on the percolation threshold energy \( E_p \) for a tracer under "soft" percolation conditions, i.e. when more or less conductive (or rapid) bonds (channels) are encountered by the hopper according to the PDF \( P(E) \). In this second aspect our work confirms and extends some recent results of Solovev [22].

2. Description of the theoretical model

The transition rate \( v \) within the framework of the activated complex theory [18,19] is related to some activation energy barrier \( E \) by means of

\[
v = v_0 \cdot \exp(-E/RT),
\]

where \( v_0 \) is the frequency of vibration of a particle within an energetic cell (\( v_0 = 10^9 - 10^{12} \)) and \( R \) is the gas constant. The shear viscosity \( \eta \) is defined as \( \eta = G/v \) with \( G \) the infinity frequency shear modulus, and, respectively, the diffusion coefficient, \( D \), as \( D(T) = a^2 \cdot v(T)/2d \) where \( a \) is the mean jumping distance and \( d \) stands for the dimension of space.

While in a perfectly ordered crystal one could assume the existance of a single activation energy, \( E_c \), spatial disorder in an amorphous system should imply the activation energy \( E \) being a random variable, subject to some probability distribution \( P(E) \). Assuming that the hopping particle may use \( Z \) different channels to move out from one coordination sphere into another (\( Z \) could be viewed as roughly proportional to the coordination number of the lattice), it is clear that both \( Z \) and \( P(E) \) will determine the average transition rate of particles between neighbouring sites. Evidently, in the case of dynamic disorder, \( \tau_{ren} \leq \tau_{hop} \), after each successive jump the particle faces the same degree of disorder as before, if one neglects for the time being "correlated renewal" of the local environment.

In ref. [20] we determined the probability \( W(E) \) that the hopper will overcome a barrier of height between \( E \) and \( E + dE \), if the energy barriers are distributed over \( Z \) escape channels with a probability \( P(E) \), and the particle goes as a rule over the lowest barrier:

\[
W(E) \cdot dE = \left[ \int_{E}^{E_{max}} P(e) \, de \right] Z - \left[ \int_{E + dE}^{E_{max}} P(e) \, de \right] Z \nonumber
\]

\[
= Z \cdot P(E) \cdot \left[ \int_{E}^{E_{max}} P(e) \, de \right] Z^{-1} \cdot dE, \quad (2)
\]

where \( E_{max} \) is the largest possible value of \( E \). Using eq. (2) one obtains for the mean transition rate

\[
\langle v \rangle = \int_{0}^{E_{max}} W(E) \cdot v(E) \cdot dE, \quad (3)
\]

which can be solved for any specified choice of \( P(E) \). It is to be expected that the form of the PDF \( P(E) \) should depend on the peculiarities of the particular system under consideration. We try several analytical forms for \( P(E) \) which could reproduce the unknown genuine PDF with a varying degree of accuracy (fig. 1). Activation barriers, much larger than those in the ordered state, \( E_c \), appear highly improbable since neighbouring particles can hardly get any nearer than in the crystal. In all trial PDF we assume therefore \( E_{max} = E_c \). Moreover, it appears reasonable to restrict the
PDF to cover non-negative activation barriers only, i.e. we have \( 0 \leq E \leq E_c \) as a definition range of \( P(E) \).

Making use of eqs. (1)–(3) one obtains in the case of:

(a) Uniform distribution (fig. 1(d))

\[
P(E) = \frac{1}{\sigma}, \quad \sigma \leq E \leq E_c, \tag{4}
\]

\[
W(E) = Z E_c - E \frac{Z - 1}{\sigma} \left\{ \frac{E_c - \sigma \leq E \leq E_c}{\sigma \leq E} \right\}, \tag{5}
\]

\[
\langle v \rangle = \frac{v(E_c)}{(\sigma/RT)^Z} \times \left\{ \e^{-\sigma/RT} \sum_{k=0}^{Z-1} \frac{(-1)^k (Z-1)!}{(Z-1-k)!} \right. \\
\left. \times \left( \frac{\sigma}{RT} \right)^{Z-1-k} - (-1)^{Z-1} (Z-1)! \right\}. \tag{6}
\]

(b) Poisson distribution (fig. 1(b))

\[
P(E) = e^{(E-E_c)/\sigma} \left[ 1 - e^{-E_c/\sigma} \right]^{-1}, \quad 0 \leq E \leq E_c, \tag{7}
\]

\[
W(E) = Z e^{(E-E_c)/\sigma} \left[ 1 - e^{-E_c/\sigma} \right]^{-1} \times \sum_{k=0}^{Z-1} (-1)^k \binom{Z-1}{k} e^{k(E-E_c)/\sigma}, \tag{8}
\]

\[
\langle v \rangle = \frac{Z v(E_c)}{(1 - e^{-E_c/\sigma})^Z} \times \sum_{n=0}^{Z-1} (-1)^n \binom{Z-1}{n} \frac{e^{n(1/(RT - n - \sigma - 1/\sigma))} - 1}{\sigma/RT - n - 1}. \tag{9}
\]

(c) Gaussian distribution (fig. 1(c))

\[
P(E) = \frac{2 e^{-(E_c-E-E_c/\sigma)^2}}{\sigma \sqrt{\pi} \erf(E_c/\sigma)}, \quad 0 \leq E \leq E_c, \tag{10}
\]

\[
W(E) = Z \left[ \frac{\erf(E_c/\sigma - E/\sigma)}{\erf(E_c/\sigma)} \right]^{Z-1} \times \frac{2 e^{-(E_c-E-E_c/\sigma)^2}}{\sigma \sqrt{\pi} \erf(E_c/\sigma)}, \tag{11}
\]

\[
\langle v \rangle = \int_0^E W(E) \cdot v(E) \cdot dE, \tag{12}
\]

whereby in eqs. (10)–(12) we use the error function,

\[
\erf(x) = \frac{2}{\sqrt{\pi}} \int_0^x e^{-t^2} dt. \]

In eq. (12) one can derive a close analytical expression only for the case of \( Z = 1 \), while higher \( Z \) should be dealt with numerically. However, the final results in eqs. (9) and (12) for all \( Z \) may be put in a very simple form if one takes into account that the expressions in square brackets in eqs. (8) and (11) equal very nearly unity for \( \sigma \geq RT \) and \( E \) varying within that part of the interval where the rest of the integrand contributes most to the value of the integral. So, one can put approximately \( \langle v \rangle \approx Z \cdot \langle v_1 \rangle \). The correctness of this approximation has been confirmed by our computations.
and, since it substantially simplifies the analytical results, we use it extensively hereafter.

The exact results for \( Z = 1 \) are:

(a) **Poisson PDF**

\[
\langle v_1 \rangle = v(E_c) \frac{e^{E_c/(\sigma/RT - 1)}/\sigma - 1}{(1 - e^{-E_c/\sigma})(\sigma/RT - 1)}.
\]

(c) **Gaussian PDF**

\[
\langle v_1 \rangle = v(E_c) \times \left[ \frac{\text{erf}(E_c/\sigma) - \sigma/2RT + \text{erf}(\sigma/2RT)}{\text{erf}(E_c/\sigma)} \right]
\]

\[
\times e^{(\sigma/2RT)^{2}}.
\]

The dispersion \( \sigma \) reflects the degree of disorder in the system and is connected with an entropy \( S \),

\[
S = -\frac{RZ}{2} \cdot \int_0^{E_c} P(E) \cdot \ln P(E) \cdot dE + \text{const.}
\]

In eq. (15) it is taken into account that a system of \( N \) particles has \( NZ/2 \) escape channels and that the various activation barriers occur statistically independently. Since in the dependence of \( S \) on \( \sigma \) in eq. (15) an unknown constant is always present, we operate rather with the entropy difference \( S - S_0 \) where \( S_0 \) is some reference entropy corresponding to dispersion \( \sigma_0 \). For the three PDFs under consideration one obtains from eq. (15) with eq. (4), or (7), or (10) the entropy as:

(a) **Uniform PDF**

\[
S - S_0 = \frac{RZ}{2} \ln(\sigma/\sigma_0), \quad \sigma \leq E_c.
\]

(b) **Poisson PDF**

\[
S - S_0 = \frac{RZ}{2} \left[ \ln(\sigma/\sigma_0) + \ln \frac{1 - e^{-E_c/\sigma}}{1 - e^{-E_c/\sigma_0}} \right]
\]

\[
- \ln \frac{1 - e^{-E_c/\sigma}}{1 - e^{-E_c/\sigma_0}}
\]

\[
+ \ln \frac{1 - e^{-E_c/\sigma_0}}{1 - e^{-E_c/\sigma}}.
\]

It can be readily shown that in eqs. (16)–(18) the contribution of all but the first term on the rhs is vanishing, so

\[
S - S_0 = \frac{RZ}{2} \cdot \ln(\sigma/\sigma_0)
\]

is a very good approximation to eqs. (16)–(18) for every reasonable choice of the parameters \( E_c, \sigma, \) and \( \sigma_0 \).

In line with the considerations above we express the viscosity \( \eta \) in the case of a Poisson PDF, cf. eq. (13), as

\[
\eta = \frac{G}{Z \sigma_0} \frac{(\sigma/RT - 1)(1 - e^{-E_c/\sigma})}{e^{-E_c/\sigma} - e^{-E_c/RT}}.
\]

For a constant heat capacity, \( C_p = A \), one has for the entropy,

\[
S - S_0 = A \cdot \ln \left( \frac{T}{T_0} \right),
\]

where \( T_0 \) is the temperature at which the entropy is \( S_0 \). With eq. (21), eq. (20) may be further simplified for \( RT < \sigma < E_c \). Then one has in dimensionless temperature, \( \theta = T/T_m \),

\[
\ln \eta = \ln \eta_0 + \beta/\theta^\alpha,
\]

where

\[
\eta_0 = \frac{G}{Z \sigma_0} \left( \frac{\sigma_0 T_0^\alpha - 1}{RT_0^\alpha} \right), \quad \beta = \frac{E_c}{\sigma_0} \left( \frac{T_m}{T_0} \right)^\alpha
\]

and

\[
\alpha = 2A/RZ
\]

distinguishes through the value of \( C_p \) whether the system is in a liquid or in a crystalline state. In eq. (12) one has three parameters, \( \alpha, \beta \) and \( \eta_0 \), which
can be estimated to be confined within the following range of variation, $A/2R \leq \alpha \leq A/6R$, respectively, $10/Z \leq \alpha \leq 50/Z$, $0.1 \leq \beta \leq 5$ and $10^{-2} \leq \eta_0 \leq 10^2$ (poise).

An analogous simple relation for $\eta$ and a Gaussian PDF (making use of eq. (14)) has not been derived. Our numerical calculations have shown, however, that the $\eta$ vs. $T$ relationships calculated with Gaussian and with Poisson PDFs do not differ significantly; moreover, they can be made to coincide for a slightly different choice of parameters.

3. Comparison with experiment

Concerning the physical meaning of $S$, one should bear in mind that it reflects the degree of spatial disorder of the structural building units whereby their internal structure is considered irrelevant. For that reason contribution to entropy, due to internal degrees of freedom of the building units should not be taken into account. So we assume that $S$ in our model comprises the configurational part of the entropy of the disordered system, $\Delta S$, plus the vibrational entropy of an ideal classical crystal with $C_p \approx 3R$. In ref. [23] it has been shown that $\Delta S$ of undercooled melts can be calculated with sufficient accuracy under the assumption that the excess heat capacity, $\Delta C_p$, is constant above $T_0$,

$$\Delta C_p = \begin{cases} 0, & \text{for } 0 < T < T_0 \\ C = \text{const}, & \text{for } T_0 < T < T_m \end{cases} \quad (23)$$

$T_0$ is usually identified as marking the point where the entropy curves of the undercooled melt and of the respective crystal merge [24–26]. For a great variety of glassforming melts $T_0$ is known to be of the order of 0.3–0.5 of the melting temperature $T_m$.

Taking into account eq. (23), one has for the entropy:

$$S - S_0 = A \cdot \ln\left(\frac{T}{T_0}\right), \quad (24)$$

where

$$A = \begin{cases} C + 3R, & \text{for liquids,} \\ 3R, & \text{for crystals.} \end{cases} \quad (25)$$

Using eq. (19), one obtains for $\sigma$,

$$\sigma = \sigma_0 \left(\frac{T}{T_0}\right)^{2A/RZ}, \quad (26)$$

which should be inserted into the expressions for $\langle v \rangle$. At $T_0$ the system in equilibrium is assumed to be characterized by dispersion $\sigma_0$. According to the theory of thermodynamic fluctuations, one could expect $\sigma_0 \approx RT_0$.

Usually there are two experimentally observed quantities which directly reflect the rate of molecular motion in condensed media – viscosity $\eta$ and self-diffusion coefficient $D$ – both expressed in terms of $\langle v \rangle$. Since $D$ is reciprocal to $\eta$, we confine our study to the temperature course of viscosity where a huge body of experimental data exists [27].

A comparison of our results with existing experimental data can be made, provided the parameters of the model are known or can be estimated. Quite generally one can argue that:

1. $E_c$ is expected to be between 10 and 100 kcal/mol.
2. The value of $G/Z\theta_0$ is between $10^{-2}$ and 10 poise.
3. The number of escape channels $Z$ is practically the coordination number, $2 \leq Z \leq 12$, as far as short range order is retained even in liquids.

In fig. 2 the temperature course of viscosity as following from eq. (20), combined with eqs. (25), (26), is plotted for a typical glassforming system, glucose, both for the crystal (dashed line) as well as for the undercooled melt (full line), and is compared with the simplified result, eq. (22) (dots). Evidently eq. (20) not only reproduces fairly well the existing data, but for the first time predicts also a finite viscosity in the vicinity of $T_0$, as has recently been observed [28]. In contrast, other models [13–17] predict $\eta = \infty$ at $T = T_0$ and a vanishing self-diffusion coefficient in the respective crystalline state at any $T$. In the present model the viscosity of the crystal appears always high enough so that it is solid up to the melting point, resulting, however, in a non-zero value for the self-diffusion coefficient $D_0$, in agreement with experimental evidence.
Figure 2 also shows that in coordinates \( \log \eta \) vs. \( \theta^{-1} \) the simplified result, eq. (22), does not deviate significantly from the solution (20) within the temperature interval of practical interest. In order to examine the ability of eq. (22) of reproducing experimental data on viscosity, it has been applied to different types of glassforming substances – an organic (glucose [30]), semimetal (Se [31]), oxide glass (\( \text{B}_2\text{O}_3 \) [27]) and two methaphosphate glasses (\( \text{NaPO}_3 \) [32] and \( \text{LiPO}_3 \) [33]). In fig. 3 it is shown that eq. (22) describes fairly well the experimental data with values of the parameters \( \alpha, \beta \) and \( \eta_0 \), given in table 1.

### Table 1

<table>
<thead>
<tr>
<th>Substance</th>
<th>( \log \eta_0 )</th>
<th>( \alpha )</th>
<th>( \beta )</th>
</tr>
</thead>
<tbody>
<tr>
<td>Glucose</td>
<td>-1.48</td>
<td>6.26</td>
<td>3.82</td>
</tr>
<tr>
<td>Se</td>
<td>-0.47</td>
<td>4.42</td>
<td>3.27</td>
</tr>
<tr>
<td>( \text{B}_2\text{O}_3 )</td>
<td>1.26</td>
<td>3.15</td>
<td>9.52</td>
</tr>
<tr>
<td>( \text{NaPO}_3 )</td>
<td>0.94</td>
<td>8.33</td>
<td>0.48</td>
</tr>
<tr>
<td>( \text{LiPO}_3 )</td>
<td>0.38</td>
<td>5.53</td>
<td>2.26</td>
</tr>
</tbody>
</table>

4. Diffusion of foreign particles

As mentioned in the Introduction, in the case of \( \tau_{\text{ren}} \gg \tau_{\text{hop}} \) one deals with a situation when foreign particles diffuse through a "frozen in" host structure in a state of static disorder. Now the activation energy barriers may still be distributed over the channels according to the same PDF, \( P(E) \), but the "easy escape" probability \( W(E) \) eq. (10) cannot be used to calculate \( \langle v \rangle \) because successive jumps are strongly correlated and a percolation theory consideration of the problem appears more justified. Solovev [22] has suggested to define a percolation threshold barrier \( E_p \) as

\[
\int_0^{E_p} P(E) \cdot dE = \rho_c,
\]

where the critical concentration of conducting channels, \( \rho_c \), depends both on the number of available escape channels \( Z \) and on space dimension \( d \). There exist a number of estimates for \( \rho_c \) in the literature (see Zimán [34]) and here we adopt the formula

\[
\rho_c = \frac{d}{(d-1)Z},
\]

which is shown to be satisfied within a few percent in all cases [35]. For the variation of the percolation energy threshold, \( E_p \), one obtains then:

(a) Uniform PDF

\[
E_p/E_c = 1 - (1 - \rho_c) \alpha/E_c, \quad 0 \leq \alpha \leq E_c.
\]

(b) Poisson PDF

\[
E_p/E_c = (\alpha/E_c) \ln\left( \rho_c e^{E_c/\alpha} + 1 - \rho_c \right).
\]
(c) Gaussian PDF
\[
\text{erf}\left[\left(1 - E_p/E_c\right)(E_c/\sigma)\right] = \left(1 - p_c\right) \text{erf}(E_c/\sigma).
\]
(28c)

It is evident from eqs. (28a–c) that \(E_p/E_c \rightarrow 1\) for \(\sigma \rightarrow 0\) and that \(E_p/E_c \rightarrow p_c\) for \(E_c/\sigma \gg 1\).

The average transition rate \(\langle v_p \rangle\) depends on \(\sigma\) by means of \(E_p\):
\[
\langle v_p \rangle = v_0 \cdot e^{-E_p/RT}.
\]
(29)

With eqs. (28b) and (23) the temperature course of \(\langle v_p \rangle\) is shown in fig. 4 for \(\sigma_0 = 1000\), \(E_c = 6 \times 10^4\) cal/mol, \(Z = 4\), \(T_0 = 500\) K and \(T_m = 1000\) whereby curve 1 corresponds to an undercooled amorphous phase, \(A = 10R\), while curve 2 represents the respective crystalline state \(A = 3R\). The inlet shows data measurements on specific resistivity of lithium acetate as reported in ref. [36].

It is of interest to compare \(\langle v \rangle\) as obtained for the case of dynamic disorder in sect. 2 to \(\langle v_p \rangle\) corresponding to static disorder for the same set of parameters. This is illustrated in fig. 5. It is seen that both curves have qualitatively the same shape with \(\langle v \rangle \gg \langle v_p \rangle\). In both cases, for vanishing \(\sigma\) it is only \(E_c\) which determines the energetic barrier for diffusion while for very large dispersion \(\sigma\) there is a sufficient number of channels with low energy barriers, so that hopping is not thermally activated. In these two extreme cases both curves actually merge. There is an intermediate range of variation of \(\sigma\), however, where dynamic disorder strongly favours (up to 9 orders of magnitude) the transport process.

In general the system renewal in the case of dynamic disorder may not necessarily occur completely at random, but rather in correlated steps. If has been suggested by Druger et al. [21] that in this case lower diffusion rates should be expected for the same number of overall renewals, since bottlenecks would be less efficiently bypassed. The result of fig. 5 clearly confirms this prediction. Moreover, curves 1 and 2 of fig. 5 may be regarded as envelope curves for a number of intermediate cases of quasi-static disorder in which \(\tau_{\text{ren}}\) is larger but still comparable to \(\tau_{\text{hop}}\).

5. Concluding remarks

In the present work we have developed a simple model describing transport processes in condensed systems with varying degree of disorder. Based on the assumption that structural disorder results in a random probability distribution of the activation energy for molecular transport, the model predicts a non-Arrhenius dependence of viscosity of undercooled melts on temperature. As a merit of the model one obtains a nonzero value of the self-diffusion coefficient in crystals as well as a finite viscosity of the undercooled melt in the vicinity of \(T_0\). For practical use we suggest the simplified eq.
(22) which may be used to describe the temperature course of viscosity with sufficient accuracy for $\eta$ varying within 15 orders of magnitude. As a matter of fact, this equation may be viewed as a generalization of the empirical result of Skornya-
kov [37] where $\alpha = 2$.

All parameters, except $\sigma_0$, which are used in the model, may be calculated in principle from ther-
modynamic and structural data. The temperature behaviour of viscosity of different classes of gla-
sforming systems is well reproduced, provided the heat capacity of the reference ordered state is ad-
to be $3R$ and the entropy of the dis-
ordered state is calculated from $3R$ plus the mea-
sured excess heat capacity of the latter. Although the determination of $\sigma$ for various classes of sub-
stances is a tempting problem for further investiga-
tion, here we note only that for metals and noble gases, where $R \leq \Delta C_p \leq 2R$ and $Z = 10$, one obtains $\alpha = 1$ in agreement with the observed Arrhenius behaviour of $\eta(T)$. From our investiga-
tion it turns out also that the final results are rather insensitive to the particular shape of the PDF and that even PDFs, symmetrically distribu-
ted around $E_\sigma$, lead to the same result, provided some sort of structure is still preserved, i.e., $\sigma < 5E_\sigma$.

The diffusion of foreign particles in a “frozen in” medium is described in terms of a dispersion-
dependent percolation threshold barrier, $E_\sigma(\alpha)$. The consideration is confined to the case of low concentra-
tions of tracers so that interactions be-
tween them are neglected. An increasing degree of disorder $\sigma$ reduces the percolation threshold and may again be expressed in terms of entropy of the system. In this way the predicted temperature behaviour of ionic conductivity in crystalline and amorphous systems qualitatively agrees with experimental evidence.

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