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Geometric distribution of Dynamically Accessible Volume in the Bond Fluctuation Model

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Abstract

The geometry of the Dynamically Accessible Volume, DAV, in polymeric materials has been observed by means of the Bond Fluctuation Model. Simulations were conducted in conditions that yield varying amounts of accessible volume in the lattice, acting on the density of the system, and introducing geometrical restrictions to motion. A parameter has been defined to characterise the connectivity of holes characterising the number and size of hole clusters. The mobility of the accessible cells during athermal simulations has also been characterised concluding that all the holes in the lattice are in motion, contributing to the diffusion of the molecules even for the lowest values of DAV. The number of clusters increases with decreasing DAV reaching a maximum. For values of DAV below that of the maximum number of hole clusters, the diffusion coefficient becomes smaller than that predicted by the quadratic ratio $D = \gamma|\mathrm{DAV}|^2$ The differences in the dynamics of individual molecules and polymer chain systems are studied.

Keywords: Bond Fluctuation Model; Dynamically Accessible Volume; Diffusion

1. Introduction

Conformational mobility plays a central role in the physical properties of glass-forming liquids. In the temperature interval around the glass transition, the conformational rearrangements of the molecules in a low-molecular weight glass-former, or the segments of the polymer chain in a linear polymer or a polymer network, are cooperative movements. The close packing of the molecules makes that the motion of any of them requires the motion of a neighbour molecule that successively needs that of another, thus producing a cascade of events that stops when enough free space is found to lodge a molecule in its destination site without disturbing the rest of the material. Obviously, in this phenomenon not only the amount of free volume, but also its distribution and mobility, are of great importance.

Molecular dynamics calculations using coarse grained lattice models and Monte Carlo techniques can help to understand the influence of the spatial distribution of molecules and holes on the macroscopic behaviour. The Bond Fluctuation Model [1–24], BFM, is a lattice model, which has been widely employed in order to simulate polymer dynamics [1–4,6–10]. In BFM, as in other lattice models, a number of molecules or polymer chains occupy the cells of a lattice. The density of occupied cells is high enough to simulate the properties of a liquid but compatible, in the case of polymer chains, with the difficulty of finding consecutive empty cells to situate the chains. Nevertheless, the total number of cells and the total number of occupied cells are fixed and consequently, the fraction of empty cells, which is a constant, is not a measure of free volume. In fact, the definition of a parameter that quantifies the space situated around the molecules available for their motion is not trivial.

The Dynamically Accessible Volume (DAV) is a parameter which was recently introduced in order to characterise free

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volume in lattice models [25–28]. This parameter distinguishes two kinds of empty cells. The cells that can be occupied in one Monte Carlo Step (according to the rules of the model) are called holes, whereas the rest of the cells are called vacancies. The Dynamically Accessible Volume is the fraction of holes of the system while vacancies are not taken into account. The mathematical definition of DAV can be expressed in the form [29]:

$$\mathrm{DAV} = \frac{1}{L^3}\sum_{j=1}^{L^3}\left(\min\left(1, \sum_{i=1}^{\mathrm{nn}}\delta_{ij}\right)\right) \tag{1}$$

where L is the box simulation size, nn is the number of nearest neighbours of a cell and δ_{ij} is a function defined as 1 when a monomeric jump from the cell i to the cell j is allowed and 0 in any other case.

Dawson and coworkers proposed the following relationship between DAV and the diffusion coefficient [25–28]:

$$D = \gamma|\nu|^2 \tag{2}$$

where D is the diffusion coefficient, γ is a non-universal factor that depends on the model and ν is the DAV. They showed that the diffusion coefficients calculated with the Kob–Andersen [30] and Biroli–Mezard [31] models followed Eq. (2) although significant deviations were found for low values of DAV when the calculated diffusion coefficient is smaller than expected by Eq. (2). The explanation for this behaviour in the case of the Kob–Andersen model was the existence of isolated holes unable to diffuse, and thus not contributing to the mobility of the polymer chains. In fact, when these isolated holes are not taken into account in the calculation of DAV, the quadratic behaviour prevails in the whole DAV range [28].

In our previous work [29] we have tested the quadratic law for polymeric materials with the Bond Fluctuation Model under athermal conditions, showing that the non-universal parameter γ depends on the chain size. On the other hand, a departure from quadratic law was found in the Bond Fluctuation Model for low values of DAV analogous to those shown by the Kob–Andersen and Biroli–Mezard models. Our goal in this work is to study the geometrical distribution and mobility of DAV in the Bond Fluctuation Model depending on molecular size, which gives some insight into the DAV dependence of the diffusion coefficient.

2. Model and simulations

In the Bond Fluctuation Model every molecular group, that represents 3–5 carbons of the polymeric chain, consists of eight occupied sites forming a cube. The distance of the bonds between molecular groups can fluctuate between 2 and $\sqrt{10}$. Distance $\sqrt{8}$ is not allowed in order to avoid bond crossing [3,32].

The dynamics of the model consists of randomly choosing a molecular group and a local monomeric jump of 1 lattice unit. The movement, in the athermal version of the model, will be accepted if volume exclusion is respected and bond lengths are between allowed margins.

All the simulations in this work consisted of a 3D box ($L = 40$) with periodic boundary conditions [1]. No Hamiltonian was employed in order to simulate athermal conditions. All simulations began with an initial period of 10^5 Monte Carlo Steps (MCS) in order to equilibrate the system, followed by a period of 10^4 MCS in which the value of the calculated parameters was averaged. All system configurations were replicated 20 times in order to obtain reliable values. Computer simulation was conducted with polymer chains consisting of $N = 5$, 10, 15 and 20 consecutive segments and on individual molecules ($N = 1$) as well.

Systems with different amounts of Dynamically Accessible Volume were generated by varying the density of occupied cells φ and also using geometrical restrictions to molecular motions, as in our previous work [29]. To allow the motion of a molecule or chain segment from an original position to a neighbour hole, a number of the cells that surround the origin and destination sites, what we called [29] the influence cells, must be empty. The criterion to select the influence cell of a given motion is that the sum of the distance of an influence cell to the molecular group at the beginning and at the end of the movement must be equal or less than $(1+\sqrt{2})$ lattice units. In other words, the distance from the influence cell to the original position of the polymer segment or to the destination site must be 1 lattice unit and must not exceed $\sqrt{2}$ to any of them. This gives a total of 24 influence cells per motion in a 3D lattice. So we define a parameter c in such a way that a movement can only be performed if c or more influence cells of a movement are empty. A value $c = 0$ represents the standard model with no geometric restriction, while increasing c between 0 and 18 (greater values for c led to frozen systems where no diffusion was observed) contributes to the geometric frustration. On the other hand, the interval of variation of the density depended on the size of the chains, due to the difficulty to find enough consecutive empty space to allocate long chains. For chains sized 20 and 15 maximal density was $\phi = 0.55$ and $\phi = 0.56$ respectively. Parameter c was employed in order to obtain different values of DAV for a fixed density value, changing its values between 0 and 18, as in Ref. [29].

The diffusion coefficient was calculated from the expression [1,2,4,6]:

$$D = \lim_{t\to\infty}\frac{g_3(t)}{6t} \tag{3}$$

where $g_3(t)$ represents the evolution in time of the centre of mass of the polymeric chains [1,2,4,6]:

$$g_3(t) = \left\langle\left[\vec{r}_{\mathrm{CM}}(t) - \vec{r}_{\mathrm{CM}}(0)\right]^2\right\rangle \tag{4}$$

3. Results

The relationship between Dynamically Accessible Volume and diffusion coefficient, according to BFM, is shown in

Fig. 1. This plot has been built with the data given in Ref. [29]. The zone of low DAV values is emphasised in this plot. We can observe that the points corresponding to the simulations with all systems, polymer chains of different lengths and individual molecules as well, converge for high DAV values on the straight line that represents the quadratic behaviour of Eq. (2). Nevertheless, the points separate from this behaviour for DAV values below a certain critical value that is difficult to specify. It is worth noting that all the points corresponding to the simulations of polymer systems fall on the same line, but the simulation of a low-molecular weight substance gives diffusion coefficients higher than those of the polymers for the same DAV for values below 0.1.

3.1. The distribution of Dynamically Accessible Volume

As explained above, one of the possible causes of the separation of the diffusion coefficient from the quadratic relationship is the existence of part of the holes that are counted as DAV but, in fact, are isolated and do not contribute to the diffusion of molecules or polymer segments. The following procedure has been used in this work to quantify the connectivity of DAV and detect the presence of isolated holes in BFM. When the accessibility of a cell in a MCS was determined, and thus considered as a hole, we considered how many of the six nearest neighbours could be regarded as a hole as well. This connectivity offers a first approach to know whether the cells that constitute the Dynamically Accessible Volume are isolated or, on the contrary, clustered. Fig. 2 shows a linear relationship of the connectivity between holes while DAV decreases. This linear relationship only seems to be lost when DAV reaches very low values, but there is no change in the observed slope at the DAV values that correspond to the loss of the quadratic law of Eq. (2).

The asymptotic limit for the least possible value of connectivity is 2. This is due to the topology of the model. Every molecular group forming a cube needs four free adjacent lattice cells forming a square to move. A small number of empty cells cannot form a hole because the molecular groups occupy more space. This unit of four free accessible cells is the minimal expression for DAV in the Bond Fluctuation Model, and every empty cell in this square is connected to two other neighbours; thus, minimal possible connectivity for DAV is 2. On the other side, a great difference between monomeric beads and polymeric chains is observed in Fig. 2, but all curves corresponding to polymers collapse in a unique master curve independent of the chain size. This shows the importance of the study of polymer dynamics.

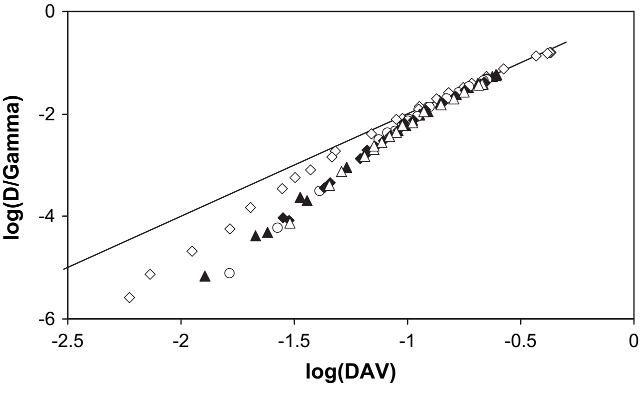

Fig. 1. Double logarithmic representation of DAV against the normalised diffusion coefficient (see Ref. [29]) for chains sized 1 (◇), 5 (▲), 10 (○), 15 (◆) and 20 (△). The straight line represents the quadratic law.

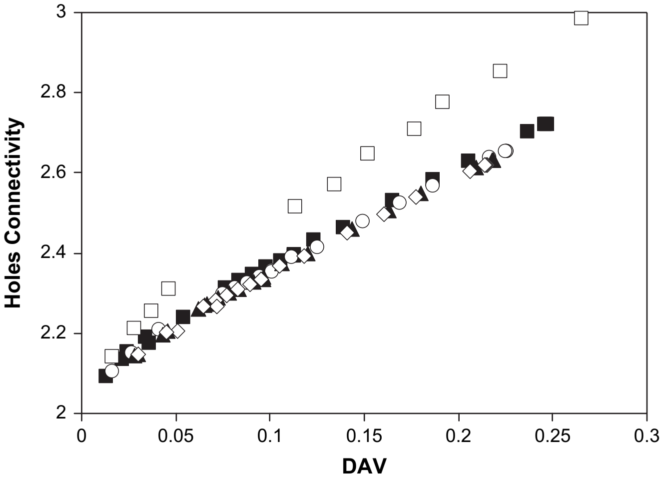

Fig. 2. Holes connectivity (see text) as a function of DAV for chains sized 1 (□), 5 (■), 10 (○), 15 (▲) and 20 (◇).

Once confirmed that in BFM the connectivity of the holes is not directly related to the loss of the quadratic law, we studied the topological distribution of DAV characterised by the number of hole clusters. We consider that two holes belong to the same cluster if they are directly connected or they are connected by means of a path formed by other holes. So, two cells may belong to the same cluster even if they are not directly connected.

The number of clusters was found to depend not only the value of DAV, but also on a number of parameters that characterise the system, such as density and geometric restrictions. In all systems, this number of clusters presents a maximum for a given value of DAV. To facilitate the comparison, Fig. 3 shows the values of the number of clusters normalised by dividing all of them by the maximal number of clusters found for a given system. It shows that the value of DAV for which the number of clusters is maximum is independent of the chain length in the case of polymer systems, but in the system consisting of not bonded molecules the whole curve is shifted towards lower values of DAV.

For high values of DAV, the number of holes is so large that it is very easy to find paths connecting them, so holes are distributed in a small number of large clusters. As DAV decreases these clusters are fragmented, thus the average size of the clusters decreases and their absolute number increases. At the point when the maximal value of clusters is reached, the empty space is totally ‘atomised’. Further decrease of DAV, below that of the maximum, continues diminishing the mean size of the clusters as shown in Fig. 3, but their number is decreasing as well.

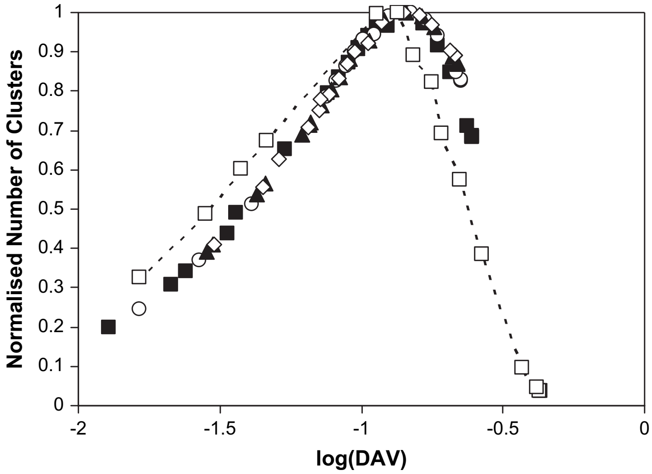

Fig. 3. Normalised number of clusters of Dynamically Accessible Volume (see text) represented against DAV for chains sized 1 (□), 5 (■), 10 (○), 15 (▲) and 20 (◇). Lines are only an intended guideline for the eye.

The maximum number of hole clusters in a system consisting of non-bonded molecules is reached for a DAV value of approximately 0.12, while in polymers this value increases to 0.14. It is not easy to calculate the exact point at which the quadratic behaviour is lost, but the values of DAV in those points are close to those of the maximum in Fig. 3. Thus, the absolute values of DAV in the critical point are smaller in the individual molecules than in polymer systems and the behaviour of the polymer systems seems to be independent of the chain length. These features support the existence of a relationship between the quadratic law and the evolution of the total number of clusters of the system.

3.2. Mobility of Dynamically Accessible Volume in the Bond Fluctuation Model

Apart from the distribution of holes in the lattice, what seems essential to the evolution of the diffusion coefficient is the ability of the holes to change their position contributing to the diffusion of the molecules or polymer segments. As mentioned above, in the Kob–Andersen model, the isolation of holes from the network of vacancies [28] makes those holes become blocked and unable to contribute to the diffusion of the system. In order to test this feature with the Bond Fluctuation Model, we calculated which part of the holes was blocked and which one was on movement, contributing to the diffusion of particles. Following the evolution of DAV is not so easy as following the molecular groups. A hole can be created or disappear depending on the dynamics of the model, and an oscillating movement can create and destroy a hole while there is no diffusion.

In order to tackle this task, we have taken an ‘image’ of the simulation box for every 100 Monte Carlo Steps during 10^4 MCS, so we have 100 configurations showing the evolution in time of the system. If the system is really blocked and no diffusion is observed, all configurations must be similar (holes stay at the same position). In order to compare this, we took the first configuration as a reference and calculated its difference (cell by cell) with the configuration of the system in other time instants:

$$\mathrm{Dif\,DAV}(t) = \frac{1}{2L^3}\sum_{i}^{L}\sum_{j}^{L}\sum_{k}^{L}\left|\mathrm{DAV}(t,i,j,k) - \mathrm{DAV}(0,i,j,k)\right| \tag{5}$$

where the function DAV(t,i,j,k) will have a value of 1 if the cell (i,j,k) at time t is a hole and 0 if it is a vacancy or has been occupied by a molecule or chain segment. The factor 1/2 is included because each change of position of a hole is counted two times in the sum. Due to the fluctuation of DAV by the contribution of created or disappeared holes, we averaged these values during 10^4 MCS for 20 simulations in order to cancel random fluctuations caused by Monte Carlo method.

On the other hand, there is a number of holes that even having changed their position during the time elapsed between 0 and t, their position at the time t is the original one, and so do not contribute to the sum of Eq. (5). The probability of such a situation is just DAV^2. Thus, the final fraction of holes that are moving in the lattice is:

$$\mathrm{Mob\,DAV} = \left\langle \frac{1}{2L^3}\sum_{i}^{L}\sum_{j}^{L}\sum_{k}^{L}\left|\mathrm{DAV}(t,i,j,k) - \mathrm{DAV}(0,i,j,k)\right| \right\rangle + \mathrm{DAV}^2 \tag{6}$$

We have calculated this amount of mobile DAV for every simulation and compared it with the total DAVs (Fig. 4). The slope of all lines is between 1.0048 and 1.0188, very close to the unity. This indicates that almost all DAVs are really moving and changing its position during the simulation. This feature can be observed independently of having monomers or different chain sizes, and is valid also for very low values of DAV where the quadratic law is not fitted. This fact indicates that in the Bond Fluctuation Model most of DAVs are

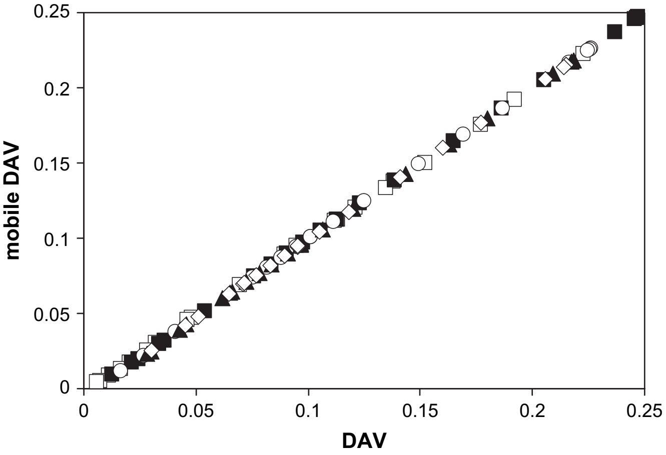

Fig. 4. Mobile DAV (see text) represented versus total DAVs calculated in simulations for chains sized 1 (□), 5 (■), 10 (○), 15 (▲) and 20 (◇).

always mobile, even for low values, contributing to the diffusion of the molecules.

4. Discussion

Dynamically Accessible Volume is, in some way, a measure of the interface between the material and the empty space in the lattice, as seen from the point of view of the empty space. The relationship between the number of accessible cells and molecular mobility is not as direct as it could appear. As an example, an isolated hole surrounded by z occupied cells counts as a unit of DAV (in fact $1/L^3$) while allows the displacement of any of the z molecules in the next Monte Carlo trial, whereas in the case of a plane surface that separates occupied cells and holes there is a hole per molecule with capacity to move. This is just to state that not only the number of accessible empty cells, but also the spatial distribution of holes, vacancies and occupied cells may be important in molecular mobility in lattice models.

Eq. (2) establishes a simple and quite natural relationship between molecular mobility and DAV. Thinking of individual molecules (not polymer chains) and taking into account Eqs. (3) and (4), this relationship expresses that the mean squared distance in the motion of a molecule in the equilibrate system is proportional to the square of the fraction of accessible cells.

In athermal simulations, the distribution of holes, vacancies and molecules in the lattice is just governed by probability criteria according to the mobility of the molecules. During the equilibration of the system, starting from an arbitrary initial distribution of the molecules in the lattice, the system chooses a distribution of holes that depends on the amount of DAV in the system, as has been discussed in Section 3.1. And, with this distribution, if the value of DAV is high enough, Eq. (2) is satisfied. The amount of DAV can be varied in the simulation obviously changing the fraction of occupied cells. In this work, in addition, we have used a geometrical restriction to motion to vary the accessibility of the empty cells. Thus, with the same fraction of empty cells, the system may have different fractions of holes, or different values of DAV. The fact that the points in Fig. 1, corresponding to the same density but different values of the geometrical restriction, fall on the same line than the points corresponding to different densities without any geometrical restriction is a strong support of the adequacy of DAV as a parameter characterising the dynamics of the system.

The relationship given by Eq. (2) determines the highest efficiency of the existing number of holes to facilitate the mobility of the molecules. The dynamics of the system yields to the adequate spatial distribution of holes. As shown in Section 3.1, the system reacts to the decrease of DAV distributing the holes in a higher number of smaller clusters (Fig. 3). According to the example given in the first paragraph of this section, the same number of holes can facilitate the motion of a higher number of molecules. For values of DAV higher than around 0.1 (roughly speaking) the system reaches in this way the maximum mobility determined by Eq. (2).

Situations in which the diffusion coefficient is smaller than that predicted by using Eq. (2) for a given value of DAV can be expected if a part of the empty cells is not useful for the diffusion of the molecules. In thermal systems, the change of the energy of the total systems produced by the motion of a molecule to a neighbour hole plays a role in accepting or not such movement, according to Metropolis criterion. In that case, for instance, a hole in vicinity of an ordered, crystalline surface (accessible according to geometrical criteria) could in fact be never accessed because of the low probability of any molecule that separates from the rest moving to it. In these systems, the definition of DAV used in this work no longer describes properly the dynamics of the system, and a modification was proposed in Ref. [33] to take into account the probability with which a hole can be acceded. But there is no similar situation in athermal systems. In athermal systems the accessibility of a hole is controlled just by geometrical reasons. The possibility mentioned in Ref. [28], with respect to the Kob–Andersen model, that some isolated holes could not contribute to the molecular diffusion, being in practice immobile, has been analysed in Section 3.2 for BFM. Clearly in this model there is no change of behaviour in the mobility of the holes, even for the smallest values of the Dynamically Accessible Volume. A linear relationship was found between the parameter defined in Section 3.2 to characterise the mobility of a hole, Eq. (6), and DAV. In the Bond Fluctuation Model holes propagate quite easily in athermal conditions.

The reason for the decrease of the diffusion coefficient below the quadratic relationship of Eq. (2) must then be found in the impossibility of the dynamics of the system (below a critical value of DAV) to distribute the accessible cells in the whole volume occupied by the molecules. In this way, the local fraction of holes in the vicinity of a number of molecules can become smaller than the average and its mobility reduced. As a consequence, the average diffusion coefficient becomes smaller than that expected. The start of this phenomenon seems to be related to the maximum number of hole clusters (Fig. 3) that establish a change in the trend of distribution of accessible cells in the lattice.

Another interesting feature is the clearly different behaviour of individual molecules and polymer chains. The differences with respect to diffusion coefficient can be due to the fact that D takes into account the mobility of the centre of mass of the polymer chain, and a number of molecular motions of the chain segments cannot actually contribute to the chain diffusion. This can justify that the critical value of DAV for the loss of the quadratic behaviour occurs in polymer chains at DAV values higher than in the individual molecules. Nevertheless, the different behaviour regarding mobility of beads and polymer chains can have more significance, since the distribution of the holes in the lattice is different as well, as shown in Figs. 2 and 3, although the mobility of the holes is exactly the same in both kinds of systems. It is worth noting with respect to this point that the chain connectivity that introduces a restriction to the accessibility of the empty cells in the vicinity of a polymer chain is taken into account when determining the value of DAV.

5. Conclusion

In this work, we have studied the spatial distribution of Dynamically Accessible Volume in the Bond Fluctuation Model. The minimal unit of the Dynamically Accessible Volume is formed by four empty cells forming a square. Two parameters have been used to characterise the influence of the fraction of holes in the lattice on their distribution: one of them is a connectivity parameter that determines the average number of accessible cell sites connected to each other in a cluster, and the other is the number of such DAV clusters. It has been found that the connectivity parameter decreases linearly with DAV until very low values (its minimum value is 2). On the other hand, the number of clusters goes through a maximum when represented against DAV. The mobility of the holes has also been characterised concluding that in BFM there are no immobile holes even for the smallest values of DAV. The quadratic relationship between the diffusion coefficient and DAV determines the maximum efficiency of the existing holes to facilitate the mobility of the molecules. At low values of DAV, the diffusion coefficient becomes smaller than expected by the quadratic law when the number of hole clusters cannot be further increased by the dynamics of the system and a part of the molecules has no direct access to them. There is a significant difference in the critical value of DAV for which the diffusion coefficient separates from the quadratic law in the dynamics of low-molecular weight substances and in polymer chains, but qualitatively the behaviour is the same.

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