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Stochastic matrix description of glass transition in ternary chalcogenide systems

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Abstract

A method using stochastic transfer matrices is applied to description of microcluster growths in typical ternary chalcogenide glasses (Ge–As–Se and Ge–Sb–Se systems) during glass transition. By finding the unit eigenvalue and eigenvector of the stochastic matrix, an analytical relation between the glass transition temperature and the concentration of atomic species is found. In the chalcogen-rich region this formula reproduces the experimentally observed modified Gibbs–DiMarzio equation, which is a semi-empirical rule that relates the average coordination number with the glass transition temperature, using a parameter β which is fixed by the experimental data. The present approach allows us to predict a β close to the observed one.

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1. Introduction

There is still no consensus about which thermodynamical and structural factors are important in determining the glass transition temperature [1] (T_g), although the subject has considerable practical and technological importance. Furthermore, this problem must be intimately related to the question of how the glasses do form [1], i.e., how the complex structures forming the glass network grow as one decreases the temperature of a liquid. In particular, for chalcogenide glasses, attention has been devoted to correlations of T_g with other physical or chemical properties [2], since such glasses have electric and infrared transmission properties that make them useful in several technological applications [3]. Among these chalcogenide glasses, ternary systems such as $Ge_xAs_ySe_{1-x-y}$ and $Ge_xSb_ySe_{1-x-y}$ have been extensively studied [3–5] not only for their applications, but also for testing the Phillips constraint theory [6], since the bonding numbers (valencies) of Ge, As, Sb and Se (4, 3, 3, and 2, respectively) allow the realization of the rigidity threshold (attained when the average coordination number, $\langle r\rangle = 4x + 3y + 2(1 - x - y)$ is 2.4) in many different chemical compounds. Also, since T_g is one of the most important parameters for a description of the glassy state, much work has been done on ternary glasses to determine the dependence

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of T_g on composition [7]. For these systems, it has been observed by Sreeram and co-workers [3] that T_g follows a modified Gibbs–DiMarzio equation, which can be expressed as

$$T_g = \frac{T_{g0}}{1 - \beta(\langle r \rangle - 2)}, \tag{1}$$

where T_{g0} is the limit of T_g when the concentration of Ge, Sb, or As tends to zero, and β is a constant that depends on the system and which is fitted from experimental data [3]. The magnitude of β has not received much attention, although it is fundamental for determining T_g. To our knowledge, the first successful attempt that presents a theoretical model for determining β in binary glasses can be found in Refs. [8,9]. In this article we present a model of agglomeration and growth that can in a simple way predict β for ternary glasses. The advantage of using a model of growth for this propose, is that one can obtain information about some characteristic parameters of glass by explaining how it was formed, since we assume that both questions are intimately related.

In this article, the study of agglomeration in ternary glasses will use the recently introduced stochastic matrix method (SMM) [10,11]. The SMM is based on the assumption that glass is created when some basic entities (atoms or clusters) that are present in a liquid agglomerate to form bigger clusters as the temperature decreases. At any stage of this process, each cluster is divided into two parts, the interior and the surface. The elementary growth step is achieved by adding a new entity at a given site of the surface. Clearly, this process occurs with certain probability at each site of the surface depending on the physical parameters involved, such as the bonding energies, temperatures, and chemical composition of the liquid. Thus, since after the addition of a new entity to the cluster its surface changes, the distribution of sites on the surface evolves following these probabilities. The idea behind our method is to represent this change as a matrix that acts on a vector, which contains the 'state' of the surface at any stage of the agglomeration process. We shall show that the SMM always converges to a final configuration of probabilities for the glass, irrespective of the initial conditions. It also predicts an oscillatory behavior for the first steps of agglomeration. These oscillations are damped exponentially as the average cluster size grows. All these properties are derived from the behavior of eigenvectors and eigenvalues of the stochastic matrix.

2. The stochastic matrix method

In this section we shall construct the stochastic matrix for a ternary system, and we also show some of its properties. For this purpose, we observe that chalcogenide glasses grow out of a liquid which contains the basic entities, the local microscopic agglomerates of a few atoms or microclusters, which later on, as the temperature decreases, agglomerate and form the glass network. These basic entities can represent either single atoms or clusters that were already present in the liquid before the formation of the glass. For example, in the case of a binary glass such as amorphous Sb_2Se_3, the intramolecular bonds of Sb_2Se_3 remain in the liquid state [12]. A similar phenomenon can be observed in amorphous As_2Se_3, and thus, the ternary $As_xSb_ySe_{1-x-y}$ glass can be viewed as a system formed of Sb_2Se_3 and As_2Se_3 structural units, diluted in the extra amount of Se atoms [5] in the chalcogen-rich region ($\langle r \rangle < 2.4$). Another example is the As_xSe_{1-x} compound, where the As atoms are paired via an intermediate Se (as is shown in Fig. 1), and thus, the effective coordination number of As as a cross linking agent of Se chains is *four* instead of three [6]. On the other hand, it is known that in the Ge_xSe_{1-x} compound, Ge atoms do not form this kind of microcluster [6].

Taking into account all these considerations, we shall assume that the liquid which forms $Ge_xAs_ySe_{1-x-y}$ contains three types of basic entities: clusters of two As atoms joined by one Se atom, and Ge, Se atoms, which will be labelled b, c and a units correspondingly, their respective concentrations being C_b, C_c and C_a.

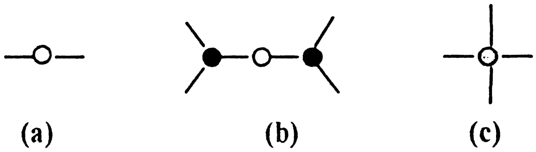

Fig. 1. Basic units (a) Se atoms, (b) As–Se–As clusters and (c) Ge atoms.

These concentrations of basic entities are related with those of the atomic species by the following equations:

$$C_c = \frac{x}{1-y}, \qquad C_b = \frac{y}{2(1-y)}, \qquad C_a = 1 - C_b - C_c. \tag{2}$$

For the $\mathrm{Ge}_x\mathrm{Sb}_y\mathrm{Se}_{1-x-y}$ compound the same relations hold, except that C_b represents the concentration of two Sb atoms joined by a Se atom.

With these three basic entities forming covalent bonds between them, there are five elementary processes of single bond creation. Each process involves its characteristic energy of activation for creating the bond between two units; these energies will be denoted by E_1, E_2, E_3, E_4 and E_5 for *a–a*, *a–b*, *a–c*, *b–b*, *b–c* and *c–c* bonds, respectively.

During the cooling process, clusters of different sizes appear as a result of covalent bond formation between the smaller entities that are already present in the liquid, and when a new basic entity comes close to a cluster, it can stick to one of the free valencies available on the surface. Since the coordination numbers of *a*, *b*, and *c* units are 2,4 and 4, respectively, the new entity may encounter seven types of configurations, as shown in Fig. 2 (observe that in Fig. 2 it is supposed that the two or three membered rings do not appear, i.e., the growth is dendritic; this assumption allows us to decrease the size of the stochastic matrix but it is valid only in the chalcogen-rich region).

We shall refer to different configurations that appear on the surface of a cluster calling them *sites*. For example, a *c* unit can appear on the surface,with one, two or three free valencies. These configurations are identified as three different sites denoted, respectively, by u, v and w. The *b* units with one, two and three free valencies give rise to the sites t, s, r, respectively. A single Se atom belonging to a cluster can have only one free valency, which represents a site called q. Using all these notations, the state of the surface is specified by the vector $\mathbf{v} = (q, r, s, t, u, v, w)$ whose components are the *relative frequencies of each kind of site*; therefore its trace is normalized to one: $q + r + s + t + u + v + w = 1$. The main task of the SMM is to determine the evolution of this vector as the clusters continue to grow.

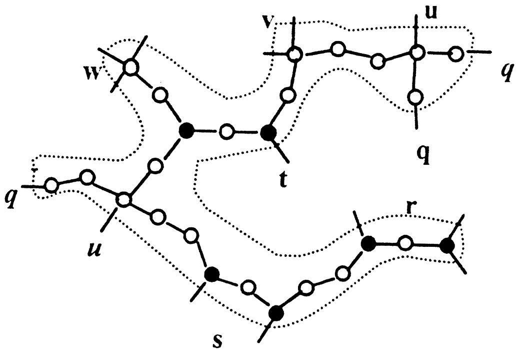

Fig. 2. A typical cluster with seven types of sites on the surface.

We shall represent the growth of clusters divided into elementary steps, which occur when a new entity is attached to any of the free valencies available on the surface. The probability of joining the new entity to each of the sites depends basically on two factors, the frequency of the given kind of site on the surface, represented by the corresponding component of the vector **v**, and the probability of the sticking process between the new entity and the surface, which depends on physical parameters such as temperature and bond activation energy. Once the new entity is attached to a site, a new site on the surface is created. For example, if we add an *a* unit to a *q* site, a new *q* site is created. Thus, the frequency of *q* sites in the new layer is given by

$$q' = P(q,q)q, \tag{3}$$

where $P(q,q)$ is the probability of the process, denoted symbolically as

$$q + a \rightarrow q. \tag{4}$$

As it was said, $P(q,q)$ depends on physical parameters involved in the process of creation of an *a*–*a* bond. In this case, $P(q,q)$ is

$$P(q,q) \sim 2C_a \mathrm{e}^{-E_1/KT}, \tag{5}$$

where the probability of the sticking process is a product of two factors, one of which is purely statistic (the number of ways available for joining the 2 valencies of Se to one of the free bonds of a *q* site, multiplied by the concentration of the units of type *a* in the melt), the other one is the Boltzmann factor which takes into account the corresponding activation energy barrier necessary to form that bond. Similar expressions can be formed when a Ge, As–Se–As and Se are added to each of the sites. However, in the $\mathrm{Ge}_x\mathrm{As}_y\mathrm{Se}_{1-x-y}$ and $\mathrm{Ge}_x\mathrm{Sb}_y\mathrm{Se}_{1-x-y}$ systems, the Ge–Ge, Ge–As and Ge–Sb bonds are almost never observed, and thus the probability of these kinds of bonds is close to zero. This assumption allows us to reduce the number of possible transformations of sites according to the following scheme:

$$q + b \rightarrow r\colon\ P(q,r) \sim 4C_b \mathrm{e}^{-E_2/KT},$$

$$q + c \rightarrow w\colon\ P(q,w) \sim 4C_c \mathrm{e}^{-E_3/KT},$$

$$r + a \rightarrow q, s\colon\ P(r,q) = P(r,s) \sim 6C_a \mathrm{e}^{-E_2/KT},$$

$$s + a \rightarrow q, t\colon\ P(s,t) = P(s,q) \sim 4C_a \mathrm{e}^{-E_2/KT},$$

$$t + a \rightarrow q\colon\ P(t,q) \sim 4C_a \mathrm{e}^{-E_2/KT},$$

$$u + a \rightarrow q\colon\ P(u,q) \sim 2C_a \mathrm{e}^{-E_3/KT},$$

$$v + a \rightarrow q, u\colon\ P(v,q) = P(v,u) \sim 4C_a \mathrm{e}^{-E_3/KT},$$

$$w + a \rightarrow q, v\colon P(w,s) = P(w,v) \sim 6C_a \mathrm{e}^{-E_3/KT}.$$

The transformation of the surface of an average cluster is encoded in the matrix that acts on the vector **v**, because the total probability for an entity to stick to a certain site is equal to the probability of the sticking process multiplied by the frequency of occurrence of the corresponding type of site at the surface. The components of the matrix are the probabilities of transformation of each kind of site onto another. Inserting all the contributions we get the explicit matrix as follows:

$$M = \begin{pmatrix} 2C_a\mathrm{e}^{-E_1/kT} & 6C_a\mathrm{e}^{-E_2/kT} & 4C_a\mathrm{e}^{-E_2/kT} & 4C_a\mathrm{e}^{-E_2/kT} & 2C_a\mathrm{e}^{-E_3/KT} & 4C_a\mathrm{e}^{-E_3/KT} & 6C_a\mathrm{e}^{-E_3/KT} \\ 4C_b\mathrm{e}^{-E_2/kT} & 0 & 0 & 0 & 0 & 0 & 0 \\ 0 & 6C_a\mathrm{e}^{-E_2/kT} & 0 & 0 & 0 & 0 & 0 \\ 0 & 0 & 4C_a\mathrm{e}^{-E_2/kT} & 0 & 0 & 0 & 0 \\ 0 & 0 & 0 & 0 & 0 & 4C_a\mathrm{e}^{-E_3/KT} & 0 \\ 0 & 0 & 0 & 0 & 0 & 0 & 6C_a\mathrm{e}^{-E_3/KT} \\ 4C_c\mathrm{e}^{-E_3/KT} & 0 & 0 & 0 & 0 & 0 & 0 \end{pmatrix}.$$

This matrix acts on a vector whose components represent the probabilities of finding each type of site, and thus is normalized to one. The vector obtained after applying the matrix also must be normalized since it should represent a new distribution of probabilities. To assure this normalization, the sum of elements in each column of the matrix must be equal to one. After normalizing each column, we obtain the matrix in which only three entries are functions of the concentration and temperature, while all the others are constant:

$$M = \begin{pmatrix} A & \frac{1}{2} & \frac{1}{2} & 1 & 1 & \frac{1}{2} & \frac{1}{2} \\ B & 0 & 0 & 0 & 0 & 0 & 0 \\ 0 & \frac{1}{2} & 0 & 0 & 0 & 0 & 0 \\ 0 & 0 & \frac{1}{2} & 0 & 0 & 0 & 0 \\ 0 & 0 & 0 & 0 & 0 & \frac{1}{2} & 0 \\ 0 & 0 & 0 & 0 & 0 & 0 & \frac{1}{2} \\ C & 0 & 0 & 0 & 0 & 0 & 0 \end{pmatrix}. \tag{6}$$

The entries A, B and C are defined as

$$A = \frac{2C_a}{2C_a + 4(C_b\xi + C_c\eta)}, \qquad B = \frac{4C_b\xi}{2C_a + 4(C_b\xi + C_c\eta)}, \qquad C = 1 - A - B \tag{7}$$

and where, $\xi = \exp((E_1 - E_2)/kT)$, and $\eta = \exp((E_1 - E_3)/kT)$.

The consecutive agglomeration and growth of clusters is represented by successive application of the above matrix to the initial vector $\mathbf{v}_0$. After applying N times the matrix, the final configuration of the surface is

$$\mathbf{v}_N = \sum_{m=1}^{7} a_m(\lambda_m)^N \mathbf{e}_m, \tag{8}$$

where $\mathbf{e}_m$ are the eigenvectors of M corresponding to the eigenvalue λ_m, and a_m are the projections of $\mathbf{v}_0$ onto the eigenvectors of M.

It is easy to prove that a matrix with all the columns normalized to one has at least one eigenvalue equal to one, while all other (in general, complex) eigenvalues have their norm always less than one. This condition means that only the eigenvectors with eigenvalue one remain after many successive applications of the stochastic matrix. If we suppose that M has a single eigenvalue, 1 (corresponding to $\lambda_1 = 1$), then, in the limit of big N, $\mathbf{v}_N$ converges to

$$\mathbf{v}_N = \mathbf{e}_1 \tag{9}$$

since a_1 must be one due to conservation of probability. Thus, the surface attains stable statistical regime after many successive steps of agglomeration, with the statistic given by the eigenvalue 1 eigenvector. The explicit form of this eigenvector 1 is obtained by solving the system of equations:

$$(M-1)\mathbf{e}_1 = 0, \tag{10}$$

which in our particular case yields the following solution:

$$\mathbf{e}_1 \equiv \begin{pmatrix} q_\infty \\ r_\infty \\ s_\infty \\ t_\infty \\ u_\infty \\ v_\infty \\ w_\infty \end{pmatrix} = \frac{1}{13-7A} \begin{pmatrix} 4 \\ 4B \\ 2B \\ B \\ C \\ 2C \\ 4C \end{pmatrix}. \tag{11}$$

Before the process attains the stable regime, oscillations can be observed in first generations, due to the eigenvalues different from 1, which are in general complex. This fact can be seen if in Eq. (8) we rewrite λ_m in its polar form ($\lambda_m = \|\lambda_m\| \mathrm{e}^{\mathrm{i}\theta(m)}$),

$$\mathbf{v}_N = \mathbf{e}_1 + \sum_{m=2}^{7} a_m \|\lambda_m\|^N \mathrm{e}^{\mathrm{i}N\theta(m)} \mathbf{e}_m = \mathbf{e}_1 + \sum_{m=2}^{7} a_m \mathrm{e}^{N\ \ln\|\lambda_m\|} \mathrm{e}^{\mathrm{i}N\theta(m)} \mathbf{e}_m. \tag{12}$$

Then, the factor $\mathrm{e}^{\mathrm{i}N\theta(m)}$ represents an oscillation, which is exponentially damped by the norm of λ_m, which acts as a characteristic time of damping for each eigenvector. (Note that $\ln\|\lambda_m\| < 0$ because $\|\lambda_m\| < 1$.)

The statistical distribution of various sites on the surface of an average cluster enables us to evaluate the final concentration of the corresponding elementary building blocks in the resulting interior matter. This concentration need not be in principle the same as in the surrounding medium, especially if the agglomeration process takes place in a vapor or from a solution. But in the case of glass transition one should suppose that the concentration of chemical species remains exactly the same as in the liquid at a temperature above the glass transition temperature, because if the contrary were true, one would observe noticeable local departures from the homogeneity (fluctuations of density and chemical composition), which is clearly not the case in real glasses.

This constatation enables us to introduce a new constraint in the model, which yields a very useful equation. Let us denote by X, Y, Z the asymptotic values of the respective concentrations of c, b and a-type units on the surface of an average cluster. Then, the concentration of new c units after the creation of a new layer is given by $x = w_\infty$ (observe that in order not to count the c unit twice, u and v sites do not contribute to x, since the creation of these sites requires more than one step of agglomeration; we count only the first step which always leads to the formation of a w site). For free Se atoms a similar condition holds, $Z = q_\infty$. This leads to the following equation when the values of q_∞ and w_∞ are taken from Eq. (11):

$$\frac{Z}{X} = \frac{q_\infty}{w_\infty} = \frac{2C_a + 4C_b\xi + 4C_c\eta}{4C_c\eta}. \tag{13}$$

If we also demand the concentration of b units to attain its limit, $Y = r_\infty$ we get another equation,

$$\frac{Z}{Y} = \frac{q_\infty}{r_\infty} = \frac{2C_a + 4C_b\xi + 4C_c\eta}{4C_b\xi}. \tag{14}$$

The requirement that the asymptotic values of the concentrations X, Y and Z in the resulting bulk network be the same as in the surrounding hot liquid before the glass transition results in the following three equations:

$$X = C_c, \qquad Y = C_b, \qquad Z = C_c, \tag{15}$$

which means that during the glass transition the stationary régime is established, i.e., that the mappings, $C_c \to X(C_b, C_c, T)$ and $C_b \to Y(C_b, C_c, T)$ attain one of their *fixed points*. It is easy to determine that these conditions can be satisfied by two solutions; $C_b, C_c = 0$ (corresponding to a pure Se glass) and another fixed point, which is obtained when the following two equations are verified:

$$2C_a + 4C_b\xi + 4(C_c - C_a)\eta = 0, \tag{16}$$

and

$$2C_a + 4(C_b - C_a)\xi + 4C_a\eta = 0. \tag{17}$$

The last two equations can be also written in terms of the original concentration of atoms by using Eq. (2),

$$2\left(1 - x - \frac{3}{2}y\right) + 4\left(\frac{y}{2}\right)\xi + 4\left(2x + \frac{3}{2}y - 1\right)\eta = 0, \tag{18}$$

and

$$2\left(1 - x - \frac{3}{2}y\right) + 4(2y + x - 1)\xi + 4x\eta = 0. \tag{19}$$

As we show in Section 3, these equations lead to important relations between the glass transition temperature, concentration of atomic species and differences of various activation energies.

3. Discussion

The example we consider here contains two parameters, ξ and η, which depend on the energy difference between the two activation processes. We can fix these values by considering the limit cases of two binary glasses, i.e; when $y = 0$ we have the amorphous Ge_xSe_{1-x}, and when $x = 0$ we may have either As_ySe_{1-y} or Sb_ySe_{1-y}.

If $y = 0$, we get from Eq. (18),

$$2(1 - x) - 4\eta = 0. \tag{20}$$

In a good glass former such as Ge_xSe_{1-x}, an amorphous homogenous configuration is easily obtained with arbitrarily small modifier concentration (in this case Ge); the corresponding glass transition temperature in this limit is denoted by $T_{og} = T_g(x = 0)$. Using Eq. (20) we get η at T_{og},

$$\eta(T_{og}) = \frac{1}{2} \tag{21}$$

and thus we fix the first energy difference using the definition of η,

$$E_1 - E_3 = kT_{og} \ln(1/2). \tag{22}$$

Therefore,

$$E_1 - E_2 = kT_{og} \ln(1/2) < 0, \text{ i.e., } E_3 > E_1.$$

This situation is what should be intuitively expected from the strong glass-forming tendency: at a local level the system behaves undecidedly, in a 'frustrated' way, because while the purely statistical factor (4 vs. 2) increases the probability of agglomeration of modifier's atoms (of the *c*-type, with valency 4) versus the probability of pairing of the pure glass-former atoms (of the *a*-type, here with lower valency 2), the Boltzmann factors act in the opposite direction. The value of T_{og} is known from experiments [3] to be $T_{og} = 316°$ K, giving the difference between activation energies of the order of -0.02 eV.

A similar analysis performed on Eq. (19) when $x = 0$ leads to

$$\xi(T_{\mathrm{og}}) = \eta(T_{\mathrm{og}}) = \frac{1}{2}. \tag{23}$$

At any other temperature, η and ξ are obtained from the following identity:

$$\eta = \xi = \mathrm{e}^{(E_1 - E_3)/kT_{\mathrm{g}}} = \mathrm{e}^{T_{\mathrm{go}}\ \ln(1/2)/T_{\mathrm{g}}} = \left(\frac{1}{2}\right)^{T_{\mathrm{go}}/T_{\mathrm{g}}}. \tag{24}$$

Once the parameters are fixed, we put Eq. (24) into Eq. (18) to find the relation between T_{g} and the composition parameters,

$$\left(\frac{1}{2}\right)^{T_{\mathrm{go}}/T_{\mathrm{g}}} = \frac{(1 - x - \frac{3}{2}y)}{2(1 - 2x - 2y)}, \tag{25}$$

after taking the logarithm, we get

$$\frac{T_{\mathrm{og}}}{T_{\mathrm{g}}} \ln\left(\frac{1}{2}\right) = \ln\left(1 - x - \frac{3}{2}y\right) - \ln(1 - 2x - 2y) - \ln\left(\frac{1}{2}\right). \tag{26}$$

In the chalcogen-rich region, $x, y \ll 1$, and we can use Taylor's expansion of the logarithm, which leads to

$$T_{\mathrm{g}} = \frac{T_{\mathrm{og}}}{1 - \frac{1}{2\ \ln\ 2}(2x + y)}, \tag{27}$$

and finally, using that $2x + y = \langle r \rangle - 2$, we find

$$T_{\mathrm{g}} = \frac{T_{\mathrm{og}}}{1 - \frac{1}{2\ \ln\ 2}(\langle r \rangle - 2)}. \tag{28}$$

The last equation is exactly in the form of the Gibbs–DiMarzio law, which describes quite correctly the behavior of the glass transition temperature in many ternary glasses. Furthermore, the direct comparison between Eq. (28) and Eq. (1) shows that the system parameter, β, is given by

$$\beta = \frac{1}{2\ \ln\ 2} = 0.72.$$

Experimentally, the observed β is 0.75 for $\mathrm{Ge}_x\mathrm{Sb}_y\mathrm{Se}_{1-x-y}$ [3] and 0.73 for $\mathrm{Ge}_x\mathrm{As}_y\mathrm{Se}_{1-x-y}$ [4]; both β's are in agreement with the β obtained here via SMM. At this point it is interesting to ask about the origin of the magnitude of β. If we trace back its definition, we observe that it came from fixing η and ξ at T_{og}, and thus depends on the logarithm of the ratio between the coordination number of the elementary units. This fact can explain why its magnitude is so similar for systems with very different types of atoms.

4. Conclusion

In this paper we studied ternary covalent network glasses using the SMM. An analytical relation between relevant physical parameters of several chalcogenide based glasses has been found. The corresponding formula is similar to the Gibbs–Di Marzio law. We also obtained the system parameter β that is close to the one observed in the experiments. Its value depends on the ratio between the coordination numbers of the elementary units.

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