Relaxation phenomena in the glass transition

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A review work on some of the most important aspects on relaxation phenomena that occur both in fragile and strong glass-formers is presented. In particular, different empirical forms for the logarithmic shift factor and its relation with the specific heat that have been studied in the literature are discussed. The application of the generalized stochastic matrix method in strong glasses and the behavior of the relaxation times are indicated. Special attention is given to the α and β relaxation processes and the crossover between the different dynamical regimes that may be recognized in the vicinity of the glass transition.

Keywords: Glass transition; relaxation phenomena; transport coefficients.

Se presenta un trabajo de revisión acerca de algunos de los más importantes fenómenos de relajamiento que ocurren tanto en vidrios frágiles como fuertes. En particular, se discuten diferentes formas empíricas para el factor de corrimiento logarítmico que han sido reportadas en la literatura y su relación con el calor específico. En el caso de los vidrios fuertes, se indica la aplicación del método generalizado de las matrices estocásticas y el comportamiento de los tiempos de relajamiento. Se presta una atención especial a la fenomenología de los procesos de relajamiento α , y β y al cruzamiento de los diferentes regímenes dinámicos que se reconocen en la vecindad de la transición vítrea.

Descriptores: Transición vítrea; fenómenos de relajamiento; procesos de transporte.

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1. Introduction

The relaxation studies in supercooled liquids and glasses represent nowadays one of the most important fields in condensed matter science [1–3], particularly in polymer [4, 5], ionic [6] and metallic glasses [7], both from the theoretical and experimental points of view. At present, there is no agreement on a well accepted theory to explain the different kinds of behaviors of the glass forming materials in the neighborhood of the glass transition temperature [8–10]. Furthermore, this task has become cumbersome due to the new techniques of measurement which have increased the capability to measure relaxation times, diffusion coefficients and characterization of the materials. Thus, one has to deal with a large number of facts in order to construct a good theory. Some of these new spectroscopic methods are listed in Table I.

The most significant features on approaching the glass transition are the rapid increase in the viscosity and the slowing down of the structural relaxation [11, 12]. From the theoretical point of view, many efforts have been made to elucidate the temperature dependence of the relaxation time and other properties of several glass formers [13–17].

Along these lines, the purpose of this work is to present a review on the state of the art of several important aspects concerning the phenomenology of the glass transition both in the case of fragile and strong glass formers, and to try to summarize some of the most important experimental and theoretical results that have been reported in the last few years in the literature [18–25].

In section 2 we present a brief account on the kinetic and thermodynamic aspects of the glass transition. Section 3 is devoted to the presentation of the different forms that have been proposed to describe the logarithmic shift factor in the glass transition and its relation with the specific heat. In section 4 we present the different dynamical processes that may be recognized in the vicinity of the glass transition and the different results that may be found in strong and fragile supercooled liquids. In order to present a brief overview on some microscopic aspects such as the mode coupling theory, and experiments and computer simulations of a "glass-like" transition in colloidal suspensions, we have included in section 5 results that have been reported recently in the literature. Finally in section 6 we outline some important concluding remarks.

TABLE I. Different spectroscopic techniques and the corresponding relaxation time intervals that may be attained (M. D. Ediger, C. A. Angell and S. R. Nagel, *J. Phys. Chem.* **100** (1996) 13200, and references therein).

Spectroscopic technique	Relaxation times
Dielectric relaxation	10^{-11} to 10^4 s
Dynamic light scattering	10^{-12} to 10^2 s
Dynamic neutron scattering	10^{-12} to 10^{-7} s
Computer simulations	10^{-14} to 10^{-7} s
Optical methods	10^{-14} to 10^{-7} s
NMR	10^{-11} to 10^3 s

2. Kinetic and thermodynamical aspects of the glass transition

Regarding the kinetic aspects of the glass transition, the transformation of the supercooled liquid into an amorphous solid is a time dependent phenomenon. We have as a wellestablished fact that the viscosity of a liquid increases in a spectacular way when the liquid is supercooled below its melting point, avoiding crystallization, reaching values of about 10^{13} Poise. Below this glass transition point T_q the glassy state is reached. One of the most common methods to measure T_q is based on the change of the heat capacity. On the other hand, the value of T_q depends on the cooling rate, the controlling parameter that defines the transition. The laboratory glass point temperature T_g is identified with the temperature at which the relaxation time equals a characteristic value [13], usually taken to be $\langle \tau(T_q) \rangle = \langle \tau_q \rangle = 100 \text{ s}$ [26]. Furthermore, as a supercooled liquid attains temperatures below the minimum equilibrium temperature, T_m , the melting point temperature, the viscosity increases and molecules move very slowly attaining the equilibrium state before the temperature is lowered further. In each cooling step, the properties of the supercooled liquid deviate from the equilibrium values [14] and finally, the thermodynamic and mechanic properties of the glass depend upon how it was formed and its thermal history [27]. This was pointed out by Davies and Jones [28], Goldstein [29], Johari [30] and Heijboer [31].

As the system is supercooled, it remains in the liquid phase until the glassy state is reached at T_g . If the experimental curve for the specific heat is extrapolated for temperatures below T_g , it would seem that the specific heat of the glass would be larger than the corresponding value for the crystal. This represents a violation to Nernst's principle. Thus one must postulate the existence of a temperature T_K where the entropy of a glass would eventually have the same entropy as the corresponding crystal. This paradox was pointed out by Kauzmann [32, 33]. This fact is closely related to the need for a equilibrium thermodynamic framework to insert the description of the glass transition [34–39].

3. The logarithmic shift factor and the specific heat

Although the nature of the relaxation processes that regulate the behavior of a supercooled liquid in the glass transition vicinity has not been well established[13], several experiments that probe different response functions of the liquid by means of diverse techniques indicate that these functions may be successfully described by a non-exponential decaying function, namely, the Kohlrausch-Williams-Watts (KWW) relaxation function [40–48],

$$\phi(t) = \phi_0 \exp\left\{-\frac{t}{\tau}\right\}^{\beta},\tag{1}$$

where τ (t=0) = τ_0 , τ is the average relaxation time and β is the stretching exponent. According to the fitting of the experimental results, the KWW describes well the behavior of the relaxation phenomena for long times. In the long time scale, the fluctuations are of slow character and induce changes in the correlation times in the cell-domains. Therefore, a distribution in the correlation times is produced [49]. In that case, the KWW is obtained as a superposition of the local exponential relaxation

$$\phi(t) = \sum_{n} C_n \exp\left\{-\frac{t}{\tau_n}\right\},\tag{2}$$

where C_n is the weight factor and represents the contribution of the subset of all the cell-domain with the correlation time τ_n .

As we have already mentioned, the glass transition embodies several kinetic processes. As the liquid is supercooled, the variation of the relaxation time with temperature generally presents a non-Arrhenius behavior [50]. The departure from an Arrhenius form in the liquid behavior is related to the so called "fragility" [51]. Depending on the structural strength of the system different empirical forms may be fitted to the experimental relaxation times. The temperature dependence of the relaxation time, or equivalently of the viscosity, has been studied both empirically and theoretically for different kinds of glass forming liquids by several authors [52–54] [55, 56], and may be related with changes in the specific heat.

One of the most used forms is the Vogel-Fulcher-Tammann (VFT) equation [57, 59], in terms of the so called logarithmic shift factor (LSF), $\log a_T$, given by,

$$\log a_T = \log \frac{\tau}{\tau_0} = \frac{B}{T - T_0},\tag{3}$$

where B>0, τ_0 is a reference relaxation time and T_0 is the temperature at which τ goes to infinity, and thus where any configurational change in the system becomes extremely difficult. This equation may be rewritten as [51],

$$\log a_T = \frac{DT_0}{T - T_0},\tag{4}$$

where D is a constant that represents a measure of the structural strength of the liquid and accounts for the deviation

of the Arrhenius (ARR) law that may be obtained if we set $T_0 = 0$,

$$\log a_T = \left\{ \frac{E_A}{T} \right\}. \tag{5}$$

Another empirical relation to describe the LSF was proposed by Williams, Landel and Ferry (WLF) [60], namely,

$$\log a_T = \frac{-C_1(T - T_s)}{C_2 + T - T_s},\tag{6}$$

here, T_s represents a reference temperature and C_1 and C_2 are taken as two constants that have the same value for a great number of glass forming liquids [61,62]. Both the VFT and WLF equations lie on the values of the adjustable parameters that reproduce in each case the best possible fit of the experimental data.

The behavior of different kinds of glass forming liquids has been classified by Angell and several other authors in terms the form of the LSF and its departure of an ARR behavior in terms of the fragility [63-69]. One may find liquids that follow a conventional ARR law, mainly many metallic oxides as SiO_2 , GeO_2 and B_2O_3 ; intermediate liquids such as alcohols and glycerol, that deviate slightly from ARR; and finally liquids that drastically deviate from this behavior, such as several organic liquids as glucose, lactose and polymers. This latter kind of glasses follow the VFT empirical relation. Experimentally, the parameter that indicates the deviation from ARR lies within the interval $5 \le D \le 100$. The largest D ($D \approx 20 - 100$) values characterize those liquids which are most resistant towards temperature induced changes. These liquids display a nearly ARR behavior and are referred to as "strong" [63]. To this group belong covalent bonded network glass formers like SiO_2 (D=100) [63] and $B_2O_3(D=35)$ [70]. At the other extreme are the "fragile" glasses, those systems with the smallest D values ($D \approx 3-5$) exhibiting the most rapid changes of the relaxation time in the supercooled region. For the fragile systems these changes in the transition range imply that T_0 is close to T_g , while for strong ones T_0 is far below $(T_g/T_0 \approx 2 \text{ for } B_2O_3)$. Another way to evaluate the fragility is by defining the fragility index [62].

$$m = \left[\frac{d\log\tau}{d(T_g/T)}\right]_{T=T_g} \tag{7}$$

This slope depends on D, thus it has a small value in the case of fragile glasses and its value increases in the case of strong ones. The fragility index pattern allows us to discover important slope changes in the activation energy when the temperature gets close to the glass transition temperature.

The relation between the characteristic parameters of the VFT equation and the stretching exponent β of the KWW equation has been proposed by Hodge [25] where β depends on the difference between T_g and T_0 . On the other hand, Bohmer et al.[51] found a relation between the fragility index and the stretching exponent, namely,

$$m = 250 - 320\beta. (8)$$

From a theoretical point of view one of the most important works to present a model to find the dependence of the LSF on temperature was presented in 1965 by Adam and Gibbs [71] based on a microscopic model of the glass transition for polymeric glasses proposed by Gibbs and Di Marzio [72]. This model predicts the existence of a temperature T_0 for which the configurational entropy S_c vanishes, $S_c(T_0) = 0$. Adam and Gibbs propose a molecular kinetic theory to evaluate the temperature dependence of the relaxation phenomena in the glass transition in terms of the probabilities of cooperative rearrangements which depend on the configurational entropy of the system, that may be evaluated in terms of the specific heat of the liquid. Since the relaxation time is reciprocally related to these probabilities, they present a form to evaluate the LSF given by,

$$-\log a_T = 2.303 \left[\Delta \mu s_c^* / k_B\right] \left(\frac{1}{T_s S_c(T_s)} - \frac{1}{T S_c(T)}\right). \quad (9)$$

 $\Delta \mu s_c^*/k_B$ is evaluated from the model and the configurational entropy is calculated in terms of the change of the specific heat, that is,

$$\Delta S_c = S_c(T) - S_c(T_0) = \int_{T_0}^{T} \frac{\Delta c_p}{T} dT.$$
 (10)

In their work, they consider the approximation in which

$$\Delta c_p = const. \tag{11}$$

which leads to an expression for the LSF that may be compared with the WLF form. Besides, if they choose T_g as the reference temperature, they can evaluate the isoentropic temperature T_0 .

Several authors have explored different kinds of possibilities to include variations for the original idea of Adam and Gibbs [73–79]. Angell and Sichina [64] removed the assumption on the constancy of the C_p around the glass transition and propose,

$$\Delta c_p = \frac{const.}{T},\tag{12}$$

and they obtain the VFT equation.

In the case of polymeric glasses, García-Colín, del Castillo and Goldstein [80] use the form for the specific heat proposed by di Marzio and Dowell [81], based on the Gibbs-di Marzio model, namely,

$$\Delta c_p = A \frac{1}{T^2} + BT - CT^2. \tag{13}$$

In this case, on one hand, the LSF may be expressed in the VFT form,

$$\log a_T = \frac{K}{F(T)(T - T_0)},$$
(14)

where F(T) is a function of temperature, K is a constant that depends on the cooling rate [82–84] and K/F(T) remains constant in the region $T > T_g$. On the other hand, the LSF may also be written as an expression in terms of $1/T^2$ given by [85],

$$\log a_T = -K \left\{ \frac{1}{F(T_s)(T_s - T_0)} - \frac{1}{G(T)} \frac{1}{T^2} \right\}, \quad (15)$$

where G(T) is another function of T. This form may be directly compared with the WLF equation, and the isoentropic temperature T_0 is found through this comparison. Both forms for the LSF Eqs. (14) and (15) are parameter free expressions and are in agreement with other results presented in the literature [86]. Furthermore, Dagdug and García-Colín [87] generalize these results and are able to include non-polymeric fragile glass forming liquids in their description. These results have been confirmed in a recent work [88].

One may also find in the literature some works that present directly different fits for the experimental values of the relaxation times or the viscosity such as the superArrhenius forms proposed by Kivelson et al. [89–91].

4. Relaxation processes in the glass transition

At this point we have discussed the behavior of the relaxation times for very slow diffusion processes. This kind of processes have been defined as α -relaxation processes. In the past few years, fast dynamical relaxation processes which occur in the vicinity of T_g , referred as β -relaxation processes, have been studied. Several authors have found in different sorts of experiments solid evidence that many properties of the supercooled liquid, specially diffusion mechanisms, present drastic changes around a cross-over temperature T_c , whose values lie between 1.15 T_g and 1.28 T_g [92–94].

One of the most known evidences of this crossover is the fact that in the range of temperatures below T_c the VFT equation does no longer fit the experimental values for the relaxation times nor the viscosity. In order to probe the validity of the VFT equation, Stickel, Fischer and Richert [95, 96] proposed the so called temperature derivative analysis for the quantities,

$$x = \{f/Hz, \sigma_{dc}\varepsilon_0/s^{-1}, \eta^{-1}/poise^{-1}\},$$
 (16)

where f is the maximum frequency, $f = \tau^{-1}$, σ_{dc} is the dc conductivity, and η is the viscosity. The method consists in the evaluation of three derivatives for a given empirical form for $\log x$,

$$\left[\frac{d\log_{10}x}{dT}\right]^{-\frac{1}{2}},\tag{17}$$

$$\frac{d}{dT} \left[\left(\frac{d \log_{10} x}{dT} \right)^{-\frac{1}{2}} \right],\tag{18}$$

$$\Theta = \frac{\frac{d \log_{10} x}{dT}}{\frac{d^2 \log_{10} x}{dT^2}},\tag{19}$$

The function Θ gives us a specially useful mathematical tool to analyze different regimes in the vicinity of T_c [97]. If these derivatives are applied to the VFT equation, the results may be written as,

$$\left[\frac{d\log_{10}x}{dT}\right]^{-\frac{1}{2}} = B^{-\frac{1}{2}}(T - T_0),\tag{20}$$

$$\frac{d}{dT} \left[\left(\frac{d \log_{10} x}{dT} \right)^{-\frac{1}{2}} \right] = B^{-\frac{1}{2}},\tag{21}$$

$$\Theta = \frac{\frac{d \log_{10} x}{dT}}{\frac{d^2 \log_{10} x}{dT^2}} = -\frac{T - T_0}{2}.$$
 (22)

Using this technique, one may decide whether a set of experimental data obey the VFT form or a different empirical form must be proposed.

4.1. Strong glasses. The case of B₂O₃ and the stochastic matrix method

In the case of strong glasses, Dagdug and García-Colín [98–100] have derived a theoretical VFT equation for the average relaxation time, or viscosity, of the strong glass forming liquid B_2O_3 introducing a probabilistic temperature. The main idea of this work is to take the average relaxation time as inversely proportional to the probability of forming a boroxol ring, evaluated using the stochastic matrix method, calculated for a large number of steps of growth and to identify the type of the obtained equation using the temperature derivative method.

Through the stochastic matrix method (SMM) it is possible to describe the growth process of a solid [101]. In this method such process can be described by matrix acting on a vector. The matrix components are the probabilities of finding a given site at the border (rim) of a glass cluster of a certain size. The vector components represent the probabilities of finding a given site on the rim of such cluster. The matrix transforms this vector onto a new one after adding one atom to the cluster. The transformation of the rim depends on the site on which the new atom sticks to. Each sticking process has a certain probability of occurring, so that the matrix elements contain the probabilities of transforming each kind of site into others. The probability factors must include two contributions: (1) The statistical weight for each process, that is the number of ways leading to the same final result, and (2) the Boltzmann factor taking into account the energy barrier necessary to form a certain kind of bond. The probability of sticking a new unit in the bulk at any temperature T is taken to be proportional to $\exp(-E_i/k_BT)$, where E_i is the energy cost of sticking a unit in the i form at temperature T. Due to the fact that below T_q the glass system is unable to displace any unit to stick it in the bulk, a temperature T' is introduced such that the probability to stick a unit in the bulk may be zero. Thus, the SMM may be generalized so that the probability of sticking a new unit at the rim is proportional to $\exp(-E_i/k_B(T-T'))$. To identify T' as a physical property of the system, the relaxation time for the growth of the system is calculated as inversely proportional to the probability of forming a ring,

$$\tau \propto \frac{1}{P(\xi)},$$
 (23)

where,

$$\xi = \exp\left[\frac{\Delta E}{k_B(T - T')}\right],\tag{24}$$

and $\Delta E = E(T) - E(T')$.

In the particular case of B_2O_3 the probability of forming a ring when passing form the jth layer to the (j+1)th one, is simply given by counting the proportion of rings that were formed between the step j and the step j+1. If it is calculated for a large number of steps of growth, P_B^j can be replaced by its limiting value which according to Barrio *et. al.* [102] for B_2O_3 this is given by,

$$P_B^{\infty} = \frac{24\xi^2 + 16\xi}{84\xi^2 + 107\xi + 25}.$$
 (25)

The transition probability may be identified as the probability of forming a ring for the B_2O_3 . Furthermore, since the viscosity is proportional to the relaxation time, and the α -relaxation time is inversely proportional to the average transition probability, then

$$\tau \propto \frac{1}{P_R^{\infty}} = \frac{84\xi^2 + 107\xi + 25}{24\xi^2 + 16\xi}.$$
 (26)

Taking the derivatives $d^n \log x/dx^n$, and $d \log x/d(1/T)$ (n=1,2) of equation (26) where $x=\tau$

$$\left[\frac{d\ln(P_B^{\infty})}{dT}\right]^{-\frac{1}{2}} = (T - T_0) \left[\frac{E_2 - E_1}{k_B}\right]^{-\frac{1}{2}} L_{B_2O_3}^{-1/2}, \quad (27)$$

$$\left[-\frac{d\ln(P_B^{\infty})}{dT^{-1}} \right]^{-\frac{1}{2}} = \left(1 - \frac{T_0}{T} \right) \left[\frac{E_2 - E_1}{k_B} \right]^{-\frac{1}{2}} L_{B_2O_3}^{-1/2}, \tag{28}$$

where

$$L_{B_2O_3} \equiv \left[\xi \frac{d \ln P(\xi)}{d\xi} \right] = \frac{48\xi^2 + 16\xi}{24\xi^2 + 16\xi} - \frac{168\xi^2 + 107\xi}{84\xi^2 + 107\xi + 25}. \quad (29)$$

Taking typical values for the activation energy as quoted in Ref. [70], if $\xi \ll 1$, $L_{B_2O_3} \approx 1$, one can obtain,

$$\Theta = -\frac{T - T_0}{2}. (30)$$

If equations (27)-(29) are integrated, a theoretical VFT-like equations is obtained, namely

$$\tau = \tau_0 \exp\left(\frac{E_2 - E_1}{k_B(T - T_0)}\right) = \tau_0 \exp\left(\frac{DT_0}{T - T_0}\right), \quad (31)$$

where the constant τ_0 is the preexponential factor and D is a constant equal to $(E_2-E_1)/k_BT_0$ that can be determined comparing it with the experimental VFT equation to reproduce the experimental data for B_2O_3 The values $D\approx 35$, $T_0\approx T_g/2$ [103] and $\tau(T_g)=10^{13}{\rm P}$ to calculate τ_0 .

Using these values the activation energy turns out to be $E_2 - E_1 = 18.207 kcal mol^{-1}$ [104].

4.2. Fragile glasses. The crossover of regimes and the diffusion processes

As far as fragile glasses are concerned, experimental evidence of a crossover between two regimes is reported. One of the main features of this behavior is exhibited by the fact that a large number of fragile glasses follow the VFT equation only for temperatures above T_c . Another important evidence is the change in the diffusion mechanisms in the system for temperatures below T_c .

For temperatures below T_c , although the experimental values for $\log x$ seem to satisfy Eq. (3), the derivatives given by Eqs. (17)-(19) do not exhibit the form for VFT, namely Eqs., (20)-(22). One may thus identify the value of T_c below which the VFT equation is no longer a good fit. Goldstein, García-Colín and del Castillo [105] have analyzed the behavior of three fragile glass forming liquids, namely, phenyl salicylate (salol), phenolphthaleine-dimethyl-ether (PDE) and orthoterphenyl (OTP). Using experimental data, a mathematical form for the logarithm of the viscosity is presented in the region between T_g and T_c which does not correspond to a VFT equation, and for temperatures above T_c a form that corresponds to a VFT equation.

In the case of the $T_g < T < T_c$ region, an empirical equation to fit the experimental data of the form,

$$\log \frac{f(T)}{f(T^*)} = \log \frac{\eta(T^*)}{\eta(T)} = A(T_a - T)^2 + B,$$
 (32)

is proposed for the three glasses. For the $T>T_c$ interval, the experimental data do fit a VFT equation, namely,

$$\log \frac{\eta(T^*)}{\eta(T)} = C - \frac{E}{T - T_0}.$$
 (33)

A small region in the vicinity of T_c where both descriptions overlap may be found. In fact, one may find a temperature T_c' where Eqs. (32) and (33) intersect each other. This temperature is taken as the value where the derivatives Θ for both descriptions are intersected, that is,

$$\Theta_{T < T_c}(T_c') = \Theta_{T > T_c}(T_c'). \tag{34}$$

These temperatures for the three glass formers are in good agreement with the experimental values of T_c .

An important number of experiments on the properties of tracer diffusion in supercooled fragile glass forming liquids have been carried on [106–112] and several theoretical models [113] have been proposed which indicate that the traditional Stokes-Einstein relation between the translational diffusion coefficient and the viscosity,

$$D = \frac{k_B T}{6\pi d} \eta^{-1},\tag{35}$$

with d, the effective hydrodynamic particle radio, breaks down in the $T_g < T < T_c$ region, that is, the relation which holds in this regime between both transport coefficients is,

$$D = \frac{k_B T}{6\pi d} \eta^{-\xi},\tag{36}$$

where ξ is a coupling exponent, that depends on the tracer particle, with values in the interval $0.75 < \xi < 0.95$. The exponent ξ may be interpreted as a fractionary scaling parameter in the region where the glass former switches from a translational-rotational coupling regime in the α -relaxation region, to an uncoupled regime in the β -relaxation interval. Goldstein et al. analyzed the relationship between the values of the viscosity of salol, PDE and OTP in both the $T < T_c$ and the $T > T_c$ regions and the experimental values for the diffusion coefficients of tracer particles, and find that, in the case of temperatures below T_c , $\xi < 1$, exhibiting a case of anomalous diffusion, while for T above T_c , $\xi \approx 1$.

5. Some remarks on the microscopic approach

In recent years, Mode Coupling Theory (MCT) has been used to describe some dynamic aspects of the glass transition [114]. MCT predicts the following form for the viscosity,

$$\eta = A(T - T_c)^{-\gamma},$$

where A and γ are parameters that characterize the coupling between the molecules of the liquid. The divergence of the viscosity as the temperature approaches T_c indicates the onset of the change from the liquid-like to the solid-like behavior. For temperatures below T_c , MCT describes the scenario for the temperature dependence of the light scattering susceptibility spectrum, the so called boson peak [115]. Furthermore, MCT has been used to describe transitions in hard spheres in colloidal suspensions where the viscosity scales in a similar way replacing the temperature by the concentration. In order to understand the glass transition from a microscopic point of view, some models using hard spheres or hard disks have been proposed. These ideas are based on the fact that a system consisting on identical hard spheres may undergo an order (crystal-like state)-disorder (liquid-like state) transition. When density is increased quickly enough, crystallization may be avoided, thus, a "glass-like" transition is evoked. This transition is characterized in terms of the concentration, instead of the temperature as in a real supercooled

liquid. Computer simulations imply that this kind of transition may be of a kinetic nature such as in a glass transition [116] [117]. By using this analogy, it is possible to generate experiments and computer simulations in colloidal suspensions [118] in order to investigate some critical clues on the glass transition phenomena. Based on the Adam-Gibbs theory, and using the volume fraction as the thermodynamic variable instead of the temperature, experimental and computer simulation evidences of cooperative motion in structural relaxation in colloidal suspensions, both in the α and β relaxation time scales have been reported recently in the literature [119, 120]

6. Concluding remarks

In this work, we have presented some of the most important topics related to the glass transition phenomenon and different kinetic, thermodynamic, and dynamic aspects that have been reported in the literature from different points of view. Two topics were specifically included, namely the description of the relaxation time or the viscosity in different types of glasses and the dynamics of the relaxation processes, specially, the crossover between different regimes. Some theoretical efforts that have been developed to describe the dependence of the relaxation time on temperature, in the particular case of fragile glasses, are discussed. As far as the dynamic aspects are concerned we have shown how the temperature derivative analysis applied to strong and fragile glasses lead us to different kinds of behavior. Using the generalized stochastic matrix method, one may show for strong glasses, particularly B_2O_3 , that a non-Arrhenius behavior is expected for the average relaxation time. In the case of fragile glasses we discuss the evidence of a crossover between different dynamic regimes and its influence in the break down of the Stokes-Einstein relation

The task of explaining to a full extend the phenomenology of the glass transition is so far, as we may conclude after analyzing the facts we have exposed here, clearly unfinished. Some clues might be found in the explanation of the dynamic heterogeneity of the different cooperatively rearranging regions in the supercooled liquid and understanding how the mesoscopic scale controls the rates of the macroscopic scale in glass formers near and above the glass transition temperature.

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Dedication

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