Universal behavior of the viscosity of supercooled fragile and polymeric glassformers in different temperature regions

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H I G H L I G H T S

- Two corresponding states laws for fragile supercooled liquids for all temperatures are presented.
- A new empirical equation for the *LSF* for fragile glassformers below T_c is proposed.
- The universal behavior for the *LSF* for honeys may be extrapolated for all fragile glassformers.

A R T I C L E   I N F O

A B S T R A C T

The behavior of the viscosity of supercooled liquids with temperature has been extensively studied in different regimes. We present a universal behavior for the Logarithmic Shift Factor for fragile and polymeric glassformers in two temperature regions, above and below the crossover temperature T_c, respectively. We find two different equations, one for each region, that may be represented as master plots which show universal behaviors for both cases.

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1. Introduction

The study of the nature of the phenomenology of the glass transition remains still an open issue. The relaxation processes that take place in fragile supercooled liquids have been extensively presented in the literature for temperatures above the glass transition temperature T_g in the range of temperatures between T_g and the melting temperature T_m. It is exactly in this region that the liquid is considered supercooled. The definition of fragility is in the sense of Angell's classification of fragile and strong glasses [1].

As the supercooled liquid approaches T_g, the value of its viscosity η increases drastically with temperature. Experimentally, there is a clear evidence that the dependence of the viscosity with temperature, for fragile glass formers, presents different behaviors varying the temperature range where it is measured. We may find solid proofs that a crossover of regimes is present, below and above a phenomenological crossover temperature, T_c [2–25], which lies within the values $\left(1.15T_g, 1.28T_g\right)$ [26–30]. It is important to point out that this crossover temperature has not the same meaning as the theoretical critical temperature T_c introduced in the Mode Coupling Theory [31–34].

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The dependence of the Logarithmic Shift Factor (*LSF*),

$$LSF = \log a_T = \log \frac{\eta(T)}{\eta(T_s)} \tag{1}$$

with temperature has been widely discussed in both the two different temperature regions presented above, where T_s is a reference temperature.

The presence of this crossover of regimes became evident when two of the most widely used empirical equations that expressed perfectly well the behavior of the *LSF* with temperature, above the crossover temperature, namely, the Vogel–Fulcher–Tamann (VFT) [35–37] and the Williams–Landel–Ferry (WLF) [38] for fragile supercooled glassformers, failed to describe relaxation processes below T_c.

The VFT equation is given by.

$$\log a_T = A - \frac{B}{T - T_0}. \tag{2}$$

A and *B* are independent parameters, and T_0 is generally interpreted as the isoentropic temperature, namely, the temperature where the configurational entropy vanishes [39,40].

The WLF equation is written in the form,

$$\log a_T = \frac{C_1(T - T_s)}{C_2 + T - T_s} \tag{3}$$

where, according to first proposal of Williams, Landel and Ferry [38], C_1 and C_2 were universal constants for all glass forming liquids, given an appropriate reference temperature T_s. The WLF has been applied successfully up to the present in the study of the viscosity of glassforming liquids above T_c, although C_1 and C_2 may not have the same values for different systems [41–52].

Since neither the VFT nor the WLF equations describe the viscosity dependence on temperature below T_c, several equations have been proposed to describe the behavior of the *LSF* with temperature within the range (T_g, T_c) [5,7,8,25–28].

In previous works [53,54] we have proposed an equation for this region that satisfies Stickel's derivative analysis [7,8], that may be written as,

$$-LSF = \log \frac{\eta(T_s)}{\eta(T)} = C(T_A - T)^2 - E \tag{4}$$

where *C* and *E* are fitting parameters, and the temperature T_A is related to T_0 and T_c. This equation has proved to fit experimental values for the *LSF* for several glassformers [53,54].

Finding corresponding states laws for physical quantities has always represented a desired result in many fields of knowledge. It seems that the first efforts to find a corresponding states law for glass forming liquids began in 1972 [55]. One of the first analysis in order to obtain a corresponding states law for several supercooled fragile glassformers is presented by Rössler [27]. He works with eleven organic supercooled liquids and recognizes two kinds of behaviors between the glass transition and the melting point temperatures.

In another work [28], Rössler and co-workers on one side, and Leon and Ngai [5] on the other, study corresponding states laws using ten glassformers, including strong, polymeric and organic substances, obtaining the possibility of fitting the data in both intervals using different VFT equations, an idea that Stickel et al. [8] had already proposed. A different approach is proposed by Elmatad and coworkers [56,57] and Kivelson and coworkers [26] present corresponding states laws in different intervals of temperature from the ones we are presenting in this work.

In this work, we shall show that, through master plots for each region, corresponding states laws for several fragile glassformers, organic and polymers, may be obtained, using in each case different empirical equations. In Section 2, we present the empirical equations that have successfully been used to make the description for each case. In Section 3, we study the behavior for temperatures above T_c, and we find a corresponding states law with a proposed empirical equation for all the supercooled liquids we have considered. We find that using the same equation for temperatures below T_c, this pattern breaks down. In order to find a corresponding states behavior for this last temperature region, in Section 4, we propose a different empirical equation that results in a master plot for the same liquids, now below T_c. Finally, in Section 5 we present a complete discussion on our results.

2. The empirical equations

2.1. The LSF for temperatures above T_c

As we have already pointed out, one of the most used empirical equation to describe the relaxation processes for temperatures above T_c is the WLF equation (3). In previous works [48,58], we have shown that the WLF equation may

Table 1
Values of m and b given in Eq. (6), for the studied liquids for
temperatures above T_c.

LIQUID	m	b
PDE	14.626	−20.13
TNB	14.35	−19.379
OTP	15.93	−21.04
PIB	14.374	−8.398
PVAc	14.49	−11.129
PS	15.116	−12.426

be written in the linear form in terms of $\left(\frac{T_s}{T}\right)^2$,

$$LSF = \log \frac{\eta(T)}{\eta(T_s)} = m\left(\frac{T_s}{T}\right)^2 + b(T_s) \tag{5}$$

where T_s is a reference temperature, m is the slope of the linear form which turns out to be independent of the reference temperature, and $b(T_s)$ is the only parameter that depends on it. This kind of equation has already been considered in the literature by authors such as Bässler [59].

The expression for the logarithmic shift factor for temperatures above T_c will allow us to find a master equation for the viscosity.

2.2. The LSF for temperatures below T_c

In the last three decades, important experimental evidence shows that neither a VFT nor a WLF equation, are able to fit data for the viscosities in the complete range of temperatures between T_g and T_m, the fusion temperature [2–11,30]. We have already presented that, for temperatures above T_c, equations such as VFT and WLF are still valid nowadays to fit experimental values for the viscosity, in particular, Eq. (5).

For the case of temperatures below T_c, we have already proposed in previous works [53,54], an empirical form for the *LSF* for fragile glassformers, namely, Eq. (4),

$$-LSF = \log \frac{\eta(T_s)}{\eta(T)} = C(T_A - T)^2 - E$$

where T_s is a reference temperature, C and E are fitting parameters, and the temperature T_A is connected with T_c and T_0 through the use of Stickel's derivative analysis [7,8].

3. Corresponding states laws for the supercooled liquids above T_c

As we have exposed earlier, corresponding states behavior for glass formers has been presented in different temperature regions using always a reference temperature [27,28,60,56,57].

In this section, we shall present a corresponding states law for temperatures above T_c, through a master plot for the *LSF* for fragile glassforming organic liquids, honeys and polymers. We have already presented this result for several honeys of different countries, under the same conditions [58]. Honey is essentially an aqueous solution of different kinds of sugars such as fructose, galactose, glucose and sucrose depending on the kind of this substance we are studying. The total percentage of sugars in honey is about the 83%, and the rest of its components are different kinds of lipids and amino acids [61]. In this work, we study the *LSF* for temperature above for organic liquids, salol (phenyl salicylate), PDE (phenolphthalane-dimethyl-ether), OTP (orthoterphenil), TNB (tri-naphthylbenzene) [7,8,62,63], polymers PIB (polyisobutilene), PVAc (polyvinyl acetate) and PS (polysterene) [64,65], and honeys [58].

We may fit the experimental data of these glassformers using the linear form we have already presented in Section 2.1, Eq. (5), taking for each case the corresponding value for T_g as the reference temperature T_s. We may express the *LSF* for each one of our systems as,

$$LSF = \log \frac{\eta(T)}{\eta(T_g)} = m\left(\frac{T_g}{T}\right)^2 + b(T_g). \tag{6}$$

As we have already discussed, the slope m is independent of the reference temperature, whereas, the intercept $b(T_g)$ does depend on it. Eq. (6) includes the reference temperature T_g within.

The value of both parameters of Eq. (7) are given in Table 1. As it is easily seen, the values of the slopes are very similar, while the intercepts are different.

In Fig. 1 we plot the experimental values for the *LSF* of PDE, OTP and TNB vs. $\left(\frac{T_g}{T}\right)^2$ and, we clearly see straight lines with almost the same slopes. In Fig. 2, we present the same plot in the case of the polymeric glass formers PIB, PVAc, and PS and obtain similar results.
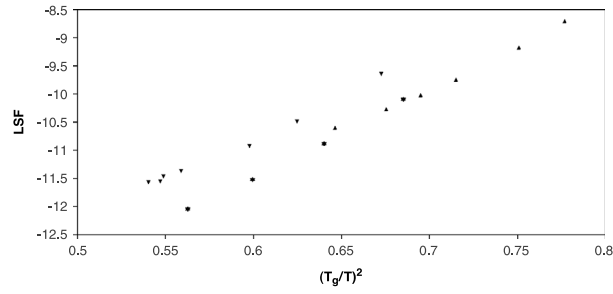
Fig. 1. Plot of the *LSF* vs. $\left(\frac{T_g}{T}\right)^2$ for PDE (▲), TNB (▲) and OTP (★) for temperatures above T_c.
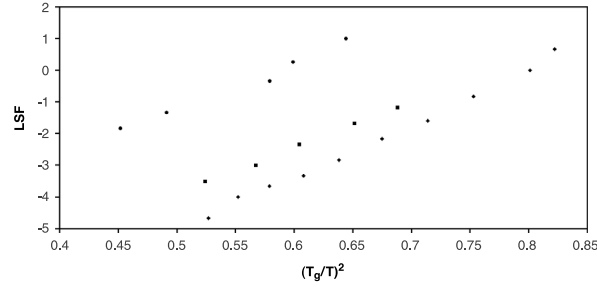

Fig. 2. Plot of the *LSF* vs. $\left(\frac{T_g}{T}\right)^2$ for PIB (●), PVAc (■) and PS (♦) for temperatures above T_c.

In order to obtain a master plot for these six glassforming liquids, we take as a reference the straight line that represents the *LSF*, Eq. (5), for PIB. We introduce a new reference temperature, T_b and evaluate the value of the *LSF* with respect to this new temperature, adding a number c to Eq. (6), namely,

$$\log \frac{\eta(T)}{\eta(T_b)} = LSF + c = \log \frac{\eta(T)}{\eta(T_g)} + c = m \left(\frac{T_g}{T}\right)^2 + b(T_g) + c \tag{7}$$

where,

$$c = \log \frac{\eta(T_g)}{\eta(T_b)}$$

thus,

$$\log \frac{\eta(T_b)}{\eta(T_g)} = m \left(\frac{T_g}{T_b}\right)^2 + b(T_g) = -c. \tag{8}$$

Using Eqs. (6) and (8), we may find the reference temperature T_b in terms of the glass transition temperature T_g,

$$T_b = fT_g \approx T_c \tag{9}$$

where,

$$f = \left(\sqrt{\frac{-c - b(T_g)}{m}}\right)^{-1}.$$

An amazing result is that the new reference temperature of Eq. (8) is related to the glass transition temperature and has a very similar value to the corresponding crossover temperature of each system.

We may finally obtain a master plot for the *LSF* for the six systems in terms of $\left(\frac{T_b}{T}\right)^2$, given by Fig. 3, from which we may conclude a corresponding states law for the viscosities above T_c.

In Table 2, we present the values of c, T_b and f for each system. From this table we may notice that T_b, in all cases, has a value very close to T_c [26–30]. Thus, we obtain a law of corresponding states for the viscosity for different kinds of glassformers above the crossover temperature T_c.

In Fig. 4, we include the values of the *LSF* for several honeys, thus finding a corresponding law for three different kinds of fragile glassformers.

We may now raise an interesting challenge: Is this corresponding states law valid for temperatures below T_c?

Table 2
Corresponding values of c, T_b and f for the master plot, above T_c, given in Fig. 3.

LIQUID	c	T_g	T_b	f
PIB	0	197	257.73	1.31
PVAc	2.73	304	399.31	1.31
PS	4.02	362	485.66	1.34
PDE	11.32	294	387.99	1.32
TNB	10.98	342	447.06	1.31
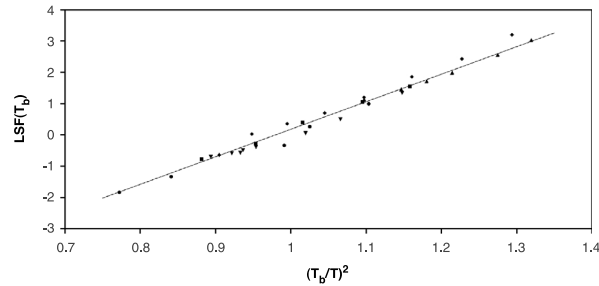
Fig. 3. Master plot of the *LSF* vs. $\left(\frac{T_b}{T}\right)^2$ for fragile glassformers and polymers for temperatures above T_c. The six glassformers PDE (▲), TNB (▲) OTP (★) PIB (●), PVAc (■) and PS (♦).
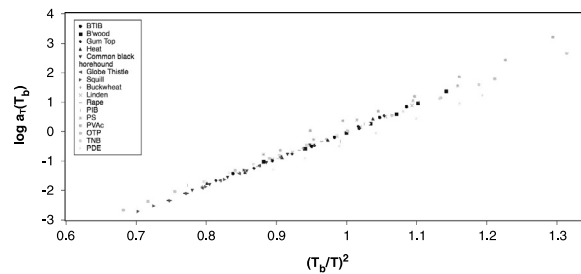

Fig. 4. Master plot of the *LSF* vs. $\left(\frac{T_b}{T}\right)^2$ for the three different kinds of fragile glassformers: organic, polymers and honeys for temperatures above T_c.
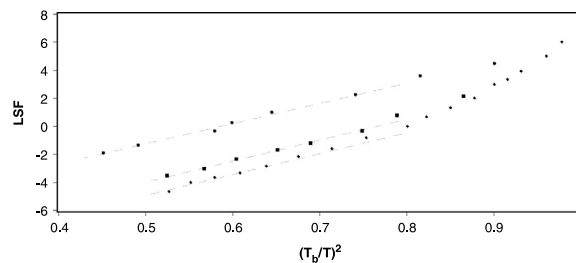

Fig. 5. Plot of the *LSF* vs. $\left(\frac{T_g}{T}\right)^2$ for PIB (●), PS (■) and PVC (♦) in the whole range of temperatures. As it is seen, for temperatures above T_c, we recover the linear form, while below T_c this behavior breaks down.

In order to answer this question, we plot the experimental data for *LSF* for our glassformers in terms of $\left(\frac{T_g}{T}\right)^2$, below T_c, Fig. 5. As it may be seen, the behavior of the viscosity for each system breaks down and thinking of a master plot in this case is really unattainable.

So, the next issue is: May we find a corresponding states law for the viscosities below T_c? The answer will be affirmative, and we shall present it in the next section.

4. Corresponding states below T_c

We shall present the following procedure to obtain a master plot for the Logarithmic Shift Factor below T_c.

Table 3
Values of C_0, T'_A, and E' given in Eq. (14), for the glass formers below T_c.

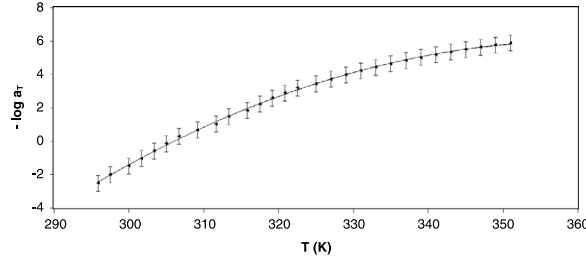| LIQUID	C_0 (K^{-2})	T_c (K)	E'	T'_A (K)
PIB	−0.00203	240	6.372	255.17
PS	−0.00203	447	10.029	444.34
PVC	−0.00203	416	9.929	419.04
PVAc	−0.00203	366	10.540	376.18
Salol	−0.00203	264	8.370	290.97
PDE	−0.00203	352	5.9985	360.36

Fig. 6. Plot of the experimental data for the *LSF* vs. T for PDE below T_c. The curve corresponds to the fit given by Eq. (10). Error bars of 5%.
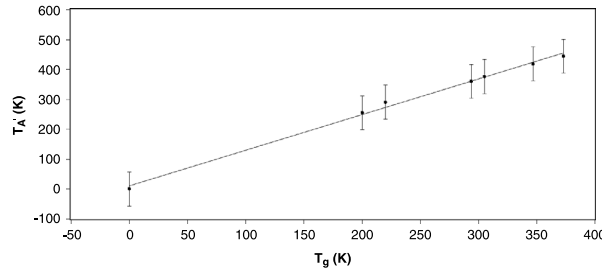

Fig. 7. Plot of T'_A vs. T_g. The slope of the straight line that fits the data is given by Eq. (11). Error bars of 5%.

The first step of our proposal consists in fitting the experimental data as a function of temperature by means of the equation

$$\log \frac{\eta(T_g)}{\eta(T)} = C_0(T'_A - T)^2 + E' \tag{10}$$

fixing the value of C_0, taking into consideration the values we obtained in a previous work [54]. We find for each glassformer the values of T'_A and E', Table 3. As an example we present the plot for PDE, namely, Eq. (10), given by Fig. 6.

Afterwards, we may consider Eq. (10) as a linear function of $(T'_A - T)^2$. We plot this linear equation for six of our glass formers, and as expected, the six straight lines differ from each other only in the intercept. We have not included data for honeys in this temperature region because they are not available. If we analyze the value of T'_A, surprisingly its value is very close to the corresponding T_c of each system. Plotting T'_A vs. T_g and find the linear relation, Fig. 7, given by

$$T'_A = f'T_g \approx T_c \tag{11}$$

where,

$$f' = (1.192 \pm 0.063).$$

Thus, within Eq. (10) the temperature T'_A is related linearly with the reference temperature T_g, thus, the reference temperature of the *LSF* appears within the proposed form given by Eq. (10).

Finally, in order to obtain a master plot we have to find a new reference temperature T^*. We add a number k to Eq. (10)

$$\log \eta(T_g) - \log \eta(T) + k = C_0(T'_A - T)^2 + E' + k. \tag{12}$$

We define

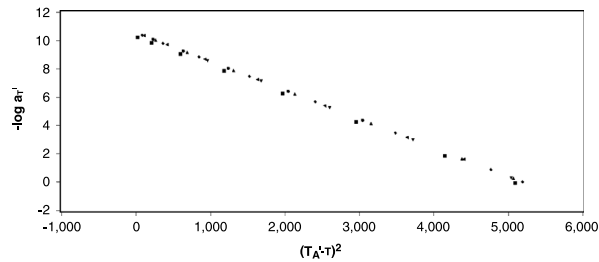$$\log \eta(T^*) = \log \eta(T_g) + k$$
$$E' + k = E''.$$

Fig. 8. Master plot for the *LSF* given by Eq. (14) below T_c. The six glassformers that are presented are PIB (●), PS (■), PVC (♦), PVAc (▲), salol (▼), PDE (◄).

We may rewrite Eq. (12) as,

$$\log \eta(T^*) - \log \eta(T) = C_0(T'_A - T)^2 + E''. \tag{13}$$

If $T = T^*$, then

$$C_0(T'_A - T^*)^2 + E'' = 0.$$

Finding that

$$T^* = T'_A - \sqrt{-\frac{E''}{C_0}}.$$

Plotting Eq. (12) we get the master plot for the *LSF* evaluated with respect to the new reference temperature T^*, Fig. 8,

$$\log \frac{\eta(T^*)}{\eta(T)} = C'(T'_A - T)^2 + E' + k = C'(T'_A - T) + E''. \tag{14}$$

5. Discussion

Achieving corresponding states laws in physics has always represented a expected result in many fields of knowledge. As we have already mentioned in the introduction, many authors have discussed this kind of behavior in different temperatures intervals for several glassformers, using different empirical equations [27,55,60,56,57].

In this work, taking into consideration that we have enough proofs to propose different behaviors in different temperature intervals between the glass transition and the melting point temperatures, we postulate two different empirical equations in the two temperature regions, basing our hypothesis in a good number of studies under these assumptions [2–30]. Our proposals are valid, respectively, for temperatures above and below the crossover temperature T_c.

In Section 3, for temperatures above T_c, we have discussed an empirical equation for the logarithmic shift factor. This new expression, Eq. (7), has allowed us to find a master plot for the values of the viscosities of fragile and polymeric glassformers.

This study has already been presented in the case of different honeys that show the same pattern [58]. Honeys, essentially, are mixtures of different kind of sugars and water, obviously completely different from polymers and other fragile glassformers. Up to our knowledge this work is one of the most complete studies of the corresponding states law for totally different kinds of glassformers in the temperature range considered. It is important to point out that for the three types of glassformers, the reference temperature for the master plot has the same value, the crossover temperature of each supercooled fluid.

As for the case of the description of the *LSF* for temperatures below T_c very few attempts may be found in the literature. In an earlier work [54] we propose the equation given in Eq. (4) in the case of fragile glassformers. Here we have rewritten this equation as a linear form Eq. (10) with a fixed slope, valid also for the polymeric systems, where the meaning of the temperature T'_A, surprisingly is the same of T_b in the former case. We have also obtain through a master plot a corresponding states law. In this temperature region we have not been able to include honeys because the experimental values of the viscosities are unavailable.

Straightforward from Fig. 5 it is evident that one phenomenological expression for the *LSF* may not fit the complete range of temperatures, thus a master plot for the viscosities that include the whole region is out of question.

6. Conclusions

A single empirical equation to describe the *LSF* for fragile and polymeric supercooled liquids for all temperatures between T_g and the melting point temperature T_m, is unattainable, as it may be seen in this work and in most of the literature on this subject. We have proposed in each temperature region, above and below the crossover temperature T_c, empirical equations, that, in terms of adequate reference temperatures, lead to corresponding states laws in terms of two master plots.

Surprisingly, in both cases, the reference temperature is very close to T_c. Consequently, we may attain the same universal behaviors for the two kinds of systems in the same temperature intervals. In the case of temperatures above T_c, it is important to point out that remarkably, we may obtain the same master plot for honeys, fragile glassformers and polymeric liquids, for they have the same slope and reference temperature as it may be seen in this work and in Ref. [58].

Acknowledgments

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